

# Measurement of the heaviest $\beta$ -delayed 2-neutron emitter: $^{136}\text{Sb}$

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**Abstract.** The  $\beta$ -delayed neutron emission probability,  $P_n$ , of very exotic nuclei is crucial for the understanding of nuclear structure properties of many isotopes and astrophysical processes such as the rapid neutron capture process ( $r$ -process). In addition  $\beta$ -delayed neutrons are important in a nuclear power reactor operated in a prompt sub-critical, delayed critical condition, as they contribute to the decay heat inducing fission reactions after a shut down. The study of neutron-rich isotopes and the measurement of  $\beta$ -delayed one-neutron emitters ( $\beta 1n$ ) is possible thanks to the Rare Isotope Beam (RIB) facilities, where radioactive beams allow the production of exotic nuclei of interest, which can be studied and analyzed using specific detection systems. This contribution reports two recent measurements of  $\beta$ -delayed neutron emitters which allowed the determination of half-lives and the neutron branching ratio of isotopes in the mass region above  $A = 200$  and  $N > 126$ , and a second experiment which confirmed  $^{136}\text{Sb}$  as the heaviest double neutron emitter ( $\beta 2n$ ) measured so far.

## 1. Introduction

The study of the decay modes of unstable nuclei is an important input to nuclear databases. It provides information needed to develop applications, improve simulations and calculations for theoretical models, and to understand the properties of atomic nuclei experimentally. In the neutron-rich region,  $\beta^-$ -decay is the dominant mode, however  $\beta$ -delayed one-neutron emission, discovered by Roberts et al. [1] in 1939, becomes the dominant decay channel for very neutron-rich isotopes. The importance in measuring the decays of neutron-emitting nuclei in certain regions of the chart of nuclides has been emphasized by international data evaluators experts [2,3] and by theoreticians, who need more realistic inputs for levels above the neutron separation energy,  $S_n$ , for their nuclear structure models, and to reproduce astrophysical environments where the  $r$ -process nucleosynthesis could take place [4–8]. In order to obtain new experimental  $\beta$ -delayed neutron emission probabilities,  $P_n$ -values, a new high efficiency  $4\pi$  neutron detector named BELEN [9,10] has been designed and

developed. Since 2009, BELEN has allowed the study of the  $\beta$ -delayed neutron emission of many species, some of them in regions with very little nuclear data information. This study presents two measurements performed at the GSI Helmholtz Center for Heavy Ion Research using the Fragment Separator (FRS) in Darmstadt (Germany) [11] and at the JYFL Accelerator Laboratory of the University of Jyväskylä (Finland) using the JYFLTRAP Penning Trap [12] at the IGISOL-4 facility. At GSI it was possible to produce the heaviest  $\beta 1n$  emitters measured so far in the region of Hg and Tl with  $N > 126$ , and in Jyväskylä the first  $\beta 2n$  emitter measured above  $A = 100$ ,  $^{136}\text{Sb}$ , was extracted without isobaric contamination. Both experiments provided new  $P_{1n}$ - and  $P_{2n}$ -values for the isotopes studied.

## 2. $\beta$ -delayed one neutron emission beyond $A > 200$ and $N > 126$

Apart from a single measurement of  $^{210}\text{Tl}$  in the sixties [13,14], the available information concerning  $P_{1n}$ -emitters reached only to masses below  $A = 150$  [15–17]. In this nuclear data conference we presented the results of a measurement in which we obtained,

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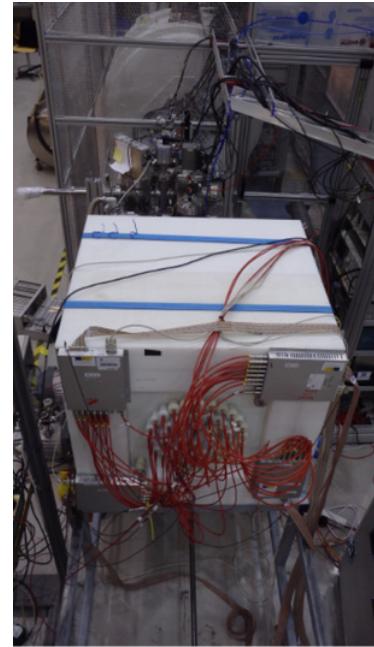
for the first time, the  $P_{1n}$ -values in the region beyond  $A = 200$  and with  $N > 126$  for several isotopes [18]. These included the half-lives of 18 isotopes ( $^{204-206}\text{Au}$ ,  $^{208-211}\text{Hg}$ ,  $^{211-216}\text{Tl}$ ,  $^{215-218}\text{Pb}$ ,  $^{218-220}\text{Bi}$ ), nine of them for the first time, and the neutron branching ratios for those which are energetically allowed neutron emitters ( $^{210-211}\text{Hg}$ ,  $^{211-216}\text{Tl}$ ).

This experiment was performed in 2011 using the accelerator complex of GSI and the FRS facility with similar settings used previously in experiments around this region [19, 20]. Nuclei from Pt ( $Z=78$ ) to Fr ( $Z=87$ ) were identified thanks to a precise time-of-flight measurement and ionization chambers on an event-by-event basis. This region comprises the aforementioned species of interest in the study with positive  $Q_{\beta n}$ -values and candidates for neutron emission [21]. The version of the BELEN detector for this experiment consisted of two rings of  $^3\text{He}$  counters, 10 in the inner and 20 in the outer ring, embedded in a polyethylene matrix. The neutron detector was placed at the end of the FRS beam-line surrounding the Silicon IMplantation Beta Absorber (SIMBA) detector [22, 23]. SIMBA is a stack of stripped silicon detectors based on SSSD and DSSSD technology, and was used to detect the implants and the decays of the nuclei, allowing the correlation analysis between implanted ions,  $\beta$ -decays and neutrons. The results from this measurement are published in Ref. [18], and a more detailed publication has also been submitted.

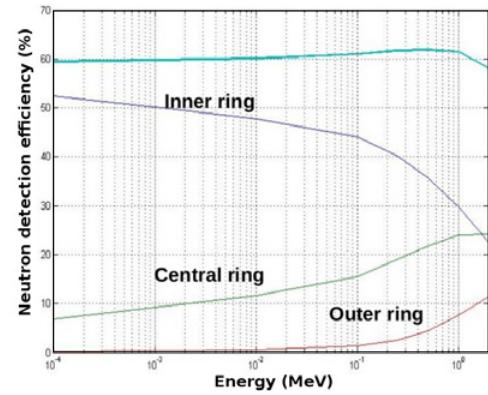
### 3. $\beta 2n$ emission of $^{136}\text{Sb}$

The BELEN detector has also been used several times at the IGISOL facility in Jyväskylä (Finland) between 2009 and 2014 [10, 24]. This facility can produce radioactive species of exotic nuclei by impinging a 25 MeV proton beam on a Uranium target. The reaction fragments are stopped in a buffer gas cell, extracted and accelerated towards a mass separator magnet, which separates nuclei according to the mass-over-charge,  $A/q$ , with a mass resolution of approximately 350. The specific isotope of interest is isolated using the JYFLTRAP Penning Trap system (see top of Fig. 1) [12, 25]. The extraction of the isotopically pure beam, from the JYFLTRAP to the implantation onto a moving tape close to the  $\beta$ -decay plastic detector located at the end of the beam-line, was through a tube in vacuum, which was surrounded by BELEN (see bottom of Fig. 1). In this experiment, the high-efficiency version of the BELEN detector consisted of 48  $^3\text{He}$  proportional counters distributed in 3 rings. We emphasize the high and constant neutron efficiency around  $\varepsilon_{1n} = 60\%$  ( $\varepsilon_{2n} = 36\%$  for  $\beta 2n$  emission) over the energy range of interest (see Fig. 2). In addition, an innovative self-triggered digital data acquisition system was specifically developed for BELEN [26]. This allows the registration of the neutrons coming after a  $\beta$ -decay, including multiple neutron emission ( $\beta 2n$ ) events.

In this conference contribution we present a measurement of  $^{136}\text{Sb}$  with the aim of determining its double neutron emission probability ( $P_{2n}$ -value). Apart of this isotope, in the latest experimental campaign at the IGISOL facility, many of the high-priority  $P_{1n}$ -values from the IAEA list [3] were remeasured with high statistics [24]. In the particular case of  $^{136}\text{Sb}$ , the motivation for measuring this isotope relates to the fact



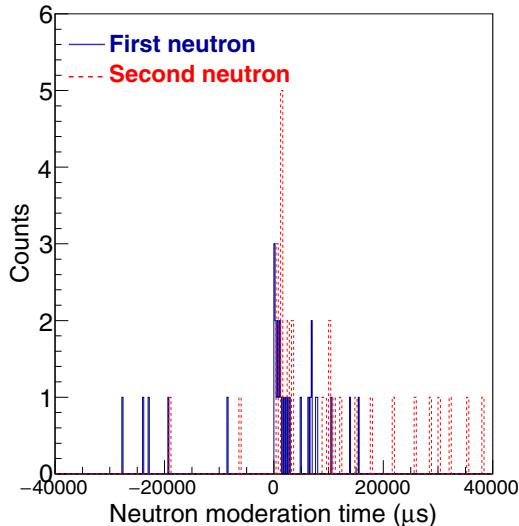
**Figure 1.** Top view of the experimental setup: IGISOL JYFLTRAP at the top of the image. BELEN detector at the bottom.



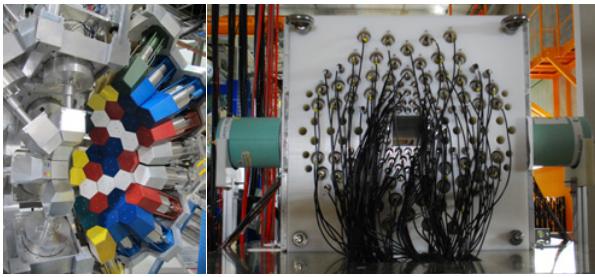
**Figure 2.** Simulation of the neutron efficiency along the energy range for BELEN. Curves represent the efficiency contribution of each ring and the upper one is the total efficiency.

that it is present in the freeze-out of the  $r$ -process and contributes to the second abundance peak around  $N = 82$ , in the  $A \sim 130$  mass region. Furthermore it is present in the fission mass distribution of several actinides. A preceding measurement of the  $A = 136$  isobaric chain suggested this isotope to be a  $\beta 2n$  emitter [27]. Although the contamination from other isobars, the double neutron emission branching ratio was determined to be  $P_{2n} = 1.4 \pm 0.2\%$ , far from the theoretical model predictions available from FRDM+QRPA which gives a branching ratio of  $P_{2n} = 6.2\%$  [28].

In this experimental campaign the  $\beta 1n$  branching ratios for the  $P_{1n}$  standards  $^{95}\text{Rb}$  and  $^{137}\text{I}$  were also measured, and they were used for the calibration of our system. We also confirmed the  $P_{1n}$ -value of  $^{136}\text{Te}$ , which is the  $\beta$ -decay daughter. Since this isotope is present in the decay chain of  $^{136}\text{Sb}$ , an accurate determination of its neutron branching ratio is needed for the data analysis. Our preliminary value for  $^{136}\text{Te}$  is in agreement with the ones previously reported in Refs. [16, 17]. Although the data analysis is ongoing, it



**Figure 3.**  $\beta\text{nn}$  correlation events. In blue is shown the correlation with the first neutron and in red the correlation with the second neutron emitted.



**Figure 4.** Left: left downstream side of the DESCANT detector coupled with the GRIFFIN germanium array. Right: BRIKEN detector setup (July 2016).

can be confirmed that a new  $P_{1n}$ -value will be reported for  $^{136}\text{Sb}$  and that it is also a double neutron emitter. In Fig. 3 the  $\beta\text{2n}$  correlations observed during 6 days of beamtime are shown. A total of 26  $\beta\text{nn}$  net correlated events yield a two neutron branching ratio of  $P_{2n} < 1\%$ . This is a lower value than those mentioned above.

#### 4. Upcoming $\beta\text{n}$ measurements

In the next few years, new measurements of  $\beta$ -delayed neutron emitting isotopes are planned at the RIB facilities of TRIUMF, Vancouver BC (Canada) and RIKEN, Wako-shi, Saitama (Japan). At TRIUMF, the DEUterated Scintillator array for Neutron Tagging (DESCANT) detector [29, 30] is ready to measure  $\beta\text{n}$  events. The system allows one to measure  $\beta\text{n}$  events in correlation with the  $\gamma$ -rays, from the excited states populated in the daughter nuclei, with the GRIFFIN germanium array [31, 32] (see Fig. 4-left). At RIKEN, the BRIKEN campaign [33] is expected to measure the most exotic neutron emitters ever produced, including dozens of  $\beta 1\text{n}$ , several  $\beta 2\text{n}$  and, for the first time,  $\beta 3\text{n}$  emitters in the heavy mass region  $A > 50$ . Currently there are three experiments approved in the regions around  $A = 130$  [34], the doubly-magic  $^{78}\text{Ni}$  [35] and in the region  $A = 100 \sim 125$  [36]. The BRIKEN array is presently under commission and will take first data in the fall of 2016. A picture of the setup is shown in Fig. 4-right.

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#### References

- [1] R.B. Roberts, R.C. Meyer and P. Wang, Phys. Rev. **55**, 510 (1939)
- [2] D. Abriola, B. Singh, and I. Dillmann, “Summary Report of Consultants’ Meeting on ‘Beta-delayed neutron emission evaluation’”, IAEA December 2011, INDC(NDS)-0599; online at <http://www-nds.iaea.org/publications/indc/indc-nds-0599.pdf> (2011)
- [3] I. Dillmann, P. Dimitriou and B. Singh, “Summary Report of 1st Research Coordination Meeting on Development of Reference Database for Beta-delayed Neutron Emission”, INDC(NDS)-0643, IAEA (2014)
- [4] P. Möller, et al., Atomic Data and Nucl. Data Tables **66**, 131–343 (1997)
- [5] I.N. Borzov, Phys. Rev. C **67**, 025802 (2003)
- [6] T. Marketin et al., Phys. Rev. C **93**, 025805 (2016)
- [7] H. Koura et al., Progress of theoretical physics **113**, 305–325 (2005)
- [8] A. Arcones and G. Martínez-Pinedo, Phys. Rev. C **83**, 045809 (2011)
- [9] F. Calviño, A. Torner et al., Universitat Politcnica de Catalunya (2014)
- [10] M.B. Gómez-Hornillos et al., Hyperfine Interactions **223**, 185–194 (2014)
- [11] H. Geissel et al., Nucl. Instr. Meth. B, **70**, 286–297 (1992)
- [12] J. Äystö, Nucl. Phys. A **693**, 477–494 (2001)
- [13] A.V. Kogan and L.I. Rusinov, Soviet Phys. JETP **57**, 365 (1957)
- [14] G. Stetter, Sci. Abstr. **16**, 1409 (1962)
- [15] R.A. Warner, P.L. Reeder, *Delayed Neutron Data from Tristan*, Nuclear Data for Basic and Applied Science, Volume 1. Proceedings of the Intern. Conference on Nuclear Data for Basic and Applied Science, held May 13–17, 1985, in Santa Fe, New Mexico. p. 701 (1986)
- [16] G. Rudstam et al., Atomic Data and Nucl. Data Tables **53**, 1–22 (1993)
- [17] B. Pfeiffer and K-L. Kratz, Progress in Nucl. Energy **41**, 39–69 (2002)
- [18] R. Caballero-Folch, C. Domingo-Pardo et al., Phys. Rev. Lett. **117**, 012501 (2016)
- [19] L. Chen et al., Phys. Lett. B **691**, 234–237 (2010)

- [20] H. Alvarez-Pol et al., Phys. Rev. C **82**, 041602 (2010)
- [21] A. Sonzogni et al., *National Nuclear Data Center (NNDC), Evaluated Nuclear Structure Data*, Brookhaven National Laboratory, USA, <http://www.nndc.bnl.gov/chart/> and [http://www.nndc.bnl.gov/nudat2/indx\\_dec.jsp](http://www.nndc.bnl.gov/nudat2/indx_dec.jsp)
- [22] K. Steiger, Diploma thesis, Technische Universität München (2009)
- [23] Ch. Hinke et al., Nature, **486**, 341–345 (2012)
- [24] J. Agramunt et al., Nucl. Instr. Meth. A **807**, 69–78 (2016)
- [25] J. Hakala et al., PRL **109**, 032501 (2012)
- [26] J. Agramunt et al., Nucl. Data Sheets **120**, 74–77 (2014)
- [27] D. Testov et al., ESP-RUS congress (2011). [http://icc.ub.edu/congress/ESP-RUS2011/Talks\\_Presentations/Parallel\\_Sessions/Seminar\\_S214/09-11-11/Testov.pdf](http://icc.ub.edu/congress/ESP-RUS2011/Talks_Presentations/Parallel_Sessions/Seminar_S214/09-11-11/Testov.pdf)
- [28] P. Möller, B. Pfeiffer and K-L. Kratz, Phys. Rev. C, **67**, 055802 (2003)
- [29] P. Garrett, Hyperfine Interactions, **225**, 137–141 (2014)
- [30] V. Bildstein et al., EPJ Web of Conf., **93**, 07005 (2015)
- [31] C.E. Svensson and A.B. Garnsworthy, ISAC and ARIEL: The TRIUMF Radioactive Beam Facilities and the Scientific Program. 127–132 (2013)
- [32] R. Dunlop et al., PRC **93**, 062801 (2016)
- [33] BRIKEN collaboration: <https://www.wiki.ed.ac.uk/display/BRIKEN>

**BRIKEN approved experiments:**

- [34] NP1406-RIBF128: <http://www.nishina.riken.jp/RIBF/NP-PAC/14thPAC.html>
- [35] NP1412-RIBF127R1: <http://www.nishina.riken.jp/RIBF/NP-PAC/15thPAC.html>
- [36] NP1412-RIBF127R1: <http://www.nishina.riken.jp/RIBF/NP-PAC/16thPAC.html>