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Experiments with Stored Highly Charged Ions at the Border between Atomic and Nuclear Physics

B. S. Gao[a,b,c], M. A. Najafi[d], D. R. Atanasov[b,e,f], K. Blaum[b,f], F. Bosch[c], C. Brandau[c,g,h], X. C. Chen[c,f], I. Dillmann[c,i], Ch. Dimopoulou[c], Th. Faestermann[d], P.-M. Hillenbrand[c], H. Geissel[c], R. Gernhäuser[d], C. Kozhuharov[c], O. Kovalenko[c,f], S. A. Litvinov[c], Yu. A. Litvinov[a,b,c,f,*], L. Maier[d], F. Nolden[c], J. Piotrowski[c,j], M. S. Sanjari[c,h], U. Spillmann[c], M. Steck[c], Th. Stöhlker[c,k,l], Ch. Trageser[c,g], X. L. Tu[a,b,c], H. Weick[c], N. Winckler[c], H. S. Xu[a], T. Yamaguchi[m], X. L. Yan[a,b,c], Y. H. Zhang[a], X. H. Zhou[a]

[a]*Key Laboratory of High Precision Nuclear Spectroscopy and Center for Nuclear Matter Science, Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China*
[b]*Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany*
[c]*GSI Helmholtzzentrum für Schwerionenforschung, Planckstraße 1, 64291 Darmstadt, Germany*
[d]*Technische Universität München, 85748 Garching, Germany*
[e]*CERN, 1211 Geneva 23, Switzerland*
[f]*Ruprecht-Karls Universität Heidelberg, 69120 Heidelberg, Germany*
[g]*Institut für Atom- und Molekülphysik, Justus-Liebig Universität, 35392 Gießen, Germany*
[h]*ExtreMe Matter Institute EMMI, 64291 Darmstadt, Germany*
[i]*TRIUMF, 4004 Wesbrook Mall, Vancouver, BC V6T 2A3, Canada*
[j]*AGH University of Science and Technology, Kraków, Poland*
[k]*Friedrich-Schiller-Universität Jena, 07737 Jena, Germany*
[l]*Helmholtz-Institut Jena, 07743 Jena, Germany*
[m]*Graduate School of Science & Engineering, Saitama Univ., Saitama 338-8570, Japan*

Abstract

Atomic charge states can significantly influence nuclear decay rates. Presented is a compact overview of experiments conducted at the Experimental Storage Ring ESR of GSI addressing β-decay of stored and cooled highly charged ions. Investigations of the two-body beta decay, namely the bound-state β-decay and its time-mirrored counterpart, orbital electron-capture, are discussed in more details and a special emphasis is given to the future experiment on the bound-state β-decay of fully-ionized $^{205}\mathrm{Tl}^{81+}$ nuclei.

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* Corresponding author. Tel.:+496159711758; fax: +496159712902.
E-mail address: y.litvinov@gsi.de

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1. Introduction

Rates of nuclear decays can significantly be modified in highly charged ions (HCI) (for more information see Rutherford and Soddy (1902), Emery (1972), Litvinov and Bosch (2011), Bosch et al. (2013)). Obvious examples are the decay modes involving bound electrons, like orbital electron capture (EC) or internal conversion (IC) decays, which are just disabled if the nuclei are fully stripped of electrons.

HCI, like, for instance, bare nuclei or hydrogen-like (H-like), helium-like (He-like) or lithium-like (Li-like) ions represent themselves as well-defined quantum mechanical systems in which influences and corrections due to many bound electrons are just absent (Bühring (1965), Dzhelepov et al. (1972)). Decays of such systems offer clean conditions for investigations of effects of the electron shell on the decay characteristics. Some decay modes known in neutral atoms can become forbidden in HCI and vice versa new decay modes can become allowed. Also in nuclear astrophysics, the understanding of decays of HCI is essential since the nucleosynthesis processes proceed at high temperatures and densities at which the atoms are ionized (Burbidge, Burbidge, Fowler and Hoyle (1957), Bahcall (1962), Blake et al. (1973), Takahashi and Yokoi (1983, 1987), Käppeler et al. (1998), Bertulani and Gade (2010), Langanke and Schatz (2013)).

However, experimental investigations of radioactive decays of HCI are not a trivial task (Bosch (1992, 2006), Bosch et al. (2006, 2013), Litvinov et al. (2011), Bosch and Litvinov (2013)). They require on the one side the possibility to produce and separate exotic nuclei in a well-defined high atomic charge state. On the other side, it is indispensable to be able to keep this charge state for an extended period of time. The latter should be sufficiently long to allow the ions to decay. Due to these experimental challenges, decay studies of radioactive HCI are conducted presently only at GSI Helmholtz Center in Darmstadt (GSI), Germany. In this contribution we briefly review the so far conducted relevant experiments. Furthermore, we give an outlook for the future experimental programs at GSI as well as at other facilities worldwide.

2. Experiment

GSI is a heavy-ion accelerator complex. The relevant part of its high-energy facility consists of a heavy-ion synchrotron SIS18 (Blasche et al. (1985)), fragment separator FRS (Geissel et al. (1992)), and an experimental storage ring ESR (Franzke (1987)). Primary beams of any stable isotope can be accelerated to a maximum magnetic rigidity of 18 Tm, extracted from the SIS18 and focused on a thin production target in front of the FRS. Projectile fragmentation and, in the case of Uranium primary beam, also projectile fission reactions are typically used to produce secondary nuclei of interest (Bernas et al. (1994), Geissel et al. (1995), Enqvist et al. (1999), Mei et al. (2014)). Owing to relativistic energies, the fragments emerge the production target as highly charged ions (Scheidenberger et al. (1998), Scheidenberger and Geissel (1998)). The production of a specific ionic charge state can be optimized by varying the energy of the primary beam, target thickness, and target material. The FRS is employed to efficiently separate the ions of interest from inevitable contaminations by other produced fragments as well as from other ionic charge states. On the one hand, the FRS is capable to efficiently collect the produced fragments and to transmit them as a cocktail beam. On the other hand, by employing the atomic deceleration of ions in specially shaped energy degraders, purification of clean mono-isotopic beams is possible as well (Geissel et al. (1992, 2002), Weick et al. (2002), Scheidenberger et al. (2006)).

The separated beams of HCI of interest are injected into the ESR (Geissel et al. (1992), Geissel (1999)). The ESR has a circumference of 108 m and is an ultra-high vacuum machine with a rest gas pressure of $\sim 10^{-11}$-10^{-12} mbar. The latter is an essential prerequisite for preserving the atomic charge state of the stored ions. The ESR is equipped with stochastic (Nolden et al. (1997, 2004), Nolden (2009)) and electron (Poth (1990), Steck et al. (1996, 2004)) cooling systems, which allow for reduction of the inevitable velocity spread of injected ions due to the reaction process. For beam intensities of below a few thousand ions, the initial velocity spread is reduced by first stochastic pre-cooling and then electron cooling to about $\delta v/v \sim 10^{-7}$ (Steck et al. (1996, 2003)) within a very few seconds (Geissel et al. (2004, 2006)). Ions with sharp velocity distributions can unambiguously be identified by their revolution frequencies. This is the basis of the so-called Schottky Mass Spectrometry (Borer et al. (1974)) which is successfully applied at the ESR for high-precision mass measurements of exotic nuclides (Radon et al. (1997, 1999, 2000), Geissel et al. (1999, 2001, 2007), Litvinov et al. (2001, 2004, 2005, 2005, 2006, 2007), Attallah et al. (2002), Novikov et al.

(2002), Geissel and Litvinov (2005, 2008), Bosch et al. (2006), Knöbel et al. (2007), Weick et al. (2007), Chen et al. (2009, 2012), Shubina et al. (2013)).

The frequencies are measured either with non-destructive Schottky detectors, which provide simultaneous information on the frequencies and intensities of all stored ions (Nolden et al. (2011), Sanjari et al. (2013)), or with particle detectors, which block specific orbits in the storage ring (Klepper et al. (1992), Klepper and Kozhuharov (2003)). The former detectors have no restrictions on the particle numbers and can work with single stored ions as well as with milli-Ampere-beams (Nolden et al. (2006)). Furthermore, they allow for redundant measurements of the decay of the parent ions and the growth of the number of the daughter ions at the same time (Litvinov et al. (2007)). The latter detectors are typically employed to detect daughter ions after the decay and are often used in the cases when the orbits of the daughter ions lie outside the storage acceptance of the ring (Ohtsubo et al. (2005)).

Alternatively, Isochronous Mass Spectrometry can be applied to measure the frequencies of the stored ions (Hausmann et al, (2000, 2001), Stadlmann et al. (2004), Geissel et al. (2006), Franzke et al. (2008), Sun et al. (2008, 2009), Münzenberg et al. (2010)). The IMS does not require electron cooling and is ideally suited to investigate the shortest-lived nuclei with half-lives as short as a few tens of microseconds. The frequencies of each individual stored ion are measured with a dedicated secondary-electron detector (Trötscher et al. (1992)). Due to energy loss in the detector, the ions can survive in the ring only a few hundreds of revolutions (Mei et al. (2010)). With the development of highly sensitive Schottky detectors, their possible application in the IMS is being discussed (Sanjari et al. (2013)). It is necessary to note that the IMS is successfully applied not only at the ESR but also at the experimental cooler-storage ring CSRe (Xia et al. (2002), Xiao et al. (2009)) at the Institute of Modern Physics in Lanzhou (Tu et al. (2011, 2011, 2014), Zhang et al. (2011, 2012, 2013), Yan et al. (2013), Xu et al. (2013), Shuai et al. (2014)) and will become the main operation mode of the RI-RING at RIKEN (Yamaguchi et al. (2008, 2013)).

3. Previous results

In this section we summarize the results obtained in various experiments at the ESR. The values were collected in the 2011 Nuclear Wallet Cards and can be found at the National Nuclear Data Center (NNDC, http://www.nndc.bnl/gov/).

3.1. Half-lives of long-lived isomeric states

Isomers are metastable nuclear states (Walker and Dracoulis (1999, 2001), which can de-excite to the corresponding ground states by either internal conversion (IC) or internal transition (IT) or can undergo beta decay. In fully ionized atoms, all bound electrons are removed and the de-excitation through IC is impossible. Therefore, the lifetimes of isomers can dramatically increase, which was accurately measured in the ESR for ^{144m}Tb, ^{149m}Dy, and ^{151m}Er isomeric states (Litvinov et al. (2003)). Such measurements allow for high precision determination of the conversion coefficients. Furthermore, weak gamma decays can be studied.

Interesting cases are the $0^+ \rightarrow 0^+$ de-excitations, which, e.g., connect the ground and the first excited states in neutron-deficient lead nuclei (Andreyev et al. (2000)). Such transitions are highly converted and in the absence of bound electrons shall significantly be hindered. For instance, in the rapid proton capture nucleosynthesis process (rp-process) in Novae, such excited 0^+ states in bare nuclei can have sufficiently long lifetimes and thus significantly modify the processing speed (Novikov et al. (2001)).

New decay modes can open up in few-electron ions, like, e.g., bound internal conversion, which was observed in HCI in single pass experiments (Phillips et al. (1989, 1993), Attallah et al. (1997)). In a storage ring, the ions are stored in the ground hyperfine states having thus well-defined total angular momentum (Seelig et al. (1998), Nörtershäuser et al. (2013)). The latter leads to the fact that the conservation of the total angular momentum has to be considered and – allowed in neutral atom transitions – may become forbidden in HCI (Folan and Tsifrinovich (1995)). In future experiments, one can consider investigating of bound electron-positron decay, where the created electron is captured on a free orbital while the positron is emitted to continuum.

Single particle sensitivity of the storage ring spectrometry allows for the search of new isomers. The advantage is that very long-lived isomers with very small production yields can unambiguously be identified. Several isomers were discovered meanwhile at the ESR (Irnich et al. (1995), Liu et al. (2006), Sun et al. (2007, 2010, 2010), Chen et al. (2010, 2013), Reed et al. (2010, 2012, 2012)).

3.2. Beta decay of highly-charged ions

Already at the time of the conceptual design of GSI accelerator facility, investigation of beta decay of HCI was one of the main motivations to construct the ESR. First experiments at the ESR in 1992 addressed beta decay of bare ^{19}Ne where a pure three-body β^+ decay channel is measured in the absence of electrons (Geissel et al. (1992), Bosch (1992)). Meanwhile the decays of several fully ionized systems were studied on both sides of the valley of beta stability (Geissel et al. (1992), Attallah et al. (2002), Reed et al. (2010), Chen et al. (2010)).

Special attention is devoted to investigations of two-body decays, since here the storage rings offer unprecedented experimental conditions. A striking example of such studies is the bound state beta decay, β_b-decay (Daudel et al. (1947), Bahcall (1961), Takahashi and Yokoi (1987)). In this β^--decay mode the emitted electron occupies one of the free bound orbitals instead of being emitted into continuum. It is clear that any significant decay probability is only existent in highly charged ions, which offer electron vacancies in the inner shells. This leads to the situation that some decay energy is saved and the decay rates can change dramatically as compared to the ones known in neutral atoms. For instance, fully ionized ^{163}Dy^{66+} nuclei decay within merely 50 days while the neutral ^{163}Dy atoms are stable (Cohen et al. (1987), Jung et al. (1992)). The measured β_b-decay of ^{163}Dy^{66+} nuclei allowed for the determination of the temperature during the slow-neutron capture process of nucleosynthesis (Bosch and Litvinov (2008)). Neutral ^{187}Re atoms have a very long half-life of 42 Gy, which changes to 33 years if all electrons are removed (Bosch et al. (1996)). As a consequence, the previous consideration to employ ^{187}Re/^{187}Os pair as a clock to determine in a model-independent way the age of the Universe had to be revised (Arnould et al. (1984), Takahashi (1997), Bosch (1999)). Recently, simultaneous measurements of the three-body β^--decay and the two-body bound state beta decay channels in ^{207}Tl^{81+} and ^{205}Hg^{80+} nuclei allowed for the determination of β_b/β^- ratios, which shall be analogous to the well-known EC/β^+ ratios (Ohtsubo et al. (2002, 2005), Faber et al. (2008), Kurcewicz et al. (2010)). In section 4 we discuss in detail the experiment on the bound state beta decay of ^{205}Tl^{81+} nuclei, which was proposed about two decades ago (Henning et al. (1985), Pavicevic (1988)) and which is now being prepared.

The two-body beta decay mode on the neutron-deficient side of the nuclidic chart is orbital electron capture, EC (Bambynek et al. (1977)). EC is disabled in bare nuclei. Recently, EC of H- and He-like ions was accurately measured in ^{122}I, ^{140}Pr, and ^{142}Pm ions (Litvinov et al. (2007), Winckler et al. (2009), Atanasov et al. (2012, 2013)). Surprisingly, H-like ^{140}Pr^{58+} and ^{142}Pm^{60+} ions decay by a factor ~1.5 faster than the corresponding He-like ^{140}Pr^{57+} and ^{142}Pm^{59+} ions, and even neutral atoms. This counterintuitive effect can be traced down to the conservation of the total angular momentum and to the fact that the ions in the ESR are stored in the ground hyperfine state (Patyk et al. (2008), Kurcewicz et al. (2008), Winckler et al. (2010, 2011)). In turn this selectivity of the ESR can be used to address forbidden decays and other subtle effects in beta decay (Folan and Tsifrinovich (1995), Litvinov (2008, 2009) Siegen-Iwaniuk (2011)). We note that similar effects were seen in muon capture (Promakoff (1959)).

However, the most intriguing result remains the observation of the modulated EC decays in H-like ^{122}I^{52+}, ^{140}Pr^{58+}, and ^{142}Pm^{60+} ions (Litvinov et al. (2008), Bosch and Litvinov (2010), Kienle et al. (2013)). If the ~10 s modulations on top of the exponential decay are confirmed in future experiments, this result can lead to new interesting physics, since such modulations are not expected within the present understanding of the electro-weak interaction.

3.3. Alpha-decay of highly-charged ions

We like to note also the proposed investigations of alpha decay of HCI. It is suggested to address possible tiny variations in the Q-values and half-lives of fully-ionized alpha emitters due to the effect of electron screening. For more details see Musumarra et al. (2009), Nocifiro et al. (2012), Patyk et al. (2008).

4. Bound-state β-decay of bare ^{205}Tl^{81+}

A highly desirable but still missing experiment is the determination of the half-life of β_b-decay of bare ^{205}Tl^{81+} (see Fig. 1) (Henning et al. (1985)). ^{205}Tl in lorandite ($TlAsS_2$) at Allchar mine is used as a long-time solar neutrino dosimeter in the LOREX project (Pavicevic (1988)). ^{205}Tl nuclei transmute into ^{205}Pb nuclei by (solar) neutrino capture via the (Kondev (2004)) ^{205}Tl + ν_e ($E_{\nu e}$ > 52 keV) → ^{205}Pb* (2.3 keV) + e^- reaction, where the threshold of the neutrino energy (52 keV) is by far the lowest compared to other experiments measuring solar neutrino flux. The LOREX project renders the product of mean solar neutrino flux $\Phi_{\nu e}$ and the capture cross section $\sigma_{\nu e}$ within 4.3 My $<\Phi_{\nu e} \cdot \sigma_{\nu e}>$, where both the neutrino flux and capture cross section remain unknown. However, the neutrino capture probability of the ^{205}Tl atoms and the β_b-decay of bare ^{205}Tl^{81+} nuclei share the same nuclear matrix element. Hence, the measurement of the β_b -decay probability of bare ^{205}Tl^{81+} nuclei provides the (unknown) neutrino capture cross section in ^{205}Tl atoms.

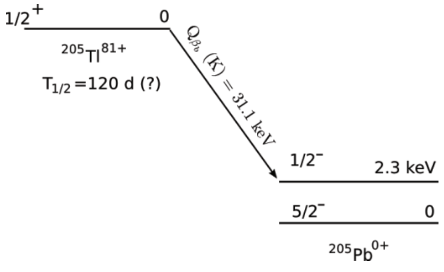


Figure 1. Bound-state beta decay scheme of bare ^{205}Tl^{81+} ions. The estimated half-life, $T_{1/2}$ ≈120 d, shall be verified experimentally. The data is taken from Kondev (2004), Litvinov and Bosch (2011) and references therein.

Furthermore, ^{205}Pb is the only purely s-process short-lived (10^7 years) radioactivity (SLR) alive in the early solar system which gives insight into nucleosynthesis prior to the Sun's birth. The expected abundance ratio of ^{205}Pb and ^{204}Pb in interstellar medium (ISM) is (Huss et al. (2009))

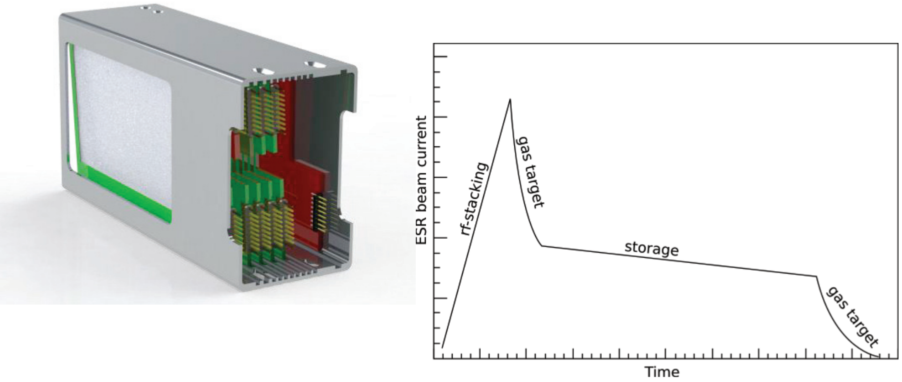
$$\frac{N_{205}}{N_{204}} = (k+2)\frac{P_{205}}{P_{204}}\frac{\tau_{205}}{T} \tag{1}$$

where N_{205} and N_{204} are the ISM abundances of ^{205}Pb and ^{204}Pb, respectively, P_{205}/P_{204} is the production ratio of the two species at their stellar source, τ_{205} is the mean lifetime of ^{205}Pb (21 My), and T is the age of the Galactic disk (~8.5 Gy). The parameter k is the infall parameter with typical values in the range 1 to 3. The production ratio P_{205}/P_{204} is expected to be in the order of unity (Blake et al. (1973)) and the abundance ratio N_{205}/N_{204} (~0.0025) agrees reasonably well with the value $(1\pm0.4)\times10^{-3}$ measured by Baker et al. (2010). However, the production rate of ^{205}Pb could be strongly reduced by free electron capture from the 2.3 keV first excited state in ^{205}Pb (Blake and Schramm (1975)). In this case, the measured value of the abundance ratio N_{205}/N_{204} would on the one hand significantly exceed what should be expected from Eq. 1. Then, an input of ^{205}Pb material from a special stellar source just prior the Sun's birth has to be assumed. On the other hand, the reduction of ^{205}Pb production rate might be counter-balanced by the β_b-decay of bare ^{205}Tl^{81+} nuclei back to the 2.3-keV first excited state of ^{205}Pb nuclide. This may in turn rule out the assumption for a special ^{205}Pb input source. The measurement of the half-life of the β_b-decay of bare ^{205}Tl^{81+} nuclei is thus crucial to clarify this issue.

The measurement of the β_b-decay of bare ^{205}Tl^{81+} nuclei will exploit a similar technique as applied for the first observation of the β_b-decay of ^{163}Dy^{66+} (Jung et al. (1992)). A secondary beam will be used in this experiment. Primary ^{206}Pb beam will be accelerated in the SIS18 to several hundreds MeV/u. Bare ^{205}Tl^{81+} ions will be produced in a Be target in front of the FRS. They will be separated by means of Bρ-ΔE-Bρ separation through the FRS and will finally be injected into the ESR where the measurement of their half-life will be performed. Among the contaminations produced in the target, a special care shall be taken of the H-like ^{205}Pb^{81+} ions (β_b-decay daughter nuclei of ^{205}Tl^{81+}). The FRS should be tuned such that the number of ^{205}Pb^{81+} ions injected into ESR is less than 1% of that of ^{205}Tl^{81+}. After the rf-stacking in the ESR, the number of stored ^{205}Tl^{81+} ions in the ESR can reach up to 10^6, which is required to produce up to a few hundreds of decays per hour assuming the half-life of $^{205}\mathrm{Tl}^{81+}$ nuclei to be about 100 d. To eliminate $^{205}\mathrm{Pb}^{81+}$ ions transmitted through the FRS, a gas target (Petridis et al. (2011)) will be used to strip the last electron and thus remove the H-like $^{205}\mathrm{Pb}^{81+}$ ions from the ESR. During the storage of the $^{205}\mathrm{Tl}^{81+}$ ions in the ESR, daughter nuclei $^{205}\mathrm{Pb}^{81+}$ are continuously produced but stay "hidden" under the frequency trace of the mother nuclei because the m/q values of these two species are too close to be resolved in the Schottky spectra. After storing the mother nuclei for an extended period of time, the gas target will have to be used again to remove the electron in the H-like $^{205}\mathrm{Pb}^{81+}$ daughter ions produced via the β_b-decay of $^{205}\mathrm{Tl}^{81+}$ nuclei.

A dedicated silicon detector will be employed to detect and count the number of daughter ions in the ESR. Due to the limitations brought about by the ultra-high vacuum environment of the ESR, special pockets have been designed and installed on a chamber after the first dipole downstream of the gas-jet target of the ESR. These pockets can accommodate particle detectors, and move near or far from the coasting beam of the ESR. The new design of the particle detector includes: a stack of eight silicon pad detectors (0.5 mm thick each), a double-sided silicon strip detector (DSSD) (0.3 mm thick), and a CsI scintillator (10 mm thick). The CsI scintillator is read out using a large-area silicon photo-diode, and the DSSD has 60 × 40 strips on the p and n sides, respectively, which are all connected to resistive chains to reduce the number of readout channels. The total thickness of the detectors is sufficient to stop bare heavy ions (Z>80) with energies up to 400 MeV/u.

This detector can combine different methods of ion identification used in the past experiments; namely, the position information from the DSSD and the energy deposit from the silicon pads (Bosch et al. (1996)), or the multiple sampling of the energy deposit (Ohtsubo et al. (2002, 2005)). In addition, for the ions that stop in the CsI scintillator, one can use the ΔE/E method. Figure 2 (left) shows the designed view of the detectors placed in a dedicated aluminum frame. The active area of the silicon detectors is 60 × 40 mm^2, which is large enough to detect both the daughter ions and possible contaminations. Moreover, the detector provides around 70 readout channels that contain detailed information about the impinging particles.



A schematic illustration of the experimental procedure to be adapted in this experiment is shown in Fig. 2 (right).

Figure 2. (Left) The design of the new detection system for the β_b-decay of $^{205}\mathrm{Tl}^{81+}$ to $^{205}\mathrm{Pb}^{81+}$. (Right) Schematic illustration of the experimental procedure to be adapted in the half-life measurement.

5. Summary and Outlook

Heavy-ion storage-cooler rings employed for storing exotic ions have proven to be excellent tools to investigate radioactive decays of the latter. Still the ESR at GSI is presently the only facility to perform such kinds of measurements. In addition to the successful mass measurements program at the CSRe in Lanzhou, also the lifetime spectroscopy is being commissioned now (Zang et al. (2011)). Furthermore, several new storage ring projects were launched which will be able to study properties of highly charged exotic nuclei. These are the TSR@ISOLDE at CERN (Grieser et al. (2012)), RI-RING at RIKEN (Yamaguchi et al. (2008, 2013)), and HIAF in China (Yang et al. (2013)). Last but not least, there is a new storage ring complex FAIR (Henning et al. (2001)) being constructed at the GSI location. We note that decay studies of HCI were foreseen in electron-ion beam traps (Elliott (1993)) as well as are planned in ion traps, like, e.g., HITRAP at the ESR (Blaum (2006), Kluge et al. (2008)).

At FAIR, the exotic nuclei will be produced and separated at the new superconducting fragment separator Super-FRS (Geissel et al. (2003), Winkler et al. (2007)) and injected into the storage rings. The physics program at future storage rings is rich and goes beyond investigations of nuclear ground-state properties, see, e.g., Krücken et al., (2005, 2006), Kalantar-Nayestanaki et al. (2009), Antonov et al. (2011), Moeini et al. (2011), Stöhlker et al. (2011), Litvinov et al. (2013), and von Schmid et al. (2014). Decays of highly charged ions will be studied within the ILIMA experiment (Walker et al. (2005, 2013)). Short-lived nuclei will be investigated in the collector ring by applying the IMS or particle detectors (Dillmann and Litvinov (2011)). For longer-lived species, it was planned to stochastically precool them in the CR and then transport to the new storage ring NESR for precision investigations. However, the NESR is presently out of the scope of the initial version of FAIR and will thus be delayed (Stöhlker et al. (2013, 2014)). Therefore, instead of the NESR, the ions will be sent to the high-energy storage ring HESR. It was shown, that the HESR can store HCI, and, e.g., EC or β_b-decay experiments can easily be conducted.

Important to note, that the FRS and ESR will remain operational until they are surpassed by the Super-FRS and NESR. If a direct connection between FRS-ESR and the HESR would exist (Stöhlker et al. (2014)), then the beams of long-lived HCI, e.g., ^{205}Tl^{81+} ions, could be purified and pre-cooled in the ESR, transmitted and accumulated in the HESR, which is then ideally suited to measure long half-lives.

Furthermore, a low energy storage ring CRYRING is being constructed downstream the ESR (Stöhlker et al. (2013)), where unique atomic and nuclear physics experiments with exotic nuclei will become possible (Zhong et al. (2010), Brandau et al. (2009, 2010, 1012, 2013)). With CRYRING, ESR, and HESR, FAIR will offer stable and radioactive HCI in a very wide and continuous range of energies from a few hundreds keV/u (CRYRING) until 5-6 GeV/u (HESR) (Stöhlker et al. (2013, 2014)).

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