PROCEEDINGS OF SCIENCE

Progress on direct mass measurements of exotic nuclei with the FRS-ESR facilities at GSI

Baohua Sun[*†]
Justus Liebig Universität Giessen, Heinrich-Buff-Ring 14, Giessen 35392, Germany
School of Physics and Nuclear Energy Engineering, Beihang University, Beijing 100191, China
E-mail: B.Sun@gsi.de

Heavy ion storage rings operated in an isochronous mode and/or equipped with the phase-shape cooling devices can be used for high-precision, high-sensitivity and high-efficiency mass measurements of stored nuclei. This has been achieved at GSI Darmstadt, where two complementary methods, Schottky Mass Spectrometry (SMS) and Isochronous Mass Spectrometry (IMS) were developed based on the combination of the fragment separator (FRS) and the storage ring (ESR). So far, the storage ring spectrometry has been successfully used in measuring about 300 unknown nuclear masses. In this contribution, I will discuss the progress on direct mass experiments of stored exotic nuclei, with emphasis on the first extension of the IMS in short-lived nuclear isomeric investigation, and the five new isotopes (^{236}Ac, ^{224}At, ^{221}Po, ^{222}Po, and ^{213}Tl) discovered with the SMS. Preliminary results from a pilot study on the application of a new Resonant Schottky pick-up for mass and lifetime measurements of nuclei are also reported.

8th International Conference on Nuclear Physics at Storage Rings-Stori11,
October 9-14, 2011
Laboratori Nazionali di Frascati dell'INFN, Italy

[*]Speaker.

[†]This talk represents recent work done by the FRS-ESR mass collaboration, see e.g. the author list in Refs. [11, 20]. Special thanks go to F. Bosch, H. Geissel, R. Knöbel, C. Kozhuharov, Yu.A. Litvinov, F. Nolden, W.R. Plaß, C. Scheidenberger, P. Walker, and H. Weick for valuable discussions in preparing this talk.

PoS(STORI11)003

1. Introduction

The study of exotic nuclei, far from the valley of beta stability, presents an important challenge for nuclear physicists, not only for updating or renewing our understanding of this complex nuclear many-body quantum system, but also for finding the key to the origin of heavy elements in nature. With the development in both detection technique and accelerators, direct mass measurements have also become an important tool for discovering new isomers and even new isotopes.

At GSI, the heavy ion storage ring (ESR) [1] can be coupled with a powerful fragment separator (FRS) [2], and for the first time experiments were carried out by employing it as a multi-turn time-of-flight mass spectrometry [3]. The principle of the storage ring mass spectrometry [4], in the first order, can be simply expressed as:

$$\frac{\Delta f}{f} = -\alpha_p \frac{\Delta(m/q)}{m/q} + \eta \frac{\Delta p}{p} , \tag{1.1}$$

where f, m/q, and p are the revolution frequency, mass-to-charge ratio and momentum for a circulated ion in the ring, respectively. $\eta = 1/\gamma^2 - 1/\gamma_t^2$ is the frequency dispersion function and $\gamma_t = 1/\sqrt{\alpha_p}$ is the transition point of the storage ring.

Two complementary methods to minimize the term containing the momentum spread in Eq. 1.1 have been developed [5]. In the first method, Schottky mass spectrometry [6, 7, 8, 9, 10, 11, 12], the electron cooler is used to force all circulated ions in the ring to have an identical velocity (with a velocity spread down to $\Delta v/v \sim 10^{-7}$) [13], and thus the revolution frequency is only related in the first order to their mass-to-charge ratio. The electron cooling time is directly proportional to the corresponding velocity difference of the cooler electrons and the hot fragments to the power of three ($t_{cool} \propto \Delta v^3$). Stochastic cooling is used for pre-cooling of the hot fragments [14], especially for half-life measurements [15]. Eventually, the revolution frequency is deduced from the fast Fourier transform (FFT) of the induced signals captured by a Schottky probe. Very high mass resolving power up to 2×10^6 is already obtained for this technique, however, the limitation is that nuclei with lifetime shorter than a few seconds can not be accessed due to the total cooling time of a few seconds.

While in the second method, isochronous mass spectrometry [16, 17, 18, 19, 20], the ESR is operated at the transition point γ_t, i.e., γ=γ_t for the reference particle, where γ is the Lorentz factor. Then the frequency dispersion function η vanishes to first order, and the revolution frequency does practically not depend on small changes of the particle energy (energy isochronous mode). This method is in principle suitable for measuring masses of nuclei with lifetime down to a few tens of μs. Presently, the revolution frequency is detected using a dedicated Time-Of-Flight detector, however this method is not suitable for monitoring the evolution of ion intensity due to the slow energy loss process in the detector. Recently, the technique was improved significantly by employing an additional magnetic-rigidity determination [21, 22]. The mass resolving power achieved so far is up to 2×10^5.

In this contribution, after introducing the characteristics of the storage ring mass spectrometry, I will show some results from two recent experiments focused on neutron-rich nuclei, and also report a pilot study of applying a resonant Schottky pick up.

2. Characteristics of the storage ring mass spectrometry

More details and results of the storage ring mass measurements at GSI can be found in the recent reviews [5, 23, 24]. Here, the most important characteristics of the storage ring mass spectrometry are summarized:

- **Highly charged state:** An important feature of the mass-measurement program with the FRS-ESR facilities is that the stored relativistic ions are highly charged, like the case of astrophysical plasma. Under this condition the observed half-lives of nuclei can be different from the neutral cases [25, 26], and nuclear isomers can survive much longer if the electron conversion branch is suppressed [20, 27].

- **Mapping power:** The large acceptance of the FRS-ESR facilities makes possible the storage of multi-component beams with accordingly different magnetic rigidity, and thus large-scale measurements on the chart of nuclei. It has been observed in previous experiments that up to 3 different charge states of the same isotope were simultaneously stored. The large momentum acceptance is extremely useful for monitoring the decay of fragment beams.

- **Sensitivity:** Sensitivity down to single ions is achieved for both the SMS and IMS techniques. This is especially important for measuring isotopes or isomers with very small production rates and short half-lives. A typical example is the first mass measurement of ^{208}Hg based on a single ion in hydrogen-like charge state, which then allows the extraction of the empirical p-n interaction of the last nucleons in ^{210}Pb [10]. The production cross section of about 1.5 μbarn is estimated for ^{208}Hg.

- **Resolving power**: The mass resolving power can reach up to 2×10^6 and 2×10^5 for the SMS and IMS, respectively. The high resolution of the SMS has been used to separate nuclear isomeric states with small excitation energies (about a few hundred keV) [9, 28] from the corresponding ground states, and moreover for searching new long-lived isomers [12, 29, 30]. Recently, the IMS has been extended to short-lived isomeric investigations as well [20]. In addition, stored nuclei present either in the ground state or in the isomeric state allow more dedicated studies of excitation energy, lifetime and isomeric production ratio, by taking into account the almost identical transmission, cooling and detection efficiency.

- **Precision:** A typical uncertainties of 30 keV and 100 keV have been obtained for the broadband frequency spectra in previous measurements employing the SMS and IMS, respectively. By restricting over a small frequency range, thus minimizing ion-optical nonlinearities, the precision can be further improved. For example, an mass accuracy of $\delta m/m$=4.3×10^{-8} [31] has been achieved for ^{93m}Mo, where the precision is sorely determined by the uncertainties of calibration masses.

- **Time-resolved:** A unique characteristic of SMS is that one can trace the fate of each species in the ring by the recording Schottky spectrum. This allows to explore the lifetime of exotic nuclei besides nuclear mass determination. Moreover, nuclear decays in the ring, which are represented by frequency shifts, can give a cross-check of particle identifications [32]. In the

next section, we show that with a new fast probe it is now possible to obtain time-resolved spectra as well for the IMS technique.

- **Lifetime restriction:** The present SMS is limited to explore nuclei with lifetime longer than a few seconds, while the IMS can in principle get access to all nuclei with lifetime down to a few tens of μs. The latter almost covers all the lifetime range of the nuclear chart. Furthermore, the highly-charged feature of the FRS-ESR facilities has presented the possibility to observe bare isomers with life-times that would be even shorter than μs in the corresponding neutral atoms [20].

3. Recent Research highlights

3.1 Selected results in recent two experiments

Recent nuclear mass experiments are largely motivated by the astrophysical rapid neutron-capture process (r-process) [33], which proceeds along very neutron-rich nuclei with one-neutron separation energy of about 2-4 MeV. Nuclear mass surface covered in recent experiments are shown in Fig. 1, including the first large-scale isochronous mass measurement of Uranium fission fragments [19] and the first Schottky mass measurement of heavy uranium fragments [11].

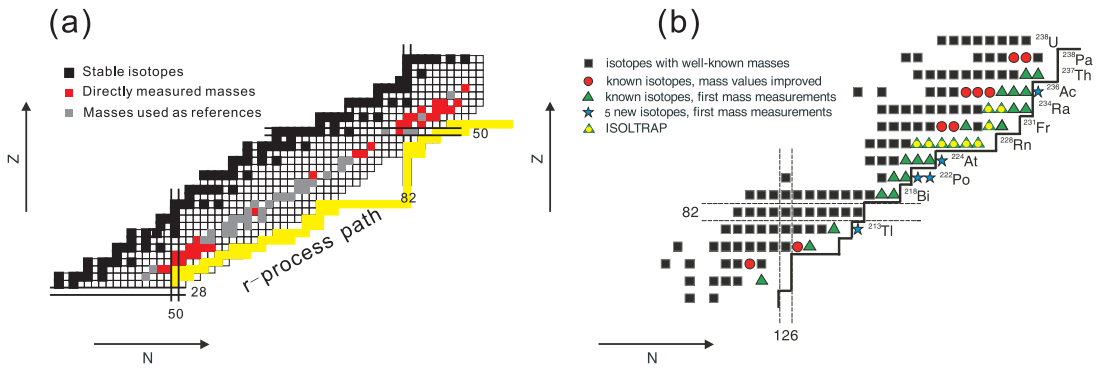

Figure 1: (a): Fission fragments measured by the IMS [19]. The references and measured nuclei in the experiment are indicated. The astrophysical r-process path [38] is shown as well. (b): Mass surface covered at the Schottky mass measurement of heavy uranium fragments [11]. The isotopes with well known masses, with masses improved, and masses first determined, as well as the five new isotopes discovered in this experiment are shown. The limit of previously known neutron-rich isotopes is indicated by the solid line. Recent results from ISOTRAP measurements [43, 44] are also shown for comparison.

One measurement is addressed to neutron-rich fission fragments with the novel IMS [19]. The masses of 35 nuclides, including 8 cases for the first time, were directly measured, and the mass surface is displayed in the panel (a) of Fig. 1. The newly measured masses indicate that the AME2003 extrapolations [34] over-bind the nuclear mass surface in the neutron rich nuclei. This systematical trend of a shifted mass surface to larger values will affect the mass model predictions which use the nuclear masses as one of their ingredients (e.g., see Refs. [35, 36]). Our new results also show a decreasing trend of shell gap energies at $N = 50$ towards ^{83}As, and this is confirmed by a penning trap measurement done at a similar time [37]. The mass values determined in this work,

furthermore, have a clear impact on the r-process nucleosynthesis calculation. A classical r-process calculation [38] under a low neutron-density astrophysical condition shows that the abundance around A = 87 has increased by a factor of 10 when including our newly measured masses [39]. A main difficulty from this experiment, however, is the availability of reliable calibration masses. With more reliable references from recent penning trap measurements included in the data analysis, mass uncertainty down to 100 keV has been achieved [40]. Possible appearance of unresolved low-lying isomeric states in neutron-rich nuclei has further complicate the data analysis.

As a part of the experiment, the IMS technique has been employed for the first time to get access to a rare nuclear isomer, 17 μs core-excited isomer in ^{133}Sb [41]. The excitation energy of 4.56(10) MeV and survival time of the isomer in the ring have been determined based on precise revolution-time measurements of a few individually stored fully ionized ions. The extended in-flight half-life of the bare ions in the ESR, which is due to the exclusion of the strong internal conversion, demonstrates that there should be another nuclear level above $17/2^+$ that identified from isomer-decay spectroscopy, thus completing the missing information on the level scheme. This measurement opens up a new half-life domain for storage-ring measurements [30].

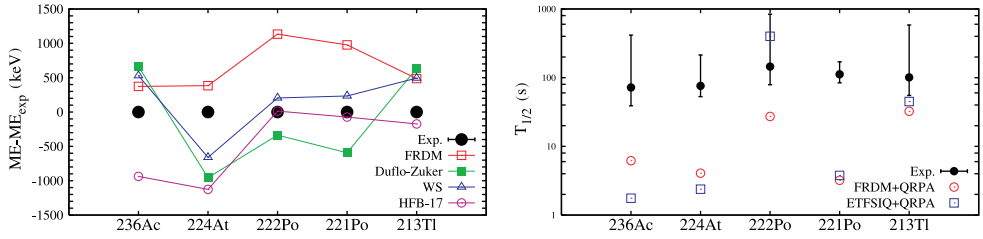

Figure 2: Left panel: Difference of the mass model predictions with experimental mass values for the five new isotopes. The mass models used are FRDM [45], Duflo-Zuker [46], WS [47] and HFB-17 [48]. Right panel: Half-lives from our experiment and the model predictions of FRDM+QRPA [49] and ETFSIQ+QRPA [50].

Another experiment was done by using the time-resolved SMS, which maps a new territory of about 150 heavy neutron-rich mass surface [11]. An overview of the harvest of this experiment is illustrated in the panel (b) of Fig. 1. About 40 nuclear masses are determined for the first time. Of them, 5 new isotopes, ^{236}Ac, ^{224}At, ^{221}Po, ^{222}Po, and ^{213}Tl, are discovered. Their masses were accurately determined with an uncertaity (1σ) of around 60 keV, and information on their decays was obtained as well. More details on the different methods of half-life determination of stored ions can be found in Refs. [15, 42]. Results from other experiments [43, 44] can serve as a cross-check for mass determinations as well as reliable references for calibration. Moreover, the important role of cold fragmentation and nuclear charge-changing reactions is manifested in this experiment by the observation of the so far most neutron-rich isotopes in the region of lead and uranium.

By taking the new isotopes as an example, we compare our new experimental mass and half-life values with the widely-used nuclear models in Fig. 2. Mass deviations from experimental data are typically around 500 keV, clearly showing the deficits of the widely-used mass models [45, 46, 47, 48]. As for the half-lives, the experimental values are in general up to one order of magnitude larger than available model predictions [49, 50]. These new experimental information will definitely contribute to the improvements of mass and half-life predictions for heavy nuclei, and thus to a more reliable theoretical modeling for the r-process nucleosynthesis.

3.2 A pilot study on the application of a new resonant Schottky pick-up

A fast and sensitive resonant pick-up [51] was built into the ESR in 2010, with the primary goal to understand the "non-exponential behavior" of electron capture decays [52]. In comparison with the standard Schottky pick-up used in previous experiments, the new probe has a significant enhanced signal to noise ratio of Schottky spectra. Consequently, much shorter measurement time is required to get one spectrum. This improvement is particularly important for detection of ions with a small charge number and/or low production rate. Another difference from the previous Schottky probe is that the working frequency of the new resonator is a factor 4 larger. Therefore the separation of neighboring frequency peaks is enhanced by the same factor, and also the time it takes to distinguish neighboring peaks is reduced by a factor of four due to the Nyquist theorem. The latter feature is critical for detecting fast changes such as short-lived nuclear decays, or for more averaging to pursue a frequency spectrum with better quality. In a pilot experiment, we have tested the application of this probe in a mass measurement. Preliminary results are report here.

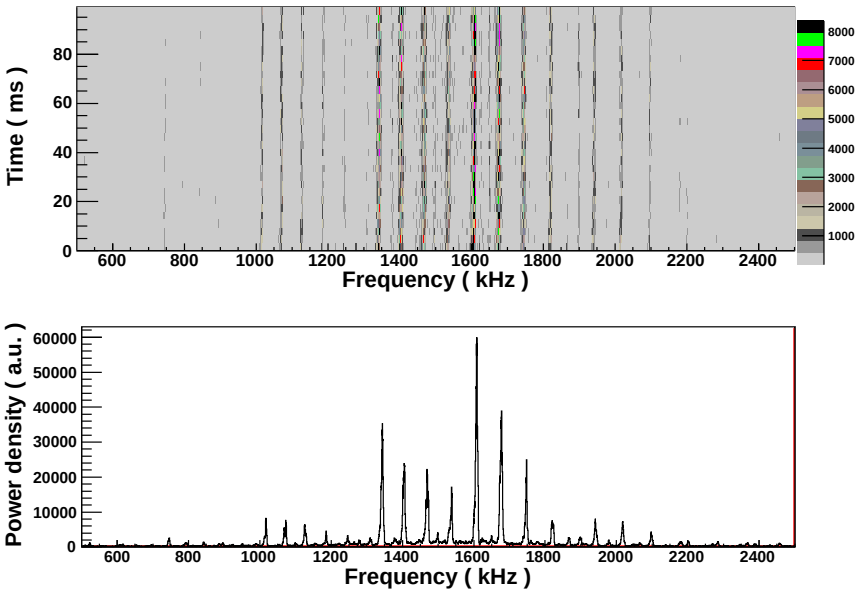

Figure 3: The time evolution of the Schottky revolution frequency spectrum and the projection of the first 16 ms measurement. Here, the raw spectrum is recorded by the new resonant Schottky pick-up.

Shown in Fig. 3 is a measured revolution frequency spectrum of ^{238}U projectile fragments centered at ^{213}Fr, where the ESR is operated at the isochronous mode with the transition point γ_t being 1.4. The advantage of the new probe is clearly shown in the Schottky spectrum. Now for the first time we are able to trace the fate of each circulated ions in the ring to the time accuracy of ms, e.g. 3.2 ms in the upper panel of Fig. 3, more than one order of magnitude reduced in comparison with previous storage ring experiments. Especially for the isochronous mass measurement, it is even possible to reveal the dispersion of momentum $\delta p/p$ of interested ions by the corresponding revolution frequency $\delta f/f$ according to $\delta p/p = 1/\eta\ \delta f/f$.

For the 16 ms spectrum shown in the lower panel of Fig. 3, a frequency resolving power of 50, 000 is obtained, and this value is almost the same to that from the standard Time-of-Flight

spectrum, where a TOF detector is used for revolution frequency determinations. A resolving power of 1.25×10^6 is obtained in the standard operational mode of the ESR with electron cooling.

As shown above, the new resonant Schottky pick-up opens a way towards precision mass and lifetime measurements of nuclei with lifetimes down to a few tens of ms. In fact, the feasibility of applying this resonator in lifetime measurements is also tested in a pilot experiment using α-emitter ^{213}Fr, but because of a very poor statistic the half-life is determined with a large uncertainty.

For the isochronous mass measurement, however, studies show that different Schottky bands will eventually overlaps due to the high working harmonic of about 125, and thus unambiguous identifications of the frequency spectra become very difficult. It is therefore essential to investigate further the application of the new probe for mass and lifetime measurements on exotic nuclei.

4. Summary

In this contribution, we have discussed the characteristics of storage ring mass spectrometries, as well as the results from recent mass measurements of neutron-rich nuclei at GSI. The newly equipped resonator Schottky pick-up has opened a new way for precision mass and half-life measurements of nuclei with lifetime down to a few tens of ms, however, further plans using well known long/short-lived nuclides need to be commissioned.

Acknowledgement: This contribution is based on the recent work of the FRS-ESR mass collaboration. BS is partially supported by NCET, NSFC(Grants No. 10975008, 11105010 and 11035007), and the Fundamental Research Funds for the Central Universities.

References

[1] B. Franzke, Nucl. Instrum. Methods B 24/25 (1987) 18.

[2] H. Geissel, et al., Nucl. Instrum. Methods B 70 (1992) 286.

[3] H. Geissel, et al., Phys. Rev. Lett. 68 (1992) 3412.

[4] F. Bosch, Lect. Notes Phys. 651 (2004) 137.

[5] B. Franzke, et al., Mass Spectrom. Rev. 27 (2008) 428.

[6] T. Radon, et al., Phys. Rev. Lett. 78 (1997) 4701.

[7] T. Radon, et al., Nucl. Phys. A 677 (2000) 75.

[8] Yu.A. Litvinov, et al., Phys. Rev. Lett. 95 (2005) 042501.

[9] Yu.A. Litvinov, et al., Nucl. Phys. A 756 (2005) 3.

[10] L. Chen, et al., Phys. Rev. Lett. 102 (2009) 122503.

[11] L. Chen, et al., Phys. Lett. B 691 (2010) 234.

[12] M.W. Reed, et al., Phys. Rev. Lett. 105 (2010) 172501.

[13] M. Steck, et al., Nucl. Instr. and Meth. A 532 (2004) 357.

[14] F. Nolden, et al., Nucl. Instr. and Meth. A 532 (2004) 329.

[15] Yu.A. Litvinov, F. Bosch, Rep. Prog. Phys. 74 (2011).

[16] M. Hausmann, et al., Nucl. Instr. and Meth. A 446 (2000) 569.
[17] M. Hausmann, et al., Hyperfine Interact. 132 (2001) 291.
[18] J. Stadlmann, et al., Phys. Lett. B 586 (2004) 27.
[19] B. Sun, et al., Nucl. Phys. A 812 (2008) 1.
[20] B. Sun, et al., Phys. Lett. B 688 (2010) 294.
[21] H. Geissel, et al., Hyperfine Interact. 173 (2006) 49.
[22] R. Knöbel, et al., AIP Conf. Proc. 891 (2006) 199.
[23] H. Geissel and Yu.A. Litvinov, Nucl. Instr. and Meth. B 266 (2008) 4176.
[24] Yu.A. Litvinov, et al., Acta Phys. Polonica B 41 (2010) 511.
[25] Yu.A. Litvinov, et al., Phys. Rev. Lett. 99 (2007) 262501.
[26] N. Winckler, et al., Phys. Lett. B 679 (2009) 36.
[27] Yu.A. Litvinov, et al., Phys. Lett. B 573 (2003) 80.
[28] C. Scheidenberger, et al., Hyperfine Interact. 173 (2006) 61.
[29] B. Sun, et al., Eur. Phys. J. A31 (2007) 393.
[30] B. Sun, et al., Nucl. Phys. A 834 (2010) 476c.
[31] Yu.A. Litvinov, et al., Hyperfine Interact. 173 (2006) 55.
[32] B. Sun, et al., Chin. Phys. C 33 (Suppl. 1) (2009) 161.
[33] E.M. Burbidge, et al., Rev. Mod. Phys. 29 (1957) 547.
[34] G. Audi, A.H. Wapstra, and C. Thibault, Nucl. Phys. A729 (2003) 337.
[35] B. Sun, et al., Sci. China Phys. Mech. Astro. 54 (2011) 210.
[36] G.J. Fu, et al., Phys. Rev. C 84 (2011) 034311.
[37] J. Hakala, et al., Phys. Rev. Lett. 101 (2008) 052502.
[38] B. Sun, et al., Phys. Rev. C 78 (2008) 025806.
[39] B. Sun, et al., Int. J. Modern Phys. E 18 (2009) 346.
[40] R. Knöbel, Ph.D. Thesis, Justus-Liebig-University Gießen, 2008.
[41] W. Urban, et al., Phys. Rev. C 79 (2009) 037304.
[42] H. Geissel, et al., Eur. Phys. J. Special Topics 150 (2007) 109.
[43] F. Herfurth, et al., Eur. Phys. J. A 25 (2005) 17.
[44] D. Neidherr, et al., Phys. Rev. Lett. 102 (2009) 112501.
[45] P. Möller, et al., At. Data Nucl. Data Tables 59 (1995) 185.
[46] J. Duflo and A.P. Zuker, Phys. Rev. C 66 (2002) 051304(R).
[47] M. Liu, et al., Phys. Rev. C 84 (2011) 014333.
[48] S. Goriely, et al., Phys. Rev. Lett. 102 (2009) 152503.
[49] P. Möller, et al., Phys. Rev. C 67 (2003) 055802.
[50] Private communication.
[51] F. Nolden, et al., Nucl. Instr. and Meth. A 659 (2011) 69.
[52] Yu.A. Litvinov, et al., Phys. Lett. B 664 (2008) 162.