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Title: Discovery and Investigation of Heavy Neutron-Rich Isotopes with Time-Resolved Schottky Spectrometry in the Element Range from Thallium to Actinium

Article Type: Letter

Section/Category: Experiments

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Order of Authors: Lixin Chen; Hans Geissel

To:

Prof. Dr. Volker Metag
Editor
Physics Letters B

Dear Volker,

Please find below the comments, answers and indicated changes in the text we have made in the manuscript:

Ms. Ref. No.: PLB-D-10-00376
Title: Discovery and Investigation of Heavy Neutron-Rich Isotopes with Time-Resolved Schottky Spectrometry in the Element Range from Thallium to Actinium
Physics Letters B

We would like to thank very much the referees for valuable comments which have improved the present paper.

Reviewers' comments:

Reviewer #1: The submitted letter reports new mass values south and southeast from 238-U. The method is explained in sufficient details and references nicely to other publications for additional details. I do recommend publication of this letter when these (minor) questions are addressed:

- In introduction, 16th line; please briefly explain "conventional detector systems"

Conventional detectors have been explicitly explained.

- In introduction where B\rho-\Delta E -B\rho method is mentioned; should add reference to [6]

The reference has been included.

- Figure 2. Square label: isotopes with well-known masses; please define what is well-known. dM < 10 keV?

The well-known masses have a mean experimental uncertainty of about 8 keV, it has been included in the legend of figure 2. Of course there are uncertainties of single values up to 50 keV.

- page 4, right column, 1st line; please also clarify "long living isomers", how long?

The lifetime of the discovered and investigated isomers are in the minute range, which is now stated in the text.

Data handling. I understand that obtaining mass values from data needs an extensive network of reference masses. Are these masses only from AME2003? Since the publication of atomic mass evaluation 2003, several new, especially Penning trap measurements, have been performed. Is the analysis using solely data from AME2003 and if other data is used, this should be clarified in the

text. Also, perhaps a little bit of explanation of the data treatment could be incorporated to text.

Yes, we have used only reference masses from AME2003. We have observed a nice agreement between the trap and the ESR measurements for a few Rn isotopes.

Masses were determined using the correlation matrix method described in details in the paper [19].

How about the knowledge of atomic binding energies? Are these known sufficiently precisely and does atomic electron binding energies contribute to the error budget of the measured atomic mass excesses? Perhaps also provide source reference of the atomic electron binding energies.

The total binding energy has never been experimentally determined in heavy atoms (e.g. in the lead region).

The electron binding energy in a neutral heavy atom (568 keV for Pb) is known with an accuracy better than one per mille (less than 1 keV).
This contribution is much less than our present experimental error. But note only in the comparison with tabulated or other published experimental data from other methods the full electronic binding energy is relevant, because in general we have the advantage of measuring bare and few-electron ions.

The binding energies calculated from Li-like ions up to a neutral atom were taken from the paper:

G. C. Rodrigues et al., At. Data Nucl. Data Tables 86 (2004) 117.

Binding energies for H- and He-like ions were calculated in the paper:

D.R. Plante, W.R. Johnson, J. Sapirstein, Phys. Rev. A 49 (1994) 3519.
K. Huang et al., At. Data Nucl. Data Tables 18 (1976) 243.
W.R. Johnson et al. At. Data Nucl. Data Tables 33 (1985) 405.

Representative references are now included in the text.

- page 6-7. Figures 5 and 6 are not referenced in the text.

The links are now made.

- Figure 6. Filled triangle symbol legend is missing. Also, please harmonise the figure with others so please make full box around the plot.

The figure, the symbols, and its motivation is better explained.

- Also for general interest, it would be good to have somewhere information about when were these measurements carried out and how much beam time was used.

The measurements were performed in 2004 (now inserted in the text), but still the new isotopes in the text have not been published in the literature. However, recent FRS experiments (without the storage ring) may cover and extend the limits.

All in all, this is a very nice letter. And as stated in text, a more comprehensive paper will be published later.

Reviewer #2: The paper describes the results of an excellent experiment with very impressive new results. The use of storage rings to study the properties of exotic nuclei is a powerful new tool for high precision measurements. The article certainly deserves to be published in Phys. Lett. B.

The authors should consider the following comments: It seems to me that the article is a little bit long for a letter. Although overall well written, it is repetitive in some instances and could be more concise. For example, the figure captions are lengthy and repeat the content of the main text. Another example is the discussion of the m/q identification. A whole paragraph stresses the point that "it was checked" without really containing any new information other than simulations were done.

The figure captions and parts of the text have been shortened where the information is anyway in the text.

It would be nice to include all known isotopes in Figure 2. This could be done by open squares.

Figure 2 shows the nuclides which have been observed in the present experiment. The limit of the presently known most-neutron-rich nuclides are indicated in the figure for orientation. The figure has been improved.

It is surprising to me that 213Tl was observed, without observing any of the neighboring nuclei (212Tl and 215Pb for example). Shouldn't 215Pb82+ show up in figure 3?

It is true that at the first glance one would expect to see nuclides with higher production cross sections as well. However, the stored new nuclides are mainly observed once and for such rare events the complex experimental setting (the complex and tiny ESR acceptance) and the stochastic atomic processes in the matter can cause the present observation. We have observed such irregularities also with stored single ions in different atomic charge-states.

I don't think that the present paper adds anything new to the discussion of dVpn. The observation of the shell gap has been published before and the new points are not even discussed. The corresponding paragraph and figure 6 could be eliminated without weakening the paper.

The figure and its motivation have been better explained now in the text. Besides this nuclear structural information the double difference of binding energies is a clear signature of the accuracy in the mass measurements. In our pioneering mass measurements such information was not possible to extract.

Discovery and Investigation of Heavy Neutron-Rich Isotopes with Time-Resolved Schottky Spectrometry in the Element Range from Thallium to Actinium

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Abstract

^{238}U projectile fragments have been created at the entrance of the fragment separator FRS, spatially separated in flight within 0.45 μs and injected into the storage-cooler ring ESR at 7.9 Tm corresponding to about 70% light velocity. Accurate new mass values and lifetime information of the stored exotic nuclei in the element range from platinum to uranium have been obtained with single-particle Schottky spectrometry. In this experiment the new isotopes of ^{236}Ac, ^{224}At, ^{221}Po, ^{222}Po, and ^{213}Tl were discovered. The isotopes were unambiguously identified and their mass measured. In addition, the time-correlated data have provided information on the lifetime of the new nuclides. The discovery of isotopes along with accurate mass measurement has been achieved for the first time at the FRS-ESR facility. The results will contribute to the knowledge of the decay products from the r-process nuclei and enable a crucial test of the predictive power of modern nuclear mass and half-life models.

Keywords: 21.10.Dr, 21.60.-n, 200<=A<=240, 32.10.Bi

1. Introduction

A basic goal of nuclear structure physics is to extend the knowledge on nuclear properties beyond the present limit of existence towards the driplines. New physical phenomena, like unexpected shell evolution at extreme neutron-proton asymmetry [1] and new decay modes [2, 3] have been discovered for the most exotic species. Presently, our knowledge is still rather scarce for neutron-rich nuclides above lead because their production cross sections are very small and the separation and particle-identification techniques for these heavy ions are very difficult and complex due to the atomic interaction, energy straggling and charge-changing collisions in conventional detector systems (e.g. tracking, time-of-flight, energy-deposition, and kinetic energy detectors). A breakthrough has been recently achieved in fragmentation studies with ^{208}Pb and ^{238}U projectiles at relativistic energies [4, 5]. In the latter experiment the fragments have been produced at 1000 MeV/u at the entrance of the in-flight separator FRS [6]. The magnetic system of the FRS can spatially separate the fragments of interest applying the Bρ-ΔE-Bρ method [6]. Redundance and verification are conventionally achieved in combination with particle identification detectors measuring the time-of-flight and the energy deposition to determine the mass number (A) and the proton number (Z). Here, we report on studies of stored highly-charged exotic nuclei applying a new combination of experimental separation tools for the discovery of isotopes with the ultimate sensitivity down to single ions.

2. Production and In-Flight Separation of Stored Uranium Fragments

A 670 MeV/u ^{238}U projectile beam extracted from the heavy-ion synchrotron SIS [7] with an intensity of 1×10^9/spill was focused on a 4 g/cm^2 beryllium production target placed at the entrance of the fragment separator FRS in an experiment performed in 2004. Fast extraction was used with a spill length of 300 ns and a typical repetition rate of 0.2 per minute determined by the chosen spectrometry cycle. The fragments of interest were separated in flight with the FRS applying the Bρ-ΔE-Bρ method [6]. The separation performance and the selected element range was checked with particle detectors and slow extraction at the direct achromatic focal plane $F4$ of the FRS, see figure 1, before we applied fast extraction and injected the fragments into the storage-cooler ring ESR [8].

In principle, we could inject monoisotopic fragment beams [9, 10] but since one of our main goals in this experiment was to perform mass measurements in a new territory, the separation conditions at the FRS were selected such that in addition to the nuclei of interest sufficient reference masses were recorded in the same spectrum. The degrader thickness of 50 mg/cm^2 (plastic) and thus

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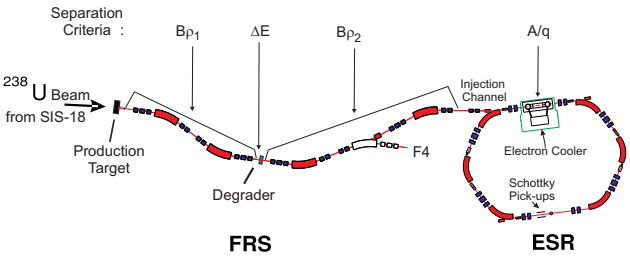

Figure 1: Experimental method and setup for the discovery of new isotopes and simultaneous measurement of their masses and lifetimes.

(ΔE) were selected to restrict the transmitted Z-range of the nuclides between gold and uranium. Electron cooling was applied to the stored ion beam which forces the circulating ions to an identical mean velocity determined by the chosen terminal voltage of the electron cooler. The different separation criteria are indicated in Fig.1. Their efficiency and performance have been reproduced by realistic Monte-Carlo simulations using the code MOCADI [11].

The revolution frequency of the cooled fragments was measured by time-resolved Schottky Mass Spectrometry (SMS) [12] using metallic pick-up plates to record the current signals induced by the stored circulating highly-charged ions at each revolution. The signals from the 30^{th} harmonics were transferred to the frequency range of 320 kHz by an image-reject mixer and then digitized and stored by the data acquisition system for off-line analysis. The frequency spectrum of the stored ions was generated by Fast Fourier Transformation. Electron cooling reduced the velocity spread of the stored fragments to approximately $3{\times}10^{-7}$. Thus the revolution frequency f is independent of the velocity spread and only proportional to the mass-to-charge ratio m/q of the orbiting ions. The revolution frequencies of two different cooled ions i and j are related by:

$$\frac{f_i - f_j}{f_i} = -\alpha_p \frac{(m/q)_i - (m/q)_j}{(m/q)_i}, \qquad (1)$$

where α_p is the momentum compaction factor which was determined over the full m/q range of the storage acceptance of the ESR. In this way each peak in the frequency spectrum corresponds to a specific mass-to-charge value. The measured m/q resolution was about $6{\times}10^{-7}$. In combination with the separation criteria of the in-flight separation described above this enables us to perform an unambiguous assignment of A and Z of the recorded ions. Although the injection channel of the ESR has a strong limitation in the Bρ and angular acceptance, nevertheless we could access previously unknown isotopes because of the high efficiency and selectivity of the in-flight separation and the performance of the time-resolved SMS. Note that in the Z-range of the present experiment SMS has the ultimate sensitivity down to single ions [12]. Single-particle spectroscopy is the key to determine very close lying ground and isomeric states because a particle can only occupy one or the other state at a fixed point in time.

Rich new information has been obtained from the present experiment: i) mapping of a new territory of the heavy neutron-rich mass surface. For 33 isotopes the masses have been measured for the first time. ii) new long-lived isomeric states in the minute range have been observed and their excitation energy has been

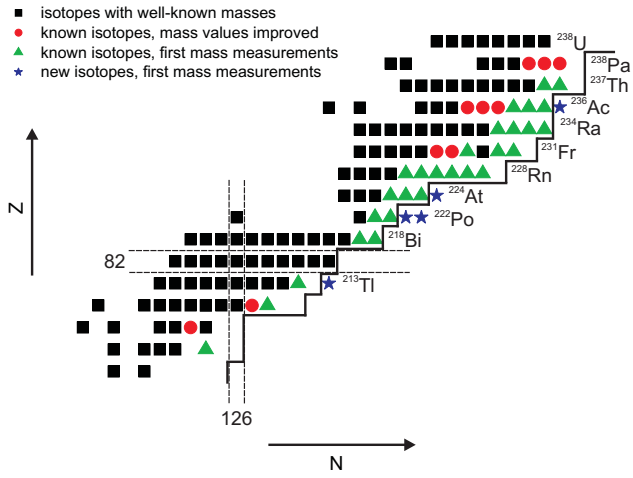

Figure 2: Illustration of new results from the present experiment. 5 new isotopes have been discovered in the element range from thallium to actinium. Their masses have been accurately determined and information on their lifetime has been obtained as well. The limit of previously known neutron-rich isotopes is indicated by the solid line. The well-known masses indicated in the figure (filled squares) have an average experimental error of about 8 keV [13].

determined. iii) 5 new nuclides have been observed in the element range from Thallium to Actinium. The latter results are the main topic of this publication.

An overview of the harvest of this experiment is illustrated in Fig.2. The access to nuclides with the same neutron number as the primary beam can be attributed to the contribution from cold fragmentation reaction [14], e.g., ^{234}Ra resulted from a removal of 4 protons while the excitation energy is still low enough that no neutrons were evaporated. Fig.2 includes also the observed isotopes formed in nuclear charge-exchange reactions. Three very neutron-rich nuclei – ^{238}Pa, ^{237}Th, and ^{236}Ac are formed via the (n,p) reaction channel. This reaction type has also been observed with ^{208}Pb projectiles at 1000 MeV/u in other FRS experiments [15].

3. Identification and Investigation of New Isotopes with Time-resolved SMS

The standard criteria for the scientific approval of the discovery of a new isotope are that A and Z are unambiguously identified and that a physical property of the new nuclide has been measured in the same experiment. In such experiments dedicated to explore the limits of presently known nuclei the full particle identification and often the production cross section have been published [16, 17]. More information is gained, if first decay properties are deduced as well. With the described experimental setup and methods we have identified new nuclides and measured their masses with an accuracy of $\Delta m/m \approx 3 \times 10^{-7}$. The information on their lifetime can be deduced by the same experimental method if their decay time matches the access window of SMS [18, 19].

The m/q identification in the frequency spectrum was based on a pattern recognition algorithm including the well-known measured masses [13] as calibration grids and for the unknown isotopes the microscopic theoretical prediction [20]. An unambiguous assignment was achieved together with the combination of the applied different separation criteria described above. In addition, we had to exclude the possibility that an ion of a well-known nuclide in a rare atomic charge-state could be

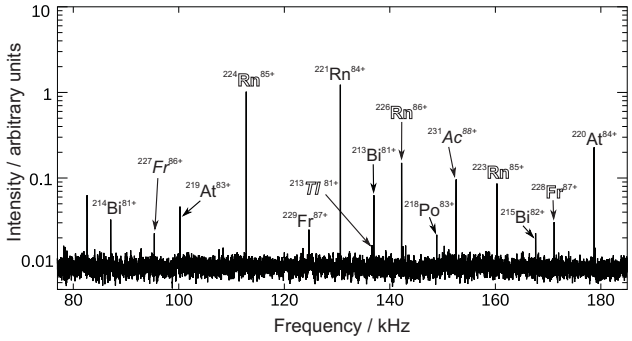

Figure 3: Characteristic Schottky frequency spectrum with isotopes of well-known masses and those with unknown masses (outlined element name). The new isotope ^{213}Tl is included in the spectrum. Note, the frequency range in the plot results from the subtraction of a fixed oscillator frequency.

misidentified as a new isotope. At the FRS-ESR this probability is rather small due to the high velocity of the fragments (v/c = 0.7) and the resulting narrow charge-state distribution. Nevertheless, in the interpretation of the frequency spectra, ions of possible charge states corresponding to up to five bound electrons were considered. In this way we checked that a very rare charge state could cause a possible wrong identification. In conclusion, we can state that our assigned particle identification down to the appearance of single ions is clear without ambiguity.

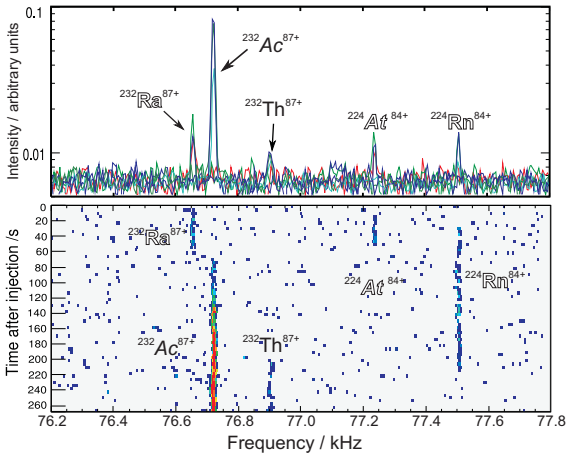

Figure 4: The time evolution of the Schottky revolution frequency spectrum and the corresponding projection. Here, the frequency information of the discovered ^{224}At isotope is illustrated.

In Fig.3 a typical frequency spectrum is shown including the new ^{213}Tl isotope, known isotopes for which the mass values have been measured for the first time in this experiment and those suitable for providing reference masses. In Fig.4 time traces and their projection into a frequency spectrum are shown for the new isotopes ^{224}At and close-lying ions. The single stored ions are easily recognized in the two-dimensional presentation.

Using known masses as reference the masses of the new isotopes have been determined. The results are listed in table 1. The mass determination has been mainly performed as described in reference [21]. However, the subdivision of the full frequency range has led in general to higher accuracies, see also ref. [19]. The atomic electron binding energies are taken into account from references [22, 23]. Their uncertainties are a negligible contribution to the errors of our mass values. More details will be given in a forthcoming paper.

Time-resolved SMS measurement can provide also half-life information along with the accurate mass measurements. Following the trace of single or few ions the decay can be observed by the change of the intensity in the trace. Especially interesting is the ob-

Isotope	ME (keV)	$T_{1/2}$ (s)
^{236}Ac	51267 ± 68	72^{+345}_{-33}
^{224}At	27706 ± 59	76^{+138}_{-23}
^{222}Po	22476 ± 67	145^{+694}_{-66}
^{221}Po	19783 ± 58	112^{+58}_{-28}
^{213}Tl	1763 ± 61	101^{+484}_{-46}

Table 1: The mass and half-life results of new identified isotopes in this work.

servation of the correlated decay events, i.e., when the mother trace disappears and the corresponding daughter trace appears. This so called single-particle spectroscopy is independent on the assumption that the area of a frequency peak is strictly proportional to the number of stored ions. The different methods of half-life measurements of stored ions are discussed in reference [19]. The errors for the half-lives were determined by using the approach pioneered in connection with the evaluation of superheavy element analysis [24].

The complete physical impact from the results of this mass measurement will be given in more detail in a forthcoming publication. Here, we illustrate the gained information with a representative comparison of our new experimental mass values with the widely-used and most accurate macroscopic-microscopic model $FRDM$ [25], the semi-empirical complex mass formula of Duflo and Zuker [26], and a microscopic Hartree-Fock-Bogoliubov $HFB-14$ theory [27]. The comparison in Fig.5 clearly shows the deficits of the models. The deviations from the experimental data systematically increase with increasing neutron number, i.e., with the distance from the previously known masses and valley of stability. Therefore, our new experimental information on masses will contribute to improvements of the mass models and thus to more reliable theoretical predictions for the r-process nucleosynthesis.

The average interaction of the protons and neutrons in their shell occupancy can be investigated by the δV_{pn} values [28, 29] which represent an averaged double difference of binding energies reflecting nuclear structure information only for accurate mass measurements. For two elements, Po and Rn, this interaction is depicted in Fig.6 for even-even isotopes as a function of the neutron number. The shell closure at N=126 is clearly manifested and possible structural changes as suggested in reference [30] are confirmed and extended by two values.

The comparison of the measured half-lives for the discovered isotopes with the models of references [31] and [32] reveals that in general our experimental values are up to one order of magnitude larger. The observed strong differences to the theory have also been observed for neutron-rich isotopes produced from ^{208}Pb primary beam [33]. Details will be discussed in a separate publication.

4. Summary

The results of this experiment clearly demonstrate the discovery potential of the unique combination of FRS and ESR for the investigation of new rare isotopes. For the elements Ac, At, Po, and Tl the most neutron-rich isotopes have been discovered in the present experiment. The importance of cold

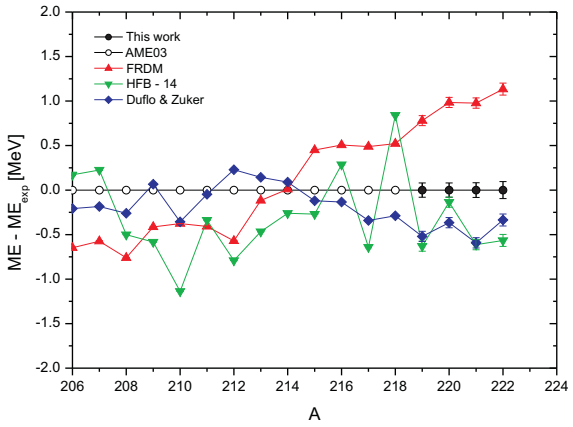

Figure 5: Comparison of experimental masses for Po isotopes with different theoretical models. The experimental data are taken from ref. [13] and the present experiment.

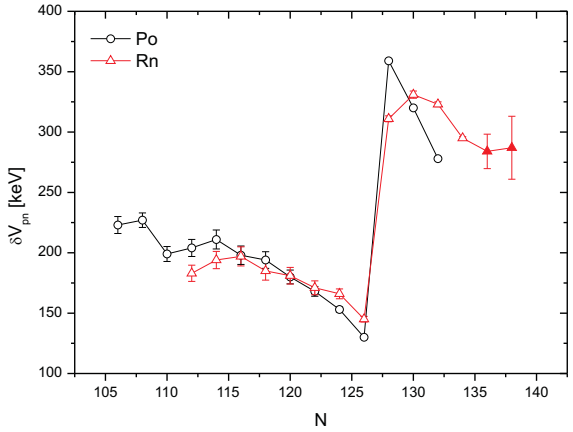

Figure 6: The shell closure at N=126 is clearly demonstrated in the proton-neutron interaction represented by the δV_{pn} values. The filled symbols indicate the new δV_{pn}-values for Rn-isotopes achieved in this experiment.

fragmentation and nuclear charge-changing reactions is manifested by the observation and investigation of the so far most neutron-rich isotopes in the region of lead and uranium. Time resolved Schottky spectrometry was applied to measure the masses of the new isotopes with an accuracy of about 70 keV. Note that the information is based on single particle observations. Representative illustration of the physical impact of these experimental results demonstrates the shortcomings of the present theoretical description especially for the new isotopes discovered. The averaged proton-neutron interactions are reflected in the deduced δV_{pn} values. The shell closure at N=126 and possible shape changes (octupole deformation) are indicated.

In future, the intensity upgrade program for the GSI accelerators will substantially contribute to improve the accuracy of the spectroscopy information for the most exotic isotopes and will also give access to more unknown nuclides.

Acknowledgement

It is a pleasure to thank the technical staffs of the accelerators, the FRS, and the target laboratory for their valuable contribution to the beam quality and experimental setups. The authors gratefully acknowledge fruitful discussions with K. Blaum, I. Borzov, and R.F. Casten. We thank very much the HGF for the support VH-NG-033 which was a basis for the strong collaboration of the university Gießen and the research center GSI.

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