β-delayed fission studies at ISOLDE-CERN

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Abstract. This contribution provides an overview of the systematic β-delayed fission (βDF) studies performed by our collaboration at the ISOLDE facility at CERN. Measurements of βDF properties of eight isotopes, including the first identification of this process for six of them, were carried out over the years. Systematic studies of fission fragment mass distributions, which led to the discovery of the new region of asymmetric fission and to identification of multimodal fission in several isotopes, are revisited. Motivation and description of the most recent experiments aimed at ^{178}Au and neutron rich actinium and francium isotopes are introduced. The detection system used until recently as well as the two newly-developed detection systems are described.

1 Introduction

The process of β-delayed fission (βDF) consists of two steps: the precursor nucleus first undergoes a β decay to an excited state close to or above the fission barrier in the daughter nucleus, which then fissions (Fig. 1). The fission occurs with the half-life of the precursor and it competes with γ-ray emission, or if the excitation energy is high enough, also with a particle emission [1]. The available excitation energy of the fissioning nucleus is limited by the Q_β value of the precursor, which is typically less than $\approx$ 10 MeV. Therefore, βDF is an example of the so-called low-energy fission, which is sensitive to microscopic effects of nuclear structure. The βDF process can be used to investigate low-energy fission properties of exotic nuclei, for which other approaches to fission studies would be difficult or impossible [1, 2].

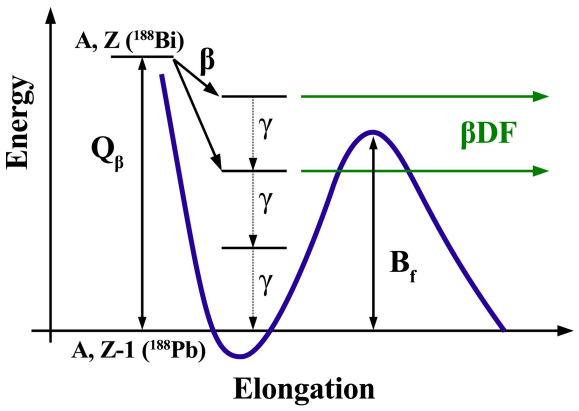


Figure 1. Simple schematics of the βDF process. As an example, β^+/electron capture decay of ^{188}Bi is used. B_f is the fission barrier height in the β-decay daughter ^{188}Pb.

The competition between Q_β values and fission barrier heights is illustrated on the example of β^+/electron capture (EC) decay of bismuth isotopes in Fig. 2. The isotopes, for which βDF was experimentally observed, have Q_β values close to or above the theoretical fission barriers of the daughter lead isotopes. All of these cases are odd-odd isotopes β-decaying to even-even nuclei. This is a general feature of the known βDF cases [1, 2] and it has two main reasons. Firstly, because of pairing, odd-odd isotopes have high Q_β values, which is apparent also in Fig. 2. Secondly, there is a strong hindrance observed for spontaneous fission of nuclei with odd neutron or odd proton compared to even-even isotopes, see for example Ref. [7]. Therefore, a similar strong hindrance is expected also for βDF, when the fissioning β-decay daughter is an odd-A or odd-odd isotope [1, 2].

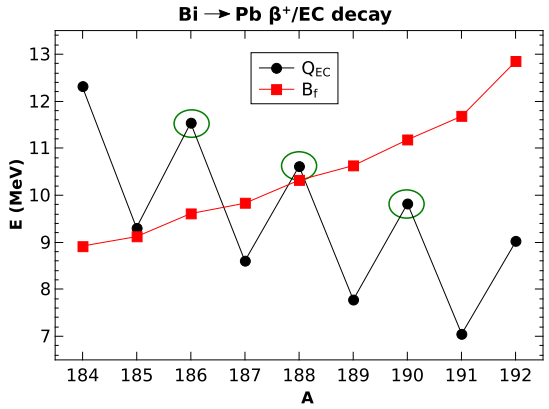


Figure 2. Comparison of Q_{EC} values for neutron-deficient bismuth isotopes and fission barrier heights (B_f) in their β-decay daughters (lead isotopes). Q_{EC} values of isotopes, for which βDF was observed (see Refs. [3, 4]), are marked by green ellipses. Q_{EC} values are taken from AME 2020 [5] and theoretical B_f values are from Ref. [6].

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One of the basic properties of βDF is probability of βDF ($P_{\beta\mathrm{DF}}$), which is experimentally determined as the number of βDF events divided by the number of β decays of the precursor nucleus. Similarly, a partial half-life of βDF ($T_{1/2\mathrm{p},\beta\mathrm{DF}}$) was defined as the half-life of the precursor multiplied by the ratio of the total number of decays of the precursor and the number of βDF events [1]. This quantity is especially useful in the neutron-deficient region, where β-decay branching ratios are often not precisely known, but expected to be small, and so the total number of decays of the precursor can be approximated to the total number of its α decays.

From an experimental point of view, βDF is a rare decay mode. Only around 30 isotopes decaying via βDF were identified so far, often with very low βDF probabilities [1, 2, 4]. However, it is an important process in the nucleosynthesis, since alongside other fission modes, it is responsible for termination of the r-process and for fission recycling, thus having a direct impact on the final abundances of elements in the Universe, see for example Refs. [8–10].

While the extremely neutron-rich regions, where the r-process takes place are experimentally inaccessible, βDF can be studied and used to explore low-energy fission properties in other nuclei, such as for example the isotopes in the neutron-deficient lead region [2]. One of the facilities, where this region can be studied, is ISOLDE [11, 12] at CERN, which often provides intense and isotopically-pure radioactive beams.

This contribution will provide a brief overview of a campaign dedicated to systematic βDF studies performed at ISOLDE by our collaboration, which started more than a decade ago. First, we will revisit the unexpected discovery of an asymmetric fission in the lead region, which was at the beginning of this campaign, and introduce the detection system used for most of the measurements. Afterwards, we will review mapping of the evolution of fission fragment mass distributions (FFMD) in the neutron-deficient region between thallium and actinium. Lastly, we will discuss the most recent experiments with laser-separated β-decaying isomers and briefly describe the newly-developed detection systems suitable for βDF measurements.

2 The new region of asymmetric fission

The campaign to investigate βDF at the ISOLDE facility started with the measurement of ^{180}Tl [13, 14]. This isotope was of a particular interest, because although βDF was reported for it already in the beginning of 1990s [15], they relied on calculated production cross-section for ^{180}Tl to determine $P_{\beta\mathrm{DF}}$. The resulting estimate seemed to be two orders of magnitude lower [13] than what could be expected based on systematics established later for βDF in the uranium region [16].

To be able to reliably determine total kinetic energy released in fission (TKE) and to deduce FFMD, Windmill (WM) detection system was used in the ISOLDE experiment [13, 14]. The vacuum chamber of this system houses a rotating wheel, on which ten thin (20 μg.cm^{-2}) carbon foils are mounted (Fig. 3). The foils are transparent to α particles and fission fragments. At two positions on the circumference of the wheel, silicon detectors are mounted on each side. One of these positions is at implantation point and the detector facing the beam-line is annular to let the beam pass through. This configuration allows a dedicated fission calibration to be made to correct the pulse height defect, which occurs when heavy ion kinetic energies are measured using silicon detectors [14, 17].

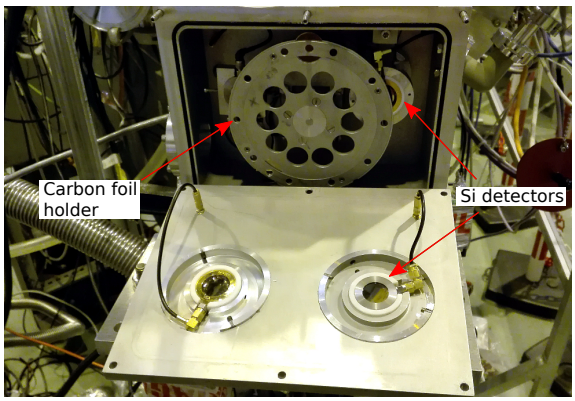


Figure 3. Opened chamber of the Windmill detection system.

The results of the ^{180}Tl experiment [13, 14] showed, that $P_{\beta\mathrm{DF}}(^{180}\mathrm{Tl})$ of $3.2(2)\times10^{-3}$% [14] is roughly in line with the systematics and the previously reported value of $10^{-(5\pm1)}$% [15] was underestimated. Moreover, although the fissioning β-decay daughter ^{180}Hg could be expected to split symmetrically into two semimagic ^{90}Zr nuclei, the measured FFMD was asymmetric with mean masses of 80(1) and 100(1) amu for light and heavy fragments, respectively [13, 14]. Shortly after, βDF was observed for the first time for ^{178}Tl. Despite very low statistics, the resulting FFMD revealed a hint of asymmetric fission also for this case [18].

The surprising discovery of asymmetric fission in βDF of ^{180}Tl brought a lot of interest both from the experimental and the theory side of the physics community. For example, complex calculations of potential energy surfaces (PES) for fissioning systems yielded an interpretation, that somewhat different mechanisms drive fission asymmetry in ^{180}Hg compared to the uranium region: while for ^{236}U the asymmetry is caused by shell effects in the fission fragments, the shell effects in ^{180}Hg itself prevent the fission fragments from entering the valley in the PES for the symmetric mass split [19]. In contrast to this, deformed shell gaps at quadrupole and octupole deformations in fission fragments were discussed as the cause of asymmetric fission for ^{180}Hg, similarly as for actinides [20, 21]. Another interpretation involves the presence of asymmetric saddle points in fission caused by coupling between specific levels, while a similar mechanism is suggested both for actinides and for ^{180}Hg [22]. Therefore, the specific cause of mass asymmetry in βDF of ^{180}Tl remains an open question even after many years since the discovery.

Calculations of FFMDs for large part the Nuclear chart between tungsten and plutonium suggested a whole new

region of asymmetric fission in neutron deficient isotopes below lead [23]. Indeed, for example fusion-fission experiments not only confirmed asymmetric fission of ^{180}Hg even at higher excitation energies of 33–66 MeV, but observed asymmetric FFMDs also for many more isotopes between osmium and lead [24, 25].

3 Mapping of the transitional region between asymmetric and symmetric fission modes

For many neutron-deficient isotopes between radon and thorium, for example for $^{206-208}$Rn and ^{209,210}Ra, symmetric fission was observed in the experiments employing electromagnetically induced fission at GSI Darmstadt [26]. The unexpected occurrence of asymmetric fission in ^{178,180}Hg implied, that a transitional region between asymmetric and symmetric fission modes should lie in the neutron-deficient nuclei between mercury and radon. Therefore, a natural step was to map this region via βDF measurements using the WM system at ISOLDE.

Isotopes ^{194,196}At and ^{200,202}Fr were studied [27, 28], where βDF was observed for the first time for ^{200,202}Fr [27]. For isotopes ^{194,196}At and ^{202}Fr, sufficient statistics was collected to deduce and analyze FFMDs [27, 28]. The broader TKE distributions compared to βDF of ^{180}Tl [13, 14] and a hint of at least three peaks convoluted in the FFMD spectra for ^{194,196}At and ^{202}Fr, suggested a presence of at least two different fission modes for the β-decay daughters ^{194,196}Po and ^{202}Rn [27, 28]. To investigate this in more detail, separate FFMD spectra were constructed for events with TKE higher and for events with TKE lower than the mean TKE value. Indeed, the resulting spectra were significantly different: while FFMD for the events with higher TKE contained distinct central peak, corresponding to a significant contribution of a symmetric fission, the events with lower TKE exhibited flat-topped FFMD spectra, revealing a significant contribution of an asymmetric fission. Therefore, ^{194,196}Po and ^{202}Rn exhibit so-called multimodal fission, in agreement with their location in the transitional region between isotopes with asymmetric and symmetric mass split [27, 28].

These conclusions were supported by the PES calculated for fission of ^{196}Po. The PES indicated that different fission paths corresponding to different symmetric and asymmetric fission modes may compete [27].

4 Laser-separated isomers decaying via βDF

The last βDF measurement performed with the WM system was the experiment dedicated to ^{188}Bi [4]. The β-decay daughter, ^{188}Pb, lies between ^{194}Po and ^{180}Hg, and thus this case is part of the mapping of the transitional region for fission modes. Another strong motivation was to investigate βDF of two β-decaying states in one nucleus independently. So far, presence of two β-decaying isomers was an additional obstacle for βDF studies. The isomers are typically produced and delivered to the detection setup simultaneously, which usually makes it impossible to reliably determine βDF properties of respective states, such as $P_{\beta\mathrm{DF}}$, or even to determined whether both isomers are decaying via βDF. This was the case for example for study of ^{202}Fr at ISOLDE [27] or experiments with ^{192,194}At [29] and ^{186,188}Bi at GSI [3], where $P_{\beta\mathrm{DF}}$ was not determined, or only limits or estimates of $P_{\beta\mathrm{DF}}$ were discussed.

On the other hand, separation of β-decaying isomers opens a possibility to study spin dependence of fission. Although the fissioning state in βDF is in the daughter nucleus, β decay is very selective in respect to spin and the populated states in the daughter will have the same or similar spin as the initial state (isomer) in the precursor. Thus, one could investigate if there are differences in $P_{\beta\mathrm{DF}}$ for the two isomers, or possibly if there are any differences even in FFMDs.

To separate the isomers in ^{188}Bi we employed Resonance Ionization Laser Ion Source (RILIS) [30]. We measured hyperfine spectra for both isomers in ^{188}Bi in our earlier laser-spectroscopy experiment [31], which allowed us to investigate βDF of ^{188}Bi in three different settings: so-called broadband (BB) mode employed broader laser linewidth, covering hyperfine components of both isomers and thus maximizing ionization efficiency at the cost of loss of selectivity. Two so-called narrowband modes employed narrower laser linewidth set to a specific hyperfine component of either the low-spin isomer [NB(^{188}Bi$^{\mathrm{ls}}$)] or the high-spin isomer [NB(^{188}Bi$^{\mathrm{hs}}$)], where only one of the two states was ionized and delivered to the detection system at a time [4].

We observed fission events in both NB modes (12 fission fragments for ^{188}Bi$^{\mathrm{hs}}$, 2 independent fission fragments for ^{188}Bi$^{\mathrm{ls}}$), directly proving that both isomers in ^{188}Bi decay via βDF [4]. Further analysis was done to attribute additional events from BB mode to specific isomer based on different half-lives of the isomers and time structure of the measurement. From the deduced final statistics we determined $T_{1/2\mathrm{p},\beta\mathrm{DF}}$ for each isomer [4].

Since the experimental β-decay branching ratios of these isomers are unknown, to deduce $P_{\beta\mathrm{DF}}$ we had to partially rely on theoretical calculations. Complex calculations of β-decay and βDF properties were performed. Isomer ^{188}Bi$^{\mathrm{hs}}$ was adopted as a specific $I^{\pi} = (10^{-})$ state in literature [32], and thus the properties were calculated for this state. To calculate βDF properties, the fission barrier of ^{188}Pb was scaled to reproduce experimental $T_{1/2\mathrm{p},\beta\mathrm{DF}}$ of ^{188}Bi$^{\mathrm{hs}}$. The character of ^{188}Bi$^{\mathrm{ls}}$ was less clear at the time and we used restrictions based on the preliminary results from our laser-spectroscopy study, pointing that it should be a prolate (1^{+}) state with a positive magnetic moment. Properties were calculated for five lowest-lying states fulfilling these conditions. Four of these states had $T_{1/2\mathrm{p},\beta\mathrm{DF}}$ consistent with the experimental value for ^{188}Bi$^{\mathrm{ls}}$ and their $P_{\beta\mathrm{DF}}$ values between 5.1×10^{-4} and 1.3×10^{-3} were considered as a possible range of $P_{\beta\mathrm{DF}}$ for ^{188}Bi$^{\mathrm{ls}}$ [4]. In the meantime, the final results from the laser-spectroscopy study are available and ^{188}Bi$^{\mathrm{ls}}$ was tentatively proposed to have $\pi 1/2[530] \otimes \nu 1/2[521]$ configuration [31]. This is consistent only with one of the four states considered before and its $P_{\beta\mathrm{DF}}$ was 1.2×10^{-3}. This is by a factor of ≈ 4

Table 1. Summary of βDF measurements performed with the WM system at ISOLDE.

Isotope	Deduced quantities	Reference
^{178}Tl	FFMD, TKE, $P_{\beta\mathrm{DF}}$	[18]
^{180}Tl	FFMD, TKE, $P_{\beta\mathrm{DF}}$	[13, 14]
^{188}Bi	FFMD, TKE, $T_{1/2\mathrm{p},\beta\mathrm{DF}}$, $P_{\beta\mathrm{DF}}$	[4]
^{190}Bi	$T_{1/2\mathrm{p},\beta\mathrm{DF}}$	[4]
^{194}At	FFMD, TKE	[27]
^{196}At	FFMD, TKE, $P_{\beta\mathrm{DF}}$	[27, 28]
^{200}Fr	$P_{\beta\mathrm{DF}}$	[27]
^{202}Fr	FFMD, TKE	[27]

lower than $P_{\beta\mathrm{DF}} = 4.7\times10^{-3}$ for ^{188}Bi$^{\mathrm{hs}}$, hinting at a lower fission probability of low-spin states in the daughter ^{188}Pb nucleus [4]. However, it is desirable to confirm this difference once the experimental β-decay branching ratios become available and fully experimental $P_{\beta\mathrm{DF}}$ values could be deduced.

The collected fission statistics in the NB(^{188}Bi$^{\mathrm{hs}}$) mode was higher than in the NB(^{188}Bi$^{\mathrm{ls}}$) mode, and also its longer half-life made it easier to attribute part of the events from BB mode to ^{188}Bi$^{\mathrm{hs}}$. As a result, we were able to produce FFMD based on the 31 coincident events (62 fission fragments) attributed to βDF of ^{188}Bi$^{\mathrm{hs}}$. Despite the relatively low statistics, the FFMD clearly showed a broad flat-topped character, which could be tentatively interpreted as consisting of at least three peaks: central peak for a symmetric fission mode and outer peaks corresponding to light and heavy fragments from an asymmetric fission mode. Incidentally, the mean values of the peaks corresponding to asymmetric fission were 20 amu apart [4], which is the same value as for βDF of ^{180}Tl [1]. Therefore, also βDF of ^{188}Bi$^{\mathrm{hs}}$ exhibits multimodal fission, similarly to other cases in the transitional region discussed in Sec. 3. During the same experiment, βDF of ^{190}Bi was observed for the first time, albeit based only on one event [4].

All βDF cases studied using the WM system at ISOLDE are summarized in Table 1. As discussed in Sec. 1, for βDF to occur, the Q_β value has to be large enough to allow population of excited states close to the top of the fission barrier (B_f) in the daughter. Therefore, the difference $Q_\beta - B_f$ is an important parameter, and $P_{\beta\mathrm{DF}}$ has an exponential dependence on this parameter, as shown for example in Ref. [33]. Similarly, also $T_{1/2\mathrm{p},\beta\mathrm{DF}}$ has exponential dependence on the $Q_\beta - B_f$ difference, as discussed for example in Refs. [1, 17]. Figure 4 shows such systematics of $T_{1/2\mathrm{p},\beta\mathrm{DF}}$ values for neutron-deficient isotopes, including the latest results for ^{188,190}Bi [4]. Values of $T_{1/2\mathrm{p},\beta\mathrm{DF}}$ deduced from results of measurements performed at ISOLDE are labeled by specific isotopes in Fig. 4. For the parameter $Q_\beta - B_f$, fission barriers from Thomas-Fermi model [34] with microscopic corrections from FRDM [35] were used. Q_β values were taken from AME 2016 [36].

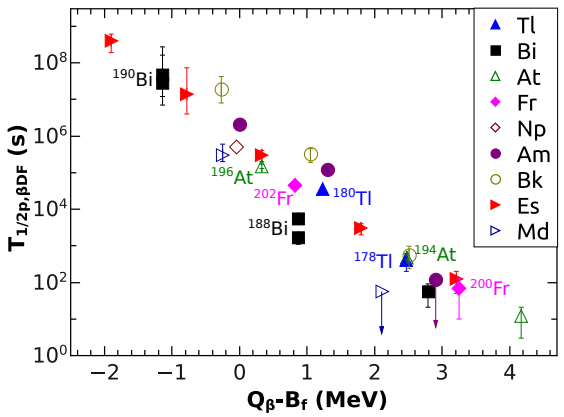


Figure 4. Systematics of experimental partial half-lives of βDF ($T_{1/2\mathrm{p},\beta\mathrm{DF}}$) for the neutron-deficient isotopes as a function of the difference $Q_\beta - B_f$. Values deduced based on results from WM experiments are labeled by specific isotopes. Figure was adapted from Ref. [4].

5 The most recent experiments: laser-separated isomers and a step towards the neutron-rich region

Further step in investigation of βDF at ISOLDE were two experiments, which took place in 2021 and 2022. The former was aimed at the search for βDF in ^{178}Au. This nucleus lies farther in the new region of asymmetric fission [23] discussed in Sec. 2. A competition of symmetric and asymmetric fission was observed for the compound nucleus ^{178}Pt (β-decay daughter of ^{178}At) in complete-fusion experiment already at relatively high-excitation energies of 40–60 MeV [37]. The βDF measurement of ^{178}Au would allow to study fission of ^{178}Pt at lower excitation energies of ≈10 MeV. Moreover, similarly to ^{188}Bi, isotope ^{178}Au also has a high-spin and a low-spin β-decaying states, but their β-decay branching ratios were experimentally determined [38] and their hyperfine components are well separated [38, 39], so broad linewidth (resulting in higher ionization efficiency) of the laser at RILIS can be used to separate these states.

The latter experiment was an attempt to move βDF studies at ISOLDE to the neutron-rich region, where βDF results are scarce and often uncertain [1]. Specifically, the goals were to confirm βDF for ^{230}Ac, for which only two fission events were reported before [40], and to search for this process in ^{232,234}Ac and ^{230,232}Fr. Actinium isotopes of interest can be produced in two ways - directly, with the use of selective ionization by RILIS, but also indirectly via β-decay chain Fr→Ra→Ac, where isotopes ^{230,232}Ra and ^{230,232}Fr are abundantly produced and readily surface-ionized in the hot ion source. The yield of ^{234}Ra will be lower compared to lighter radium isotopes, because it has the same neutron number as the target material ^{238}U, from which these isotopes are produced via spallation reactions. Still, some enhancement of ^{234}Ac production via β decay of ^{234}Ra can be expected.

Both experiments employed two different detection systems. One system was the α-decay spectroscopy setup

(ASET) designed at KU Leuven [41], which is the successor of the WM system. It has a similar configuration of two silicon detectors surrounding a thin carbon foil at the implantation position. Up to nine of these foils are mounted on a movable ladder. The system can include germanium detectors placed outside the vacuum chamber.

The second detection system employed in our measurements was the ISOLDE Decay Station (IDS) [42]. A new box-shaped detector chamber was designed at the University of York to house silicon detectors for α particle and fission fragment detection [43]. In the 2021 experiment, an array of seven silicon PIN diodes was placed in the chamber. During the 2022 run, a single annular silicon detector of the same type as used in the ASET system was mounted in the chamber. In both experiments, the setup was completed by the tape station of IDS and four germanium clover detectors.

The use of two detection systems in parallel was very beneficial, because of their different properties. The IDS can remove implanted longer-lived activities from the chamber thanks to the use of the tape station. In combination with efficient clover detectors it is ideal for yield estimates or identification of possible contamination in the beam, but also for a detailed decay spectroscopy of implanted isotopes. The ASET, on the other hand, has a high geometrical efficiency for α particles of up to 60% [41] and can detect fission fragment coincidences. The synergy of the two systems was even stronger in the second experiment, where for example an implanted sample of long-lived ^{230}Ra ($T_{1/2}$ = 93 min. [44]), decaying to isotope of interest ^{230}Ac, was measured off-line in the ASET, while the on-line ion beam was being delivered to the IDS. The results from these two experiments will be published in dedicated papers after detailed analysis.

6 Conclusion

Systematic studies of β-delayed fission (βDF) performed at the ISOLDE facility by our collaboration started more than a decade ago. So far, βDF properties of eight isotopes of thallium, bismuth, astatine and francium were investigated. For six of them, the βDF decay mode was identified for the first time. Fission fragment mass distributions (FFMD) were measured for six of these cases, where the unexpected observation of asymmetric fission for βDF of ^{180}Tl led to the discovery of the whole new region of asymmetric fission. FFMD measurements for βDF of bismuth, astatine and francium isotopes then served for mapping of the transitional region between asymmetric and symmetric fission in the neutron-deficient isotopes around lead, revealing multimodal fission of their β-decay daughter nuclei. Two states in one isotope decaying via βDF were studied separately for the first time in the case of ^{188}Bi. In the most recent measurements, we searched for βDF of the two states in ^{178}Au, employing isomerically-separated beams, and we also moved the search for βDF into neutron-rich region, specifically to isotopes of actinium and francium.

Acknowledgments

We acknowledge the support of the ISOLDE Collaboration and technical teams throughout the experimental campaign. This contribution was supported by the Slovak Grant Agency VEGA (Contract No. 1/0651/21) and Slovak Research and Development Agency (Contract no. APVV-22-0282).

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