

# Isotope Shifts in Dielectronic Recombination: From Stable to In-flight-Produced Nuclei

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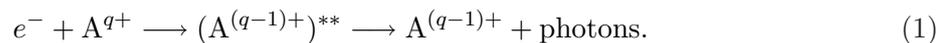
**Abstract.** The study of isotope shifts and hyperfine effects in dielectronic recombination (DR) resonance spectra strikes a conceptually new path for investigations of nuclear properties such as charge radius, spin, magnetic moment of nuclei or lifetimes of long-lived excited nuclear states. A series of DR experiments with heavy three-electron ions (Li-like) was performed at the heavy-ion storage ring ESR of the GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany. In a pilot experiment the two stable isotopes  $A=142$  and  $A=150$  of neodymium  ${}^A\text{Nd}^{57+}$  were investigated. From the displacement of DR resonances the isotope shifts  $\delta E^{142,150}(2s - 2p_{1/2}) = 40.2(3)(6)$  meV and  $\delta E^{142,150}(2s - 2p_{3/2}) = 42.3(12)(20)$  meV for  $2s - 2p_j$  transitions of the Li-like ions have been obtained. An evaluation within a full QED framework yielded a change in the mean-square charge radius of  ${}^{142,150}\delta\langle r^2 \rangle = -1.36(1)(3)$  fm<sup>2</sup>. At GSI, in addition to stable isotopes, in-flight synthesized radioisotopes can be studied as well. The production of radioisotopes of interest, the subsequent separation in the storage ring ESR and first DR experiments with the exotic nuclei  ${}^{237}\text{U}^{89+}$  and  ${}^{234}\text{Pa}^{88+}$  ( $Z=91$ ) are presented. The paper is concluded with a brief outlook at future DR experiments with heavy radioisotopes at the ESR.

## 1. Introduction

Dielectronic recombination (DR) is one of the processes that benefitted most from the advent of heavy ion storage rings at the end of the 1980s. Such DR studies can be carried out with

almost any desired charge state up to the most extreme case of one-electron uranium. At the heavy ion storage ring ESR of the GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany, in addition to stable isotopes, unstable nuclides are available for experiments as well. The understanding of the DR process and the experimental techniques at heavy ion storage rings are nowadays well-advanced, making it a recognized precision tool for application in such diverse topics as atomic physics, fundamental interactions,—in particular QED [1–3]—, plasma physics, astrophysics [4, 5] and lifetime studies of atomic levels [6, 7]. A more in-depth discussion is given in the contributions of A. Müller and A. Wolf [8, 9] as well as in the recent review article by A. Müller [4]. The latter article and [10–12] also give a broad account of DR work performed at electron beam ion traps (EBITs). In the following, we focus on the recent progress achieved at the ESR in leveraging DR as a powerful atomic physics tool for applications in isotope shift (IS) and hyperfine splitting (HFS) studies.

The process under investigation, namely DR, is an atomic resonant two-step electron-ion reaction. The initial step termed dielectronic capture, DC, is a time-inverse to Auger emission. The doubly excited intermediate state formed in the DC process can either re-autoionize or decay via the emission of photons. In the latter case, the ion is stabilized to a state below the autoionization threshold, and DR is completed:



In a typical DR storage ring experiment, the resonant character of DC is utilized to obtain spectroscopic information. On this note, DR storage ring experiments are “inverse Auger spectroscopy”. In Auger emission the de-excitation proceeds via emission of electrons with characteristic kinetic energies that are precisely determined. Conversely, in DC a free electron that possesses a matching characteristic energy can recombine resonantly with an ion and at the same time excite a bound electron. Electron-ion collision energies that meet the resonance condition lead to an enhanced number of recombined ions. In the storage ring experiments either the electron cooler or—as a distinct feature of the TSR ring in Heidelberg—a dedicated electron target, provide the free electrons for the collisions. Electron cooling conditions, i.e., equal electron and ion velocities, denote “zero” relative collision energy. Non-zero relative energies are introduced by deliberate detuning the electron beam from cooling by employing fast series of deceleration or acceleration voltages for the electrons. The detection efficiency of the recombined ions is close to unity since ions that captured an electron in the cooler/target are separated in the next ring bending magnet and are then registered using fast particle counters. Further experimental details for the set-up at the ESR storage ring can be found in [1, 13–15]. A very high energy resolution is achieved at low collision energies leading to a remarkable experimental precision [2, 3, 16]. For the three-electron (“Li-like”) ions considered in this work the energetically lowest excitations  $2s - 2p_j$  ( $j = 1/2, 3/2$ ) are  $\Delta n = 0$  intra L-shell. The DC of the free electron proceeds into Rydberg states where the gained binding energy partially cancels the excitation energy resulting in resonances typically starting already at collision energies of a few eV, —and in a few selected cases even at energies well below one eV.

## 2. DR as a tool for isotope shift and hyperfine studies of few-electron ions

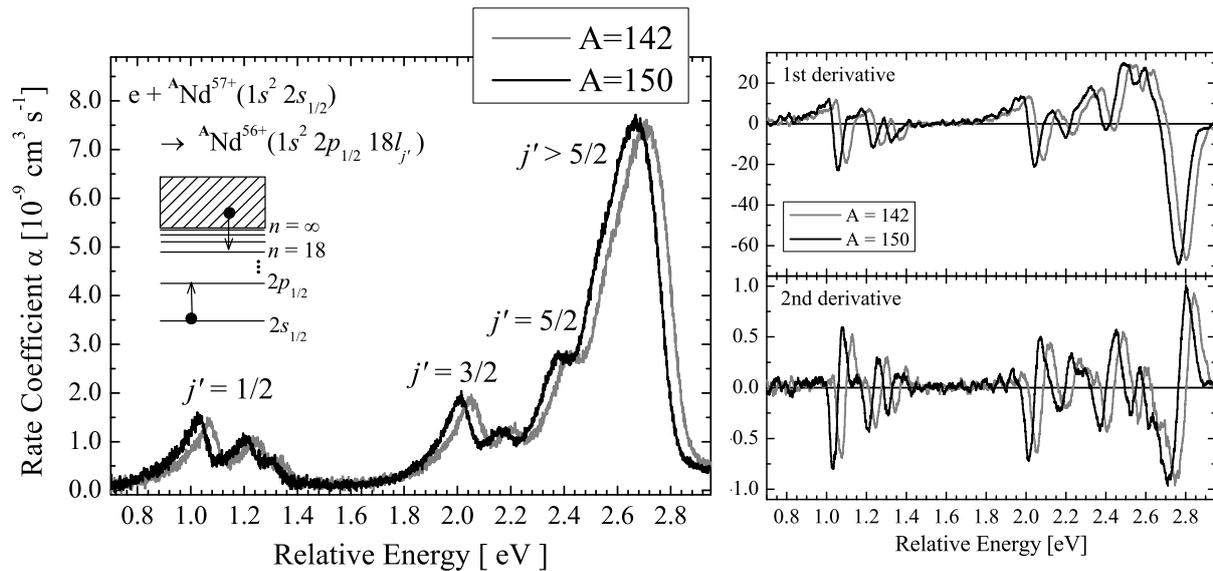
Besides the obvious impact in the field of nuclear physics we wish to emphasize the importance of a detailed knowledge of nuclear parameters for atomic physics and adjacent fields. For example, nuclear effects play an important role in determining the isotopic abundances in stellar objects [17–19], in quantum chemistry [20] and most notably in high-precision experimental tests of fundamental interactions in very heavy highly charged ions [21–23]: Highly charged ion physics ultimately aims at the understanding of matter at extreme conditions, such as QED in strong electromagnetic fields or the search for parity violation in heavy ions and, thus, testing of the

standard model. As a consequence of the large scaling of nuclear size contributions to atomic transition energies with the fifth to the sixth power of the nuclear charge,  $Z^{5\cdots 6}$ , the finite nuclear size effect exceeds those sought-after. The importance of nuclear parameters for conclusive atomic physic experiments is most easily underpinned with two examples: (a) In [24] a theory study is presented that re-examined the available experimental nuclear data for  $^{238}\text{U}$  and that explicitly includes nuclear deformation in precision QED calculations in heavy ions. For  $^{238}\text{U}$ , the heaviest naturally occurring element and the most important protagonist of strong-field QED studies an improvement in accuracy by a factor 3 in the Lamb shift calculations of highly ionized uranium is obtained. (b) Due to the near degeneracy of the  $1s2p\ ^3P_0$  and  $1s2s\ ^1S_0$  levels with opposite parity and its high nuclear charge, the two-electron uranium  $\text{U}^{90+}$  is also considered as a prospect candidate for future atomic parity violating experiments [25]. It is suggested to use the isotopic dependence of this level spacing, in order to fine-tune the effect [26]. For heavy ions ( $Z > 50$ ), isotope shift measurements by means of DR have the potential to complement existing techniques and partially remedy their shortfalls. The following list summarizes a few important benefits of DR relevant to IS and HFS studies:

- (i) Radioisotopes and isomers can be investigated, provided their lifetime is long enough for storage and cooling (present ESR  $\sim 30\text{-}60$  s). At the successor of the ESR, the upcoming NESR of the future “Facility for Antiproton and Ion Research” (FAIR) [34] lifetimes down to  $\sim 1$  s will be accessible.
- (ii) The sensitivity of DR allows for experiments with  $10^3 - 10^4$  stored ions (see below).
- (iii) The interpretation of the atomic data with respect to the nuclear parameters is clear and without ambiguity. The evaluation can be performed within a full QED framework since few-electron ions with simple atomic configurations are studied (e.g., “Li-like” with 3 electrons) [15, 24].
- (iv) In DR, the nuclear effects do not lead to a shift of a single line. Instead, a full resonance pattern of typically more than 50 well-resolved structures is displaced by the same energy value, i.e. the one of according underlying core excitation (see Fig. 1 and [15]).
- (v) Besides investigations of isotope rows of a particular element, studies along isotone chains (same number of neutrons) or measurements for different isoelectronic chains (same electron configuration) are possible. This offers the opportunity to disentangle nuclear and atomic contributions.
- (vi) Using intensity ratios of DR resonances, lifetimes of atomic metastable or nuclear isomeric states can be determined [6, 7].

### 3. Isotope shift in the DR of Li-like $^{\text{A}}\text{Nd}^{57+}$ ions (A=142, 150)

The general feasibility of the DR isotope shift technique for the measurement of nuclear physics parameters was demonstrated by our collaboration for the case of three-electron neodymium ions [15]. For both isotopes  $^{142}\text{Nd}^{57+}$  and  $^{150}\text{Nd}^{57+}$ , we determined the energy shifts of DR resonances associated with excitations of the  $2s$  valence electron, i.e.  $2s \rightarrow 2p_{1/2}$  and  $2s \rightarrow 2p_{3/2}$ . In the c.m. electron-ion collision energy range of 0–42 eV covered in the study, isotope shifts of sharp resonant structures  $^{\text{A}}\text{Nd}^{56+}(1s^2 2p_{1/2} nl_{j'})^{**}$  ( $n = 18 \dots 21$ ) and  $^{\text{A}}\text{Nd}^{56+}(1s^2 2p_{3/2} 8l_{j'})^{**}$  populated during the DR process were observed (see Fig. 1 and [15]). Characteristic values of the resonance spectrum like maxima, minima and inflection points were deduced from the 1st and 2nd derivatives (Fig. 1, right panel). This newly-developed evaluation scheme makes use of the feature-richness of a DR spectrum. In a relative measurement such as the present IS experiment the effect due to the presence of the Rydberg electron is negligibly small. Therefore, all characteristic features associated to a particular core transition, i.e., the  $2p_{1/2}$ - or  $2p_{3/2}$ -resonances possess the same IS, namely the one of the transition  $2s - 2p_{1/2}$  or  $2s - 2p_{3/2}$ , respectively. From a total of 11 independent IS data sets, 154 characteristic

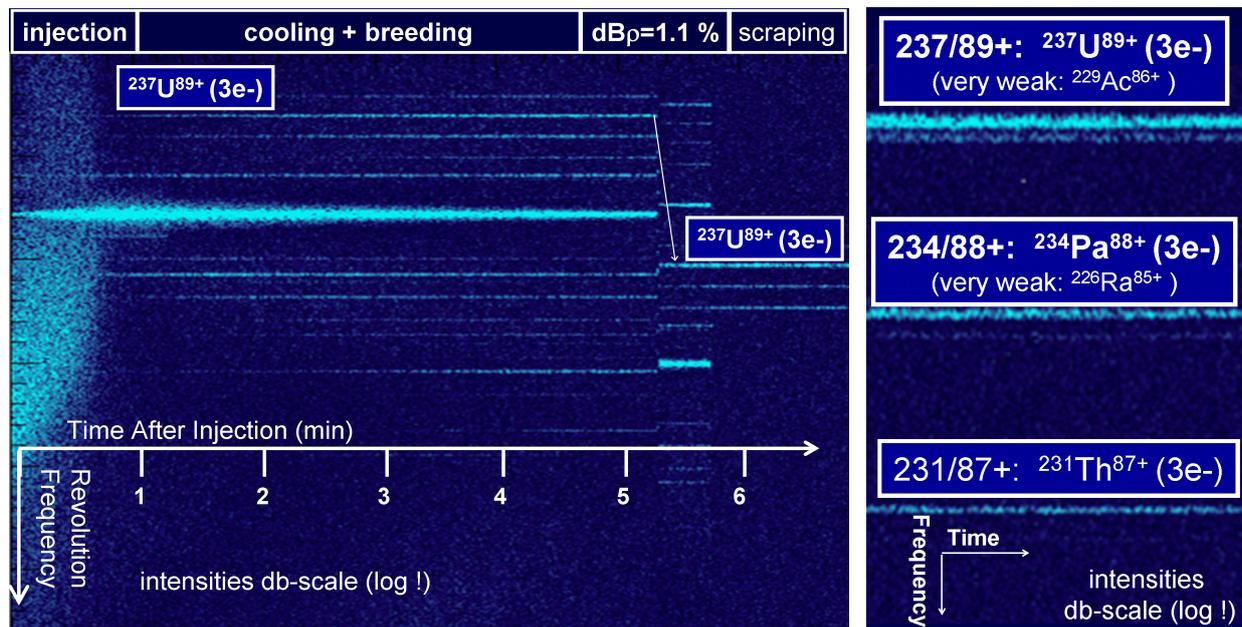


**Figure 1.** (Left panel) DR spectra of the two Li-like isotopes  ${}^A\text{Nd}^{57+}$  with  $A=142$  (grey line) and  $A=150$  (black) in the energy range of the  ${}^A\text{Nd}^{56+}(1s^2 2p_{1/2} 18l_{j'})$  resonance group. [15]. (Right panel) First and second derivative of the spectrum. The zeroes of the derivatives provide norm-independent characteristic values like maxima, minima and inflection points for the determination of the isotope shift. Additional data up to 42 eV (not shown here) was obtained as well. For further details and spectra, cf. [15].

values for the  $2s - 2p_{1/2}$  IS and 45 for the  $2s - 2p_{3/2}$  IS were obtained yielding energy shifts of  $\delta E^{142,150}(2s - 2p_{1/2}) = 40.2$  (3)(6) meV [(stat)(sys)] and a slightly higher value of  $\delta E^{142,150}(2s - 2p_{3/2}) = 42.3$  (12)(20) meV, resp. Although statistically not significant, it is interesting to see that we measured a slightly higher value for the  $2p_{3/2}$  case. This is expected, since relativistically the  $p_{1/2}$  electrons still possess a finite overlap with the nucleus while  $p_{3/2}$  electrons do not. The interpretation of the experimental shifts was performed within a full QED treatment [15, 24], resulting in a change of the charge radius of  ${}^{142,150}\delta\langle r^2 \rangle = -1.36(1)(3)$  fm<sup>2</sup>. It is noteworthy that already in this first experiment energy shifts  $\delta E^{142,150} \approx 40$  meV of atomic transitions of several hundred eV ( $E(2s - 2p_{1/2}) = 139.25$  eV and  $E(2s - 2p_{3/2}) = 729.12$  eV) were determined with an accuracy of  $< 1$  meV and  $< 3$  meV. For very heavy ions—the primary field of application of the DR method—we expect a similar experimental precision on the level of 1 meV. Since nuclear size effects scale with the 5th to the 6th power of the nuclear charge, this leads to a substantial increase of sensitivity with respect to nuclear charge distribution parameters or hyperfine effects.

#### 4. DR pilot experiment with the radioisotopes ${}^{234}\text{Pa}^{88+}$ and ${}^{237}\text{U}^{89+}$

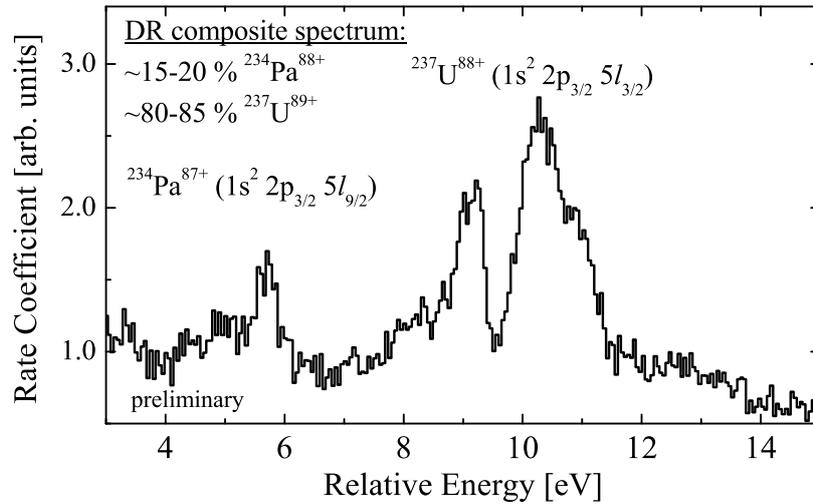
In a two-day test run in May 2008 we could perform DR experiments for the first time with in-flight synthesized radioisotopes, thus proving that the DR technique can be extended to artificially produced unstable isotopes. For our experiment, we developed a new production and separation scheme for the radioisotopes. The exotic nuclides were produced from a  ${}^{238}\text{U}$  primary beam with 381.5 MeV/u impinging on a 1 cm thick ( $1.85$  g cm<sup>-2</sup>) Be-plate located in the “stripping” area of the transfer beam line between the synchrotron SIS and the ESR. The separation of the isotopes was performed entirely in the storage ring ESR. In doing so we made use of the well-known potential of the ESR as a high resolution device for mass measurements and for isobaric or even isomeric separation [27–29]. One decisively benefits from the superb quality



**Figure 2.** Schottky noise power spectra of the preparation of a 3-electron radioisotope beam in the storage ring ESR (left panel). The parameters are optimized for a maximum yield of  $^{237}U^{89+}$ . The close-up (right panel) shows the two Li-like radioisotopes  $^{237}U^{89+}$  and  $^{234}Pa^{88+}$  with high intensity. In close vicinity of these lines, i.e., with an almost identical mass-to-charge ratios, one can see the lines of  $^{229}Ac^{86+}$  and  $^{226}Ra^{85+}$  but with much lower intensity. Well-separated from these doublets is the isotope  $^{231}Th^{87+}$ . Please note that the intensity scale is logarithmic.

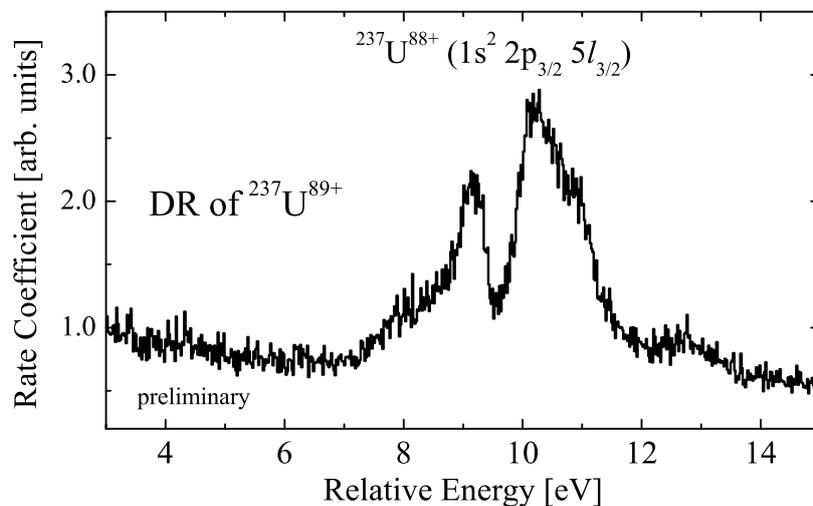
of an electron-cooled ion beam. Using this alternative scenario, we could bypass the fragment separator FRS, a more than 70m high-resolution spectrometer dedicated to the production and separation of exotic radionuclids [30]. This enabled us to run our DR-IS measurements with comparably long-lived radioisotopes *in parallel* to FRS experiments and, thus, make efficient use of the rather precious beamtime at GSI.

The isotopes created in-flight were directly injected into the ESR. Due to energy loss in the thick target (the thickness is required for a large yield of nuclear fragmentation products) the ion energies behind the 1 cm Be-plate are reduced to about 185 MeV/u depending on the isotope. At the same time, large recoil momenta of the nuclear fragmentation reaction as well as energy and angle straggling in the target deteriorate the quality of ion beam leading to a hot secondary beam. The comparatively low energy was chosen in order to facilitate the production of Li-like as well as He-like charge states with high abundance. This offers the opportunity to either directly use the Li-like ions for the experiments or alternatively charge-breed the three-electron isotopes from the He-like via recombination with the cooler electrons. Depending on the time used for cooling the one or the other method yields higher intensities of the wanted species. For the present isotopes  $^{237}U^{89+}$  and  $^{234}Pa^{88+}$  it was anticipated and verified that charge breeding will yield higher final intensities. The injection kicker of the ESR is the main device limiting the transmission of the fragment beam from the target to the ESR. Thus, a pre-separation of the isotopes is given according to the momentum acceptance of the kicker of  $\delta p/p \approx 0.3\%$ . The injected isotope cocktail (blue cloud, left in Fig. 2) is stored in the ESR and cooled to the same average velocity, namely the one of the cooler electrons. For DR experiments, within  $\sim 5$  min, Li-like ions were charge-state bred from the injected He-like ions. The Li-like ions



**Figure 3.** DR spectrum of a composite beam of predominantly  $^{237}\text{U}^{89+}$  (80-85 %) and  $^{234}\text{Pa}^{88+}$  (15-20 %). The corresponding doubly excited configurations are given in the figure. The data were obtained with the settings as shown in Fig. 2. The contamination by the three other isotopes  $^{229}\text{Ac}^{86+}$ ,  $^{226}\text{Ra}^{85+}$  and  $^{231}\text{Th}^{87+}$  is less than one percent. The total beam intensity is about  $1.5 \times 10^5$  stored ions.

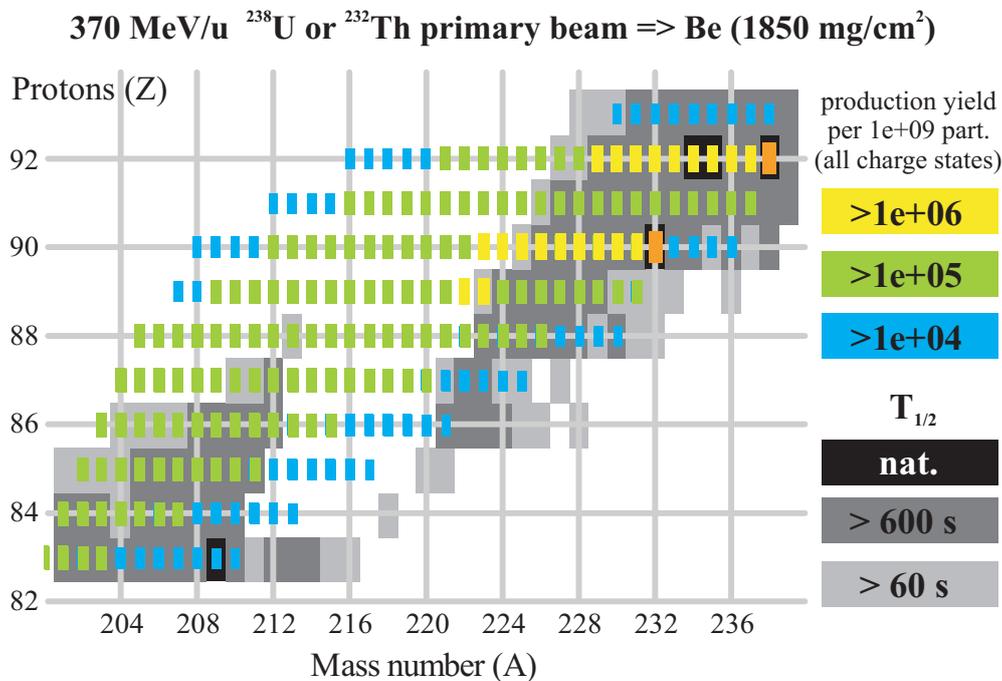
accumulated on outer orbits of the ring were then centered by increasing the magnetic field of the ring magnets according to a momentum change of  $\delta(B\rho) = 1.1$  %. By subsequent insertion of scrapers from both sides into the ring, the momentum acceptance was constricted and thus the desired isotope(s) could be separated (Figure 2). With the initial settings as shown in Fig. 2 we were able to prepare a composite beam consisting predominantly of the isotopes  $^{234}\text{Pa}^{88+}$  and  $^{237}\text{U}^{89+}$  and perform respective DR measurements. Preliminary results are displayed in Fig. 3, where the unique DR fingerprints of the resonance groups  $^{237}\text{U}^{88+} (1s^2 2p_{3/2} 5l_{3/2})^{**}$  and  $^{234}\text{Pa}^{87+} (1s^2 2p_{3/2} 5l_{9/2})^{**}$  can be clearly identified. The possible blends from  $^{229}\text{Ac}^{86+}$  and  $^{226}\text{Ra}^{85+}$ , respectively, are negligible, since their intensities are orders of magnitude lower and



**Figure 4.** DR spectrum of the radioisotope  $^{237}\text{U}^{89+}$ .

they do not possess strong DR resonances in the relevant energy range. Signatures of the approx. 1 % admixture of  $^{231}\text{Th}^{87+}$  could not be identified in the spectrum. By optimizing the yield of  $^{237}\text{U}^{89+}$  shifting the beam by an additional  $\delta(B\rho) = 0.4\%$  and further constricting the aperture we finally were able to measure a clean low-energy DR spectrum of  $^{237}\text{U}^{89+}$  (Fig. 4). In a later setting we also succeeded to measure the DR of  $^{234}\text{Pa}^{88+}$  (not shown here).

Already during this short test run we could additionally evidence that with the given procedure we produce uranium isotopes down to  $^{232}\text{U}^{89+}$  with high intensity. One can further anticipate that virtually all isotopes with sufficient half-lives for electron cooling ( $\sim 1$  min) “south-west” of the primary ions uranium or thorium down to the next stable isotope  $^{209}\text{Bi}$  are produced with sufficient intensity for DR-IS experiments. The displayed isotope production rates (Fig. 5) are given per incident  $10^9$  ions and are calculated for an initial energy of 370 MeV/u and a 1 cm Be-target as in the present test run. No momentum selection was applied. This total yield comprises the contributions from all atomic charge states. For the given parameters of energy and target, the Li-like fraction ranges from about 6 % in Bi ( $Z=83$ ) to 16 % in U ( $Z=92$ ) and for He-like ions from 60 % to 70%, respectively. In the latter case, the charge state breeding technique has to be applied to produce the three-electron ions. In fact, about 5 to 10 % of the total yield given in Fig. 5 will be finally available for the experiment. This number is further increased, since typical primary beam intensities at the synchrotron SIS are 2 to 5 times higher than the ones used for the calculations. It is planned that, later in 2009, we will have a dedicated 8 - 10 day ESR experiment with several in-flight produced uranium radioisotopes in order to study IS and HFS. It is worth mentioning that the preparation of an intense beam of Li-like radioisotopes in the ESR, as demonstrated for the DR study, is also prototypical for other atomic physics experiments with exotic ions, i.e., direct laser excitations between hyperfine levels in heavy few-electron ions [31, 32] or the extraction to the HITRAP facility [33]. For example, laser experiments between the HFS states of heavy few-electron ions that until now are only



**Figure 5.** Calculated production yields of in-flight produced isotopes per  $10^9$  primary beam ions of either 370 MeV/u  $^{238}\text{U}$  or  $^{232}\text{Th}$  impinging on a 1 cm Be target. Half-lives of the produced isotopes  $> 1$  min and  $> 10$  min are indicated as well.

performed with stable ions [31, 32] could be extended to unstable species such as  $^{237}\text{U}^{89+}$ . The accomplished and the proposed experiments will pave the road towards a new generation of DR studies at the future storage ring NESR. The NESR will be the successor of the ESR within the planned “Facility for Antiproton and Ion Research” (FAIR). A dedicated high-resolution electron target at the NESR in combination with orders of magnitude higher yields of radioisotopes and fast pre-cooling of the fragments will lead to a boost in precision and sensitivity. The combination with other in-ring equipment will furthermore expand the scope of DR based applications. For details on the FAIR project cf. [34, 35].

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