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# Experiments with stored exotic nuclei at relativistic energies

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#### **Abstract**

A review and recent progress are presented from experiments on masses and lifetimes of bare and few-electron exotic nuclei at GSI. Relativistic rare isotopes produced via projectile fragmentation and fission were separated in flight by the fragment separator FRS and injected into the storage ring ESR. This worldwide unique experimental method gives access to all fragments with half-lives down to the microsecond range. The great research potential is demonstrated by the discovery of new isotopes along with simultaneous mass and lifetime measurements. Single particle decay measurements and the continuous recording of both stored mother and daughter nuclei open up a new era for nuclear spectroscopy. The study of bare and few-electron nuclei has also important astrophysical relevance with respect to the hot stellar conditions where reactions and decay are influenced by the degree of atomic ionization. The future international NUSTAR facility at FAIR consisting of a new large-acceptance in-flight separator (Super-FRS) and a dedicated storage ring system (CR–RESR–NESR) will be an ideal tool to study nuclei with new probes and to investigate the majority of relevant r- and rp-process nuclei which are not in reach with the present-day facilities. © 2006 Elsevier B.V. All rights reserved.

*Keywords:* Storage rings; Exotic nuclei; Highly-charged ions; Mass and half-life measurements

### **1. Introduction**

Mass and half-life of a nucleus are fundamental properties which result from the interaction of all nucleons [\[1\]. N](#page-7-0)ew phenomena in nuclear physics like shell structure, pairing correlations, and decay and reaction properties have been discovered via nuclear mass measurements. Separation energies and Q-values for reactions are deduced from masses. The driplines, which determine the borders of nuclear existence, are also obtained from the mass differences of neighboring nuclei. The actual pathways of the nucleosynthesis in stars are governed by the nuclear binding energies and lifetimes.

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A very important motivation for measuring new masses of exotic nuclides is the test and improvement of nuclear theories and models. Although the progress of the theories has been enormous in recent years[\[2,3\], e](#page-7-0)specially concerning the microscopic calculations, their predictive power is still more than one order of magnitude worse compared to our presently achieved experimental accuracy [\[4,5\].](#page-7-0)

Today the challenge is to measure the masses and lifetimes of exotic nuclei close to the borders of their existence. These nuclides reveal new nuclear properties due to the strong asymmetry of their proton-to-neutron ratio. However, they are difficult to investigate due to their small production cross-sections and short lifetimes. Two methods for direct mass and lifetime measurements of stored projectile fragments were pioneered at GSI, namely Schottky mass spectrometry (SMS) [\[6–9\]](#page-7-0) and isochronous mass spectrometry (IMS) [\[10,11\].](#page-7-0) The time-

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<span id="page-1-0"></span>resolved SMS ideally enables simultaneous mass and half-life measurements down to intensities of single particles [\[12\].](#page-7-0)

In this contribution we summarize the main achievements and report on recent experiments performed at the SIS–FRS–ESR facilities. Furthermore, we outline future perspectives offered by the planned NUSTAR facility at FAIR [\[13\].](#page-7-0)

### **2. Experimental**

The combination of the in-flight separator FRS [\[14\]](#page-7-0) and the cooler storage ring ESR [\[15\]](#page-7-0) at GSI provides unique experimental conditions with bare and few-electron ions for all elements up to uranium. Relativistic fragments of several hundreds MeV/u are produced via fragmentation or fission of primary beams, provided by the heavy-ion synchrotron SIS [\[16\],](#page-7-0) and are separated in-flight with the FRS. The separated reaction products are injected into the ESR. A schematic layout of the experimental facility is presented in Fig. 1. The experimental conditions are given for a recent experiment where both stochastic and electron cooling have been applied.

The frequencies *f* of the circulating ions in the storage ring can be related to their mass-to-charge ratios  $(m/q)$  in first-order approximation

$$
\frac{\Delta f}{f} = -\frac{1}{\gamma_t^2} \frac{\Delta(m/q)}{(m/q)} + \left(1 - \frac{\gamma^2}{\gamma_t^2}\right) \frac{\Delta v}{v} \tag{1}
$$

where  $\alpha_p = 1/\gamma_t^2$  is the momentum compaction factor, which is defined as

$$
\alpha_{\rm p} = \frac{\mathrm{d}C/C}{\mathrm{d}(B\rho)/(B\rho)}\tag{2}
$$

and which denotes the relative variation of the orbital length  $(C)$ per relative variation of the magnetic rigidity  $(B\rho)$ .

The fragments have inevitably a velocity spread of the order of 1% due to their stochastic creation process. The corresponding

phase-space enlargement would aggravate precision measurements in flight. However, a storage-cooler ring presents an ideal tool to overcome this disadvantageous property. The large phasespace can be reduced by stochastic [\[18\]](#page-7-0) and electron cooling [\[19\],](#page-7-0) which forces all stored ions toward the same mean velocity and thereby reducing the velocity spread to roughly  $5 \times 10^{-7}$ at low intensities. Thus the second term in Eq. (1) becomes negligible and the measured revolution frequencies reflect directly the mass-to-charge ratios of the stored ions [\[4,5\].](#page-7-0) The velocity dependence is thereby almost completely removed, which is the basis of SMS. The time-resolved SMS is ideally suited to determine the intensity of the circulating ions, thus providing lifetime data simultaneously with the mass measurement [\[20,21\].](#page-7-0) The disadvantage of the method is the relatively long cooling time, which amounts to a few seconds and therefore limits the accessible nuclides. Exotic nuclei with half-lives shorter than the cooling time can be investigated with a time-of-flight technique by operating the ESR in the isochronous mode. For IMS [\[11\]](#page-7-0) a special ion-optical setting makes the revolution frequency of a particular ion species independent on its velocity spread. In this technique, the ions are injected at the velocity corresponding to  $\gamma_t$ , thus eliminating the second term in Eq. (1). Our experiments have demonstrated, however, that the isochronous condition is strictly fulfilled only in a small velocity and m/q-range.

#### **3. Mass measurements**

In the SMS, a frequency bandwidth of 320 kHz is simultaneously measured, which is sufficient to cover the entire frequency acceptance of the ESR. The data acquisition and the data analysis are described in Ref. [\[5\].](#page-7-0) An example of a broad-band Schottky spectrum from [\[5\]](#page-7-0) is shown in [Fig. 2,](#page-2-0) for the case of stored  $209Bi$ -projectile fragments.

Applying time-resolved SMS we can correct for possible drifts. In this way we have obtained higher resolution and have



Fig. 1. Schematic layout of the present secondary nuclear beam facility at GSI. The heavy-ion synchrotron SIS, the fragment separator FRS, and the storage-cooler ring ESR used in the experiments are highlighted. The data given for the primary beam energy, the production target, degrader, and the energy of the fragments injected into the ESR are used in the recent experiment devoted to the lifetime measurements of stored hydrogen-like <sup>140</sup>Pr<sup>58+</sup> ions [\[17\]. B](#page-7-0)oth stochastic and electron cooling have been applied, at an incident energy of 400 MeV/u being required for the stochastic precooling.

<span id="page-2-0"></span>

Fig. 2. Typical Schottky frequency spectrum with 320 kHz bandwidth, 2<sup>16</sup> frequency channels, and 30 s recording time. It is divided in four subsequent parts. The prominent lines are labelled. Up to three different charge states of the same isotope were simultaneously measured, e.g., see the peaks of  $198Bi^{81+}, 82+$ ,83+ ions at about 13, 143, and 270 kHz, respectively. A great advantage is that reference and unknown masses are ther simultaneously recorded in the spectra. The labelling of known and unknown atomic masses is shown according to the AME'95 [\[22\].](#page-7-0)



Fig. 3. Measured mass spectra analyzed from single  $147$  Dy ions in the ground and isomeric states. The ground (g) and isomeric (m) state are separated by 750.5 keV. A resolving power of  $2 \times 10^6$  (FWHM) has been achieved by tracing down to single particles [\[5\].](#page-7-0)

gained the feature to record the dynamics of the circulating ions. Tracing the peaks in time down to a single stored ion, ground or isomeric states can be assigned even for very small excitation energies, which is not possible under the condition when both states are simultaneously populated. With this so-called single-particle method described in detail in Ref. [\[5\],](#page-7-0) a mass resolving power of  $2 \times 10^6$  was achieved. The power of the method is demonstrated in Fig. 3 [\[12,23\]](#page-7-0) where the ground and isomeric states of <sup>147</sup>Dy are clearly resolved. The mass accuracy achieved in SMS is presently 30 keV [\[5,24\],](#page-7-0) which represents an improvement by a factor of about three compared to our first measurements [\[4,25\].](#page-7-0) Two hundred and eighty five new and more than 300 improved mass values of neutron-deficient isotopes in the range  $36 < Z < 92$  have been contributed by SMS (including mass values obtained indirectly via Q-values of  $\alpha$ -,  $\beta$ -, or proton-decays) to the present knowledge of the mass surface [\[26\].](#page-7-0) The research potential of SMS has been extended in our recent experiments with proton-rich 152Sm-projectile fragments [\[17\]](#page-7-0) and neutron-rich  $^{238}$ U-projectile fragments [\[27\]. I](#page-7-0)n the latter case we could measure the mass and the half-life of hitherto unknown isotopes, which is illustrated in Fig. 4.

Our large set of new mass values represents a data base for new nuclear structure studies. Examples are the shell structure of lead isotopes [\[25\], t](#page-7-0)he location of one-proton and two-proton driplines [\[25\], a](#page-7-0)nd the isospin dependence of pairing-gap energies [\[29\].](#page-7-0) In Fig. 5 the experimental pairing-gap energies are compared to the model predictions and it is clearly seen that the observed isospin dependence cannot be reproduced by the



Fig. 4. Discovery of the new isotope 235Ac along with its mass and lifetime measurements applying time-resolved SMS [\[28\]. T](#page-7-0)he mass value has been extracted by calibrating with the known mass for <sup>235</sup>Th (panel a), whereas the half-life has been extracted from the time evolution of the peak-area (panel b).



Fig. 5. Comparison of the proton (left panel) and neutron (right panel) pairing-gap energies for even–even hafnium isotopes derived with the 5-mass formula from experimental mass values and from predictions of mass models [\[30,31\].](#page-7-0)

<span id="page-4-0"></span>

Fig. 6. New mass values as a crucial test for the predictive power of mass models. The deviations ( $\sigma_{\rm rms}$ ) were calculated with 310 masses (represented by *n* in the formula, see insert) which were not experimentally known in the last atomic mass evaluation (AME) [\[26\]. F](#page-7-0)or the description of the models see Refs. [\[2,3\],](#page-7-0) and references therein.

current mass models. This conclusion holds also for the other elements in between the closed shells at  $Z = 50$  and 82.

The predictions of different mass models were tested over a large part of the nuclidic chart consisting of 310 masses that were not included in the parameter adjustments of these models (see Fig. 6) [\[24,28\].](#page-7-0) The comparison reveals that presently the best mass models are characterized by an accuracy that is about one order of magnitude worse than our measurements. For example, recent Hartree–Fock–Bogoliubov models, HFB-1 (BSk1) [\[32\]](#page-7-0) and HFB-2 (BSk2) [\[30\], d](#page-7-0)emonstrate deviations of  $\sigma_{\rm rms} = 1062$ and 650 keV, respectively.

The IMS does not require electron-cooling and is applicable to very short-lived nuclides with half-lives down to a few 10  $\mu$ s. Previously, IMS was successfully applied in small isotope regions with many reference masses in the corresponding spectra [\[33,34\]. I](#page-7-0)n these pilot experiments the mass resolution achieved was  $1.1 \times 10^5$  (FWHM) and the accuracy about 100–500 keV. An example of IMS results with astrophysical rp-process relevance can be found in [\[34\].](#page-7-0)

Recently, IMS was applied for the first time in a larger range of neutron-rich isotopes with unknown masses produced via uranium fission in-flight [\[35\].](#page-7-0) A kinematical speciality of the measurement is that by selecting a higher mean velocity than the one of the primary projectiles only the forward cone of the fission products is separated and transmitted. In this way the desired fission fragments are separated by the kinematical selection from all the numerous reaction products generated by fragmentation. The analysis of the data is still in progress and represents a special challenge due to the absence of reliable reference masses in this area and secondly due to the narrow range where the isochronous condition is fulfilled. To extend this range and improve the mass resolving power, an additional correlation to the measured revolution time is required. A possible solution could be to measure the velocity or the magnetic rigidity of each fragment. For this purpose, in addition to the one time-of-flight (ToF) detector [\[36\]](#page-7-0) positioned inside the ESR lattice, new detectors are needed either within the FRS, inside the ESR, or/and behind the extraction channel from the ESR [\[23\].](#page-7-0)

The validity of this concept was experimentally checked in a recent test run, in which the range of magnetic rigidities were restricted to about 3  $\times$  10<sup>-4</sup> at the dispersive mid-focal plane of the FRS, i.e., the FRS was used as a high resolution magnetic spectrometer. The preliminary analysis shows that the mass resolving power is improved by a factor of about 2 for the isochronous range and that this excellent resolution extends nearly over the entire time-of-flight spectrum. This is an important achievement, although at the expense of strongly reduced transmission.

### **4. Lifetime measurements**

Stored exotic nuclei circulating in the ESR offer unique perspectives for decay spectroscopy [\[8,20\]. S](#page-7-0)MS is ideally suited to measure decay properties of bare and few-electron fragments if



Fig. 7. First direct observation of bound-state  $\beta$ -decay ( $\beta_b$ -decay) [\[21\]. I](#page-7-0)n the left panel, Schottky frequency spectra of  $207T1^{81+}$  ions and their H-like  $\beta$  $^{207}Pb^{81+}$ , are shown at various times after injection into the ESR. The number of stored bare mother and daughter ions as a function of the storage time is presented in the right panel. The statistical errors for most data points are smaller than the size of the symbols.

<span id="page-5-0"></span>

Fig. 8. The measured electron capture decay of  $140Pr^{58+}$  to  $140Ce^{58+}$  ions is illustrated in the series of subsequent Schottky frequency spectra at regular time intervals. About six mother nuclei were initially stored. Two of them decayed via nuclear electron capture into  $140Ce^{58}$ . The time-correlated intensity changes are clearly seen. The other mother ions decayed via  $\beta^+$  decay or were lost, e.g., due to interaction with the residual gas.

the Q-value and the change in  $B\rho$  are not exceeding the storage acceptance of the ESR. For larger  $B\rho$  differences the daughter species leave the closed orbit and can be recorded by particle detectors in a dispersive magnetic dipole stage of the ESR lattice.

For the first time the decay of nuclei can be investigated in the laboratory at ionization stages prevailing in hot stellar plasmas.

It is obvious that such decay measurements have great astrophysical impact. Decay channels can be suppressed or opened depending on the ionization. In this way rare decay modes can be observed which are not accessible in neutral atoms. Dramatic modifications of the nuclear half-lives of bare isomers were observed, e.g., the half-life of the 0.58 s isomeric state in  $^{151m}$ Er is prolonged by a factor of 33(5) [\[20\].](#page-7-0)

For the first time bound and continuum  $\beta^-$  decay have been simultaneously measured in the laboratory [\[21\],](#page-7-0) see [Fig. 7.](#page-4-0) The combination of stochastic pre-cooling [\[18\]](#page-7-0) and electron cooling provides access to the spectroscopy of hot fragments with lifetimes down to about one second. For example, in the experiment on  $\beta_b$ -decay of  $^{207}T^{181+}$  ions, the half-life of the bare  $207$ Tl isomeric state was measured. The determined value of  $1.47 \pm 0.32$  s in the rest frame is in excellent agreement with the calculated prolongation (1.52 s) due to the complete suppression of the internal conversion decay branch [\[37\].](#page-7-0)

A new era for lifetime spectrometry is opened up with the measurements of single-particle decays, where the disappearance of each of the mother ions is correlated with the appearance of a daughter ion. An example of measured decays with only a few mother nuclei is illustrated in Fig. 8. A series of subsequent Schottky frequency spectra at regular intervals monitors the decay of  $140 \text{Pr}^{58+}$  to  $140 \text{Ce}^{58+}$  ( $Q_{\text{EC}} = 3388 \text{ keV}$ ) [\[17\]. A](#page-7-0)n aim of the experiment is to have initially no daughter ions and to store and cool only a few mother nuclei. In this way we want to study with single ions the basic decay properties independent of the



Fig. 9. Masses presently covered with FRS-ESR experiments and the future range with FAIR [\[13\]. T](#page-7-0)he classical r-process corridor is included for orientation. Different neutron densities (10<sup>20,23,26</sup>) and a temperature  $T_9 = 1.35$  have been assumed. The rp-path has been included according to reference [\[44\].](#page-7-0)

standard assumption that the evolution of the Schottky area is strictly proportional to the number of cooled circulating ions. The experimental conditions for this experiment were presented in [Fig. 1.](#page-1-0) The  $B\rho-\Delta E-B\rho$  method was applied to achieve the required monoisotopic in-flight separation. Interesting new features were observed in two pilot experiments which should be confirmed in an experimental program involving other fragments that decay via different decay modes. In these coming experiments we will also measure the influence of electron screening, which is of basic interest for astrophysical fusion reactions and heavy-ion decays in hot environment. Many experimental efforts were made in the past with decay and reaction studies under complex conditions [\[38\].](#page-7-0) The principal effect will be checked with new decay measurements of bare and He-like  $\alpha$ -emitters circulating in the ESR [\[39\].](#page-7-0)

#### **5. Future perspectives: ILIMA project**

Although the present experimental program at the SIS–FRS– ESR facilities has been quite successful and has led to several basic discoveries, we presently face severe limitations concerning primary beam intensities and the injection efficiency of the ESR for hot fragments. The future international facility for antiprotons and ion research FAIR [\[13\]](#page-7-0) will solve these shortcomings. A new double-ring synchrotron system (100/300 Tm) will accelerate ions up to uranium with intensities of  $10^{12}$  s<sup>-1</sup>. The beam of stable isotopes will be converted to rare isotopes with a large-acceptance superconducting fragment separator (Super-FRS) [\[40\],](#page-7-0) which will efficiently handle also the large phasespace of the fission fragments. A dedicated storage ring system [\[41\]](#page-7-0) will collect, store and cool the fragment beams with minor losses. These new facilities will allow us to substantially extend the nuclear physics research. Also a significant set of astrophysical relevant nuclei in the nucleosynthesis paths can be studied for the first time. The ILIMA-project (Isomeric beams, LIfetimes,



Fig. 10. Schottky frequency spectra of well resolved mother and daughter ions characterized by a Q-value of about 3.4 MeV. One hundred and seventy seconds after the injection into the ESR, the primary beam of mother ions was eliminated by mechanical scraping in the micrometer range.

and MAsses) [\[42\]](#page-7-0) aims for measuring the masses and half-lives of these exotic nuclides. The mass surface that is already measured and that we expect to cover with the new facility is shown in [Fig. 9.](#page-5-0) A clear goal is to cover the nuclides involved in the astrophysical r- and rp-processes.

Another goal of the ILIMA-project is to provide pure isomeric beams circulating in the new storage ring system to be investigated and used in reactions with the internal target or collision zones with other stored particles as electrons or antiprotons. An important step in this direction has been taken by removing one out of two nuclear species separated by the Q-value of 3.4 MeV (see [Figs. 8 and 10\)](#page-5-0). That was achieved by a precise mechanical scraper at a dispersive plane in the ESR. This technique is very similar to the one developed for the measurements of the horizontal beam size of cooled ion beams with micrometer resolution [\[43\]. M](#page-7-0)ore sensitive methods should provide a micrometer separation [\[43\]](#page-7-0) that would give access to beams of isomers with lower excitation energy.

### **6. Conclusion**

In summary, we have shown that the experiments with stored exotic nuclei at relativistic energies have opened a new era for mass and lifetime measurements. Our accurate mass measurements with cooled nuclei contribute to nuclear structure studies. The unique experimental condition of being able to select the atomic charge states of the fragments down to bare ions can yield new perspectives for decay spectroscopy in general and in particular for exotic decay as, e.g., bound-state beta-decay. For the first time experimental studies have been performed with bare and few-electron ions stored at relativistic energies, which is relevant for both basic nuclear structure and astrophysics.

An important experimental step towards the future has been achieved with the demonstration of a method to provide pure isomeric beams. The spatial separation of ground and isomeric states with excitation energies of at least 3.5 MeV is now a realistic objective.

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