β-delayed fission of ^{186,188}Bi isotopes

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By using the technique of correlating implanted evaporation residues and their subsequent fission decay, β-delayed fission (βDF) of ^{186}Bi has been identified for the first time and βDF of ^{188}Bi has been unambiguously confirmed. The experiments were performed at the velocity filter SHIP (GSI, Darmstadt). The βDF probabilities for both nuclides were qualitatively estimated, and, in particular indications for a large value in the case of ^{186}Bi are regarded.

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I. INTRODUCTION

Recent studies of β-delayed fission (βDF) demonstrated the resurgence of interest in this exotic phenomenon, both from the experimental side (see, e.g., [1–5]) and also from the theoretical side [6–10].

βDF is a rare two-step nuclear decay process, which couples β decay and fission. In the first step of βDF, a parent nuclide undergoes β decay (β^+/EC or β^-), which populates excited states in the daughter nucleus. The maximum excitation energy of the daughter nucleus is limited by the Q_{EC} or Q_β value of the parent nuclide. In the second step, if the excitation energy of these states is comparable to or even higher than the fission barrier height B_f of the daughter nuclide, the latter may undergo prompt fission, in competition with γ-ray and/or particle emission [11,12]. The βDF probability is defined as the ratio of the number of βDF decays, $N_{\beta\mathrm{DF}}$, to the number of β decays, N_β, of the parent nuclide, according to the expression $P_{\beta\mathrm{DF}} = N_{\beta\mathrm{DF}}/N_\beta$. To deduce the $P_{\beta\mathrm{DF}}$ value, knowledge of the β-branching ratio b_β of the parent nuclide is required, especially for nuclei with a small β-decay branch, which is the case of the very neutron-deficient isotopes ^{186,188}Bi discussed in this study.

Two regions of βDF-decaying nuclei are expected to exist which are accessible by presently-available experimental techniques [13,14]: the neutron-deficient nuclei in the actinide and lead regions of the nuclidic chart. While the former region is relatively well studied by now, with twelve cases known (see [4,11,12,15,16] and references therein), rather scarce data existed for the latter group of nuclei prior to our recent βDF studies. These new experiments became possible exclusively due to the systematical application of experimental techniques, which so far have been used only occasionally for βDF investigations. These techniques involve the first use of the Isotope-Separator-On-Line (ISOL) method at the mass separator ISOLDE [17] or the use of a recoil separator, such as, e.g., the velocity filter SHIP [18]. The application of ISOLDE for detailed βDF studies of ^{180}Tl was presented in our recent papers [1,2], while the use of SHIP for the first identification of βDF of ^{192,194}At was given in [3].

In the first βDF studies in the 1990s in the lead region, made at the Flerov Laboratory of Nuclear Reactions (FLNR) in Dubna, three βDF candidates were proposed: ^{180}Tl, ^{188}Bi, and ^{196}At [19,20].

In the experiments at Dubna, the so-called "rotating drum" system was used, in which a heavy-ion beam from a cyclotron irradiated a $\sim$2 mg/cm^2 thick target deposited on the lateral surface of a cooled copper cylinder. This target (serving simultaneously as a recoil catcher) rotated with a constant velocity relative to the mica fission fragment detectors arranged in close geometry around the surface of the cylinder. Measurements of α and β decays were not possible, preventing definite Z and A assignments of the produced activities. Therefore, an extended series of cross-bombardments with different target-projectile combinations and large beam doses had to be performed with the aim of limiting the possible range of A and Z of the observed fission activity and excluding neighboring isotopes produced in different xn, pxn, and αxn channels of the reactions, as well as products from reactions on the target impurities. In particular, βDF of ^{188}Bi nuclei was searched for in the ^{153}Eu(^{40}Ca, $5n$)^{188}Bi, ^{151}Eu(^{40}Ca, $3n$)^{188}Bi, ^{147}Sm(^{45}Sc, $4n$)^{188}Bi, and ^{144}Sm(^{48}Ti, $p3n$)^{188}Bi reactions [20]. Due to the limitations of such a technique, only the fact of fission itself and its half-life could be deduced, with no information on the fission fragments' energy and mass distributions. In total, several hundreds of fission events were

observed in these reactions, and the half-life value of the fissioning activity was deduced as ∼0.3 s. Typical fission cross-section values in these reactions were in the range of 10–100 pb. The conclusion was drawn in [20] that these fission events originate most probably from βDF of ^{188}Bi (the fission of ^{188}Pb produced after β decay of ^{188}Bi).

Two α-decaying states with $T_{1/2} = 210(87)$ ms and $T_{1/2} = 44(3)$ ms were known in ^{188}Bi at that time [21] (see a review of the decay properties of ^{188}Bi in Table I of [22]). As the relative excitation energies of two α-decaying states in ^{188}Bi are still not known, we will denote them as ^{188}Bi^{m1,m2} in this paper, following our study [22]. It is important to note that only one fissioning activity with $T_{1/2} \sim 0.3$ s was mentioned in the Dubna study [20], which could mean that only the longer-lived isomer of ^{188}Bi undergoes βDF according to that study. No information on the βDF probability was reported for ^{188}Bi in [20]. However, by using the fission cross-section from [20] and the measured total production cross-section of ^{188}Bi, after accounting for the *calculated* β-branching ratio, an estimate of $P_{\beta\mathrm{DF}}(^{188}\mathrm{Bi}) = 3.4 \times 10^{-2}\%$ with an uncertainty of a factor of 4 was reported in [23]. No distinction between the two isomers of ^{188}Bi was made in [23] and only one half-life value of 0.21(9) s was mentioned (see Table II of [23]).

Based on the schematic description of the βDF process given above, it is evident that the βDF probability strongly depends on the difference between the Q_{EC} value of the parent isotope and the fission barrier B_f of the daughter product. As no experimentally-deduced fission barriers are known for most of the nuclei in this region, we have to use the calculated values. Figure 1 shows a comparison of the calculated Q_{EC}(Bi) and B_f(Pb) values from the Finite Range Droplet Model + Finite Range Liquid Drop Model (called further FRDM/FRLDM) [14,25] and Thomas-Fermi (TF) [24] mass models. Evaluated or experimental (where available) Q_{EC}(Bi) values for the respective ground states are also provided for comparison [26]. The calculated Q_{EC}(Bi) values from the two mass models are in a good agreement with each other. This reflects the fact that the Q_{EC} values for a given model are derived from the difference between the calculated masses for the parent and daughter nuclei. Therefore, any systematic differences in calculated masses cancel out when Q_{EC} values are calculated. The experimental data are also in good agreement with the theoretical ones.

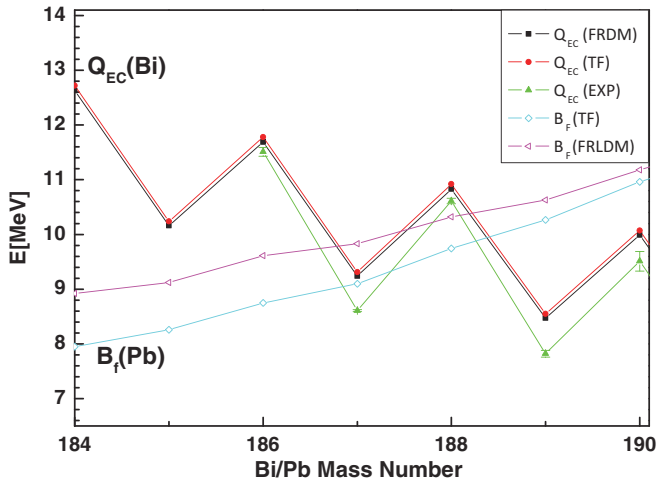

FIG. 1. (Color online) Calculated fission barriers, B_f(Pb) (open symbols), of daughter Pb isotopes and the Q_{EC}(Bi) values (closed symbols) of the β decay of the parent Bi isotopes according to the FRDM/FRLDM [14] and the Thomas-Fermi models [24].

As seen from Fig. 1, the calculated difference $Q_{\mathrm{EC}}(^{188}\mathrm{Bi}) - B_f(^{188}\mathrm{Pb}) = +0.51$ MeV, if one uses FRDM masses [25] and FRLDM fisson barriers [14], which indeed should explain the observation of the βDF in ^{188}Bi. An even higher βDF probability should be expected for ^{186}Bi, for which a larger difference of $Q_{\mathrm{EC}}(^{186}\mathrm{Bi}) - B_f(^{186}\mathrm{Pb}) = +2.08$ MeV can be deduced. Even larger $Q_{\mathrm{EC}} - B_f$ differences are expected within the framework of the TF model, which predicts somewhat lower fission barriers for the relevant lead isotopes, but the main conclusions do not change.

The present work reports on the unambiguous confirmation of βDF in ^{188}Bi and on the first identification of βDF in ^{186}Bi, which were obtained in the experiments at the velocity filter SHIP (GSI, Darmstadt) [18]. The fission data discussed in this paper result from two experiments, dedicated to detailed α-decay spectroscopy of ^{186}Po and ^{188}Bi. The βDF of ^{188}Bi has been studied in the complete fusion reaction ^{142}Nb(^{50}Cr, $p3n$)^{188}Bi, for which the α-decay data were already published in [22]. The isotope ^{186}Bi was studied in the reaction ^{144}Sm(^{46}Ti, $p3n$)^{186}Bi [27], for which, however, the α-decay data are not yet published. As these experiments used the same technique, we refer the reader to [22] for a detailed description of the experimental techniques and the setup. Here we will provide only the most pertinent information relevant for the nuclei studied.

II. EXPERIMENTAL SETUP

Pulsed beams (5 ms "beam on"/15 ms "beam off") of ^{50}Cr and ^{46}Ti were provided by the UNIversal Linear ACcelerator (UNILAC) of GSI. In both experiments, eight targets made of a specific compound (see below) were mounted on a wheel which rotated synchronously with the pulsed structure of the beam, enabling the target to cope with the high-intensity (200–500 pnA) ^{50}Cr and ^{46}Ti beams from UNILAC. In both experiments the data were collected at several beam energies to allow for excitation-function measurements of different evaporation channels [28], which provided confirmation of our conclusions.

Following separation with SHIP [18], the evaporation residues (ERs) were implanted into a 300-μm-thick, 35 × 80 mm^2 16-strip position-sensitive silicon detector (PSSD) [29], where their subsequent α and fission decays were measured. The energy resolution of each strip was typically around 18 keV at α-decay energies of 7–8 MeV. In front of the PSSSD, a system consisting of six silicon detectors, divided in 28 segments (BOX detectors), was installed, allowing the energies of α particles and fission fragments escaping from the PSSD in the backward hemisphere to be measured [29]. Three Time-of-Flight (ToF) detectors [30] in front of the BOX detectors were exploited, allowing the reaction products and scattered beam particles to be distinguished. In addition, the

ToF system allowed decay and implantation events in the PSSD to be discriminated by requiring an anticoincidence condition between the signals from the PSSD and from at least one of the ToF detectors. A clover germanium detector [31], consisting of four crystals, was installed behind the PSSD for prompt and delayed (0–5 μs) particle-γ/x-ray coincidence measurements and also for the registration of singles γ-decay data.

III. EXPERIMENTAL RESULTS

A. βDF of ^{188}Bi

In the case of the ^{142}Nb(^{50}Cr, $p3n$)^{188}Bi reaction, the main data were collected at a beam energy of 252 MeV in front of the target, which corresponds to the measured maximum of the production cross-section of ^{188}Bi (see Fig. 2 of [28]). The beam intensity reached typically up to 500 pnA (where 1 pnA = 6.24×10^9 particles/s). The 290 μg/cm^2 ^{142}Nd targets were produced by evaporating ^{142}NdF$_3$ (99.8% enrichment) onto a 40 μg/cm^2 carbon backing, which was then covered with a 10 μg/cm^2 carbon layer to increase radiative cooling while reducing the sputtering of the material.

Figure 2(a) shows the total energy spectrum of all events registered in the PSSD in the ^{142}Nb(^{50}Cr, $p3n$)^{188}Bi reaction. The spectrum was calibrated by extrapolating to higher energies the calibration based on α decays of ^{188,189,190}Bi nuclei produced in this reaction (see Fig. 1 of [22]). The low-intensity peak at ~230 MeV is due to ^{50}Cr projectiles which passed through SHIP. The observed energy of this peak is lower than the full beam energy because of the energy loss of the beam in the target, in the foils of the ToF system, and in the dead layer of the silicon detector and also due to the pulse-height defect of heavy ions in the detector, which is not accounted for when using the calibration based on α-decay energies. The large peak at ~20 MeV is due to ERs produced in the xn, pxn, and αxn evaporation channels of this

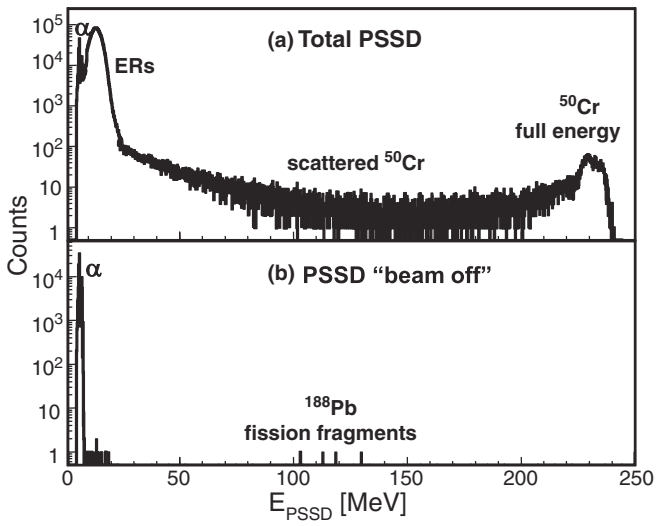

FIG. 2. (a) Energy spectrum measured during the 5-ms "beam on" and 15-ms "beam off" periods in the PSSD for the ^{142}Nb(^{50}Cr, $p3n$)^{188}Bi reaction at a beam energy of 252 MeV. (b) The same as (a), but within the 15-ms "beam off" period only.

reaction, while the events in the 20–230 MeV region are mostly due to scattered ^{50}Cr projectiles and also due to target-like nuclei. Alpha decays of implanted nuclides and their daughter products are seen in the region of <8 MeV.

For reference, the corresponding zoomed-in α-decay spectrum in the energy range of 6.4–7.4 MeV is shown in Fig. 1(b) of Ref. [22]. Complex fine-structure α-decay patterns of two α-decaying states with the tentative spin assignments of (10$^-$) for ^{188}Bi^{m1} and (3$^+$) for ^{188}Bi^{m2} were identified in ^{188}Bi. The improved half-life values for the two states were measured as $T_{1/2}(^{188}\text{Bi}^{m1}) = 265(10)$ ms and $T_{1/2}(^{188}\text{Bi}^{m2}) = 60(3)$ ms [22]. Based on observed α-decay intensities, the ratio of production rates of $\frac{I(^{188}\text{Bi}^{m2},60\text{ ms})}{I(^{188}\text{Bi}^{m1},265\text{ ms})} \sim 1.5$ was deduced. However, no correction for yet unknown and, presumably, rather small (less than 10%; see below) β-branching ratios of the two isomers was implemented for this estimate.

Figure 2(b) shows the same spectrum as Fig. 2(a) but collected only during the "beam off" intervals of 15 ms. Due to this condition, any implantation events due to the beam projectiles, target-like nuclei, and recoiling nuclei are suppressed in this spectrum. This is indeed confirmed by, e.g., the absence of the full-energy ^{50}Cr ion and ER peaks in the spectrum, with the remaining structure below 8 MeV in Fig. 2(b) being due to α decay of the implanted nuclei and their daughter products.

The four high-energy events with an apparent energy of ~100–130 MeV in Fig. 2(b) have been assigned to fission events of ^{188}Pb, being the daughter of ^{188}Bi after β decay. Two of the four fission events are also double-fold fission events, as prompt coincident signals were recorded between the signals in the PSSD and BOX detectors.

There are several arguments to support the assignment of these four events to βDF of ^{188}Bi. First, these events were recorded at the beam energy corresponding to the maximum of the excitation function of ^{188}Bi for this reaction [28]. Second, we used the ER-fission correlation analysis, by searching for time-position correlations between the implantation of an ER and its subsequent fission decay in the same position of the PSSD, within a position window of $\pm$1 mm. By using this method, the time differences Δt(ER $-$ fission) for these four events were deduced as 34.46, 44.60, 168.02, and 284.12 ms. This corresponds to a mean half-life value of 92^{+92}_{-31} ms, which is in agreement with the half-life values of both α-decaying isomers of ^{188}Bi [22].

However, these events cannot be assigned to spontaneous fission (SF) of ^{188}Bi because a partial half-life for SF of the order of $T_{1/2,\text{SF}} \sim 10^{12}$–$10^{14}$ years is expected for ^{188}Bi [32]. Therefore the expected SF branching ratio, defined as the ratio of the total and partial SF half-lives, would be by many orders of magnitude lower than the observed ratio of βDF decays and α decays of ^{188}Bi, $\frac{N_f}{N_\alpha} = 3.2(16) \times 10^{-5}$, after correction for the PSSD detection efficiency.

Such a long SF half-life is both due to the relatively high fission barrier of ^{188}Bi (~9–9.5 MeV (see Fig. 1) and also due to the so-called specialization energy, which results in a fission hindrance of several orders of magnitude for each unpaired nucleon in case of the odd-A fissioning nuclei [33]. In ^{188}Bi there are two unpaired nucleons, so the total fission hindrance is expected to be in the order of ~10^4–10^6. Furthermore, the

observed fission events cannot be due to fission of ^{188}Pb, which is abundantly produced at this beam energy in the $2p, 2n$ evaporation channel, as its half-life is 25.5 s [34] and no fission was observed in earlier studies of this nucleus.

It is important to note that, within the limited number of observed fission events, two of them have Δt(ER − fission) values consistent with the half-life of the shorter-lived 60-ms isomer in ^{188}Bi, while the two others have their time difference values consistent with the half-life of the longer-lived 265-ms isomer. This might indicate that both isomers of ^{188}Bi undergo βDF, but due to the low statistics available no unambiguous conclusion can be drawn here. Conversely, all four events could also be described as proceeding solely either from the short-lived or from the long-lived isomer in this nucleus. As mentioned above, the previous study by Lazarev *et al.* [20] attributed all observed fission events to the longer-lived (∼0.3 s) isomer only. Therefore, this issue is still open and requires further dedicated studies.

On the other hand, it is difficult to expect that only one isomer in ^{188}Bi undergoes βDF, as both of them should have comparable Q_{EC} values. A substantial difference in the β-branching ratio and/or the β-strength function for two isomers with a large spin difference of (10^-) and (3^+) could possibly explain the difference, if it indeed exists, in the βDF properties, but presently none of these quantities are known. Although measured decay data are not sufficient to draw definite conclusions, qualitative estimates will be given in Sec. IV.

Based on the ratio of $\frac{N_f}{N_\alpha} = 3.2(16) \times 10^{-5}$ and by using the production cross-section of ∼9 μb for ^{188}Bi (summed over two isomeric states; see Fig. 2 of [28]), a fission cross-section of 290(150) pb was estimated for βDF of ^{188}Bi in the ^{142}Nb(^{50}Cr, $p3n$)^{188}Bi reaction.

B. βDF of ^{186}Bi

Through the use of the same method as applied to ^{188}Bi, the βDF of ^{186}Bi was identified for the first time in the ^{144}Sm(^{46}Ti, $p3n$)^{186}Bi reaction. The data were collected at a beam energy of 239 MeV in front of the target, corresponding to the maximum production cross-section of ^{186}Bi. The beam intensity typically reached up to 200 pnA. The 400 μg/cm^2 ^{144}Sm targets were produced by evaporating ^{144}SmF$_3$ material (99.47% enrichment) onto a 40 μg/cm^2 carbon backing and then covered with a 10 μg/cm^2 carbon layer.

The energy spectrum for all events registered in the PSSD is shown in Fig. 3(a). The same features are present as in Fig. 2(a), namely, the structures due to the full-energy and scattered ^{46}Ti ions, the ERs peak, and the α-decay events. Any fission events would be hidden under the tail of scattered ^{46}Ti ions in this spectrum.

The α-decay spectrum from this reaction will be published elsewhere, but for the completeness of the discussion we mention that we previously studied α decay of ^{186}Bi by using the complete-fusion reaction ^{93}Nb(^{95}Mo, $2n$)^{186}Bi and the corresponding α-decay spectrum is given in our work [27]. Due to specific settings of the electronics, no fission events could be measured in that run. Here it is sufficient to mention that two α-decaying states, with complex α-decay patterns

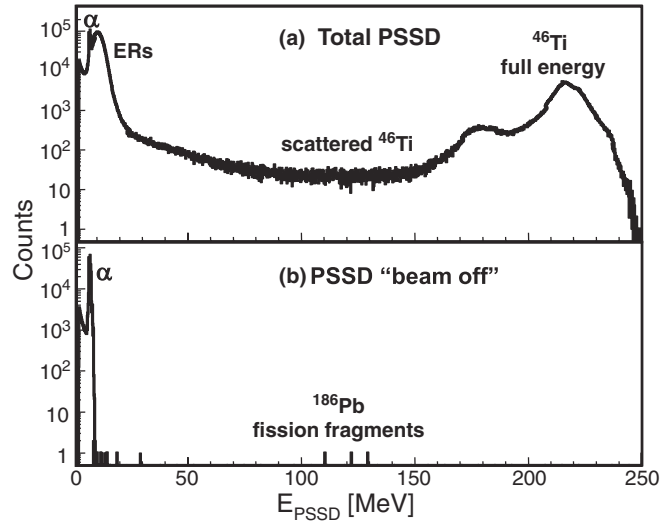

FIG. 3. (a) Energy spectrum measured during the 5-ms "beam on" and 15-ms "beam off" periods in the PSSD for the ^{144}Sm(^{46}Ti, $p3n$)^{186}Bi reaction at 239 MeV. (b) The same as (a), but within the 15-ms "beam off" period only.

and yet unknown relative excitation energies, were firmly established in ^{186}Bi, having quite similar half-life values of $T_{1/2}(^{186}\text{Bi}^{m1}) = 9.8(4)$ ms and $T_{1/2}(^{186}\text{Bi}^{m2}) = 14.8(8)$ ms [27]. Both in the study of Ref. [27] and in the present work, the shorter-lived 9.8-ms ^{186}Bi^{m1} isomer had an α-decay intensity a factor of ∼2–3 larger than the longer-lived isomer. β-branching ratios are not yet known for isomers.

Figure 3(b) shows the same spectrum as in Fig. 3(a), but with the condition of "beam off" only. The three high-energy events in the region of ∼110–130 MeV in this spectrum have been assigned to the fission of the daughter isotope ^{186}Pb following β decay of the parent nucleus ^{186}Bi. Of these three fission fragments, one was a double-fold event in which the coincident signals from the PSSD and from the BOX detectors were recorded. This event and another one were also in coincidence with γ decays registered in the clover detector.

By using the ER-fission correlations, time differences Δt(ER − fission) of 5.99, 10.21, and 30.69 ms were deduced for these three events. Due to half-life considerations, these events cannot be assigned to SF of ^{186}Pb, which is abundantly produced at this beam energy in the $2p2n$ channel, as its half-life of 4.8 s [35] is not consistent with the lifetimes of the recorded events. Based on similar arguments as discussed earlier in the case of the SF of ^{188}Bi, the SF of the ground state of ^{186}Bi can also be excluded. On the other hand, the measured time difference values are consistent with the half-lives of both α-decaying isomers of ^{186}Bi. Similar to the case of ^{188}Bi, the measured Δt(ER − fission) values might indicate that both isomers of ^{186}Bi undergo βDF, but due to the low statistics available no unambiguous conclusion can be drawn here.

The ratio of the number of observed fission and α decays (summed over two isomers) for ^{186}Bi, corrected for the respective PSSD registration efficiency, is $\frac{N_f}{N_\alpha} = 2.2(13) \times 10^{-4}$ at a beam energy of 239 MeV. This value is a factor of ∼6 larger than that for ^{188}Bi. However, to be able to perform a quantitative comparison, one needs to account for the actual

β-branching ratios and the specific population of the different isomers in both ^{186,188}Bi. A qualitative estimate of βDF probabilities will be given in the following section.

Based on α-decay rates, the production cross-section for ^{186}Bi was estimated as ∼220(50) nb, summed over two isomers, leading to the βDF cross-section of 45(25) pb.

IV. DISCISSION: QUALITATIVE ESTIMATES OF βDF PROBABILITIES FOR ^{186,188}Bi

As mentioned earlier, due to the lack of detailed decay schemes and β-decay branching ratios of both isomers in ^{186,188}Bi, their experimental βDF probabilities cannot be deduced from our data. However, in this chapter we will present a discussion resulting in reasonable estimates of the βDF probabilities, which should be correct within an order of magnitude.

A. ^{188}Bi^{m1,m2}

To be able to estimate the $P_{\beta\mathrm{DF}}$ values, we first need to calculate the β-branching ratios for both isomers of ^{188}Bi. By comparing the experimental half-life values for each isomer with the theoretical partial β-decay half-life $T_{1/2,\beta}(^{188}\mathrm{Bi}) = 5.4$ s, calculated within the Quasiparticle-Random-Phase Approximation (QRPA) framework of [36], the following values were obtained: $b_\beta(^{188}\mathrm{Bi}^{m1}, 265\ \mathrm{ms}) \sim 4.9\%$ and $b_\beta(^{188}\mathrm{Bi}^{m2}, 60\ \mathrm{ms}) \sim 1.1\%$. These values are expected to be correct within a factor of ∼2. As an example, we mention that the experimental β-branching ratios of ^{190}Bi^{m1,m2} from [37] are reproduced within a factor of 2 by using this approach. Furthermore, the theoretical partial β-decay half-life for ^{188}Bi, $T_{1/2,\beta}(^{188}\mathrm{Bi}) = 8.5$ s, deduced in [23], agrees quite well with the values of 5.4 s from [36], despite different models being used.

Obviously, due to the small β-branching ratios, the number of detected α decays, corrected for the PSSD detection efficiency, is a good measure of the total number of ^{188}Bi nuclei implanted in the PSSD. Therefore, in the second step, we assumed that only one of the isomers in ^{188}Bi undergoes βDF and compared the observed number of four fission decays to the respective number of α decays from one or another isomer, after accounting for their relative production rates. In this procedure, with the use of the calculated branching ratio of $b_\beta \sim 4.9\%$, an estimate of $P_{\beta\mathrm{DF}}(^{188}\mathrm{Bi}^{m1}) \sim 0.16\%$ could be obtained for the 265-ms isomer. Similarly, by using the calculated branching ratio of $b_\beta \sim 1.1\%$, an estimate of $P_{\beta\mathrm{DF}}(^{188}\mathrm{Bi}^{m2}) \sim 0.48\%$ could be obtained for the 60-ms isomer. The uncertainty of both $P_{\beta\mathrm{DF}}$ values could be estimated as a factor of 4, which includes a factor of 2 due to statistics (only four fission events being observed), and a typical factor of 2 due to the expected uncertainty of the calculated β-branching ratio quoted above. Clearly, if both isomers of ^{188}Bi undergo βDF, the deduced $P_{\beta\mathrm{DF}}$ estimated will become lower, depending on how many out of four observed fission decays would be attributed to each isomer.

Both $P_{\beta\mathrm{DF}}$ estimates are larger than the value of $P_{\beta\mathrm{DF}}(^{188}\mathrm{Bi}) \sim 0.034\%$ deduced in [23], also with an uncertainty of a factor of 4. To deduce their $P_{\beta\mathrm{DF}}$ value, the authors of [23] used a somewhat indirect method and compared the total production cross-section of ^{188}Bi, measured by them [and corrected for the estimated β-branching ratio of $b_\beta(^{188}\mathrm{Bi}) \sim 2.4\%$] with the fission cross-section value of 100(50) pb reported in [20]. Similar to the study in Ref. [20], in [23] only a single 0.21(9)-s isomer in ^{188}Bi is mentioned even though two α-decaying states were already known in ^{188}Bi at that time. The same complete-fusion reaction ^{153}Eu(^{40}Ca, $5n$)^{188}Bi was used in [23] and [20], but experimental techniques were very different.

In conclusion, in the present study (and also in [23]) a number of relatively crude assumptions were made to estimate the βDF probabilities for ^{188}Bi^{m1,m2}. Therefore, the obtained values in the range of ∼0.1%–0.5% with an uncertainty of at least a factor of 4 should be treated only as qualitative estimates.

B. ^{186}Bi^{m1,m2}

An estimate for the βDF probability of ^{186}Bi was performed in a way similar to that for ^{188}Bi. In this case, only three fission events were observed, with the half-life properties compatible with those for both isomers in ^{186}Bi. Therefore, due to the large uncertainty in the relative production of both isomers, we made only a single $P_{\beta\mathrm{DF}}$ estimate for both isomers.

We used a single 'average' experimental half-life value of 12 ms for both isomers in ^{186}Bi. By comparing this value to the theoretical partial β-decay half-life $T_{1/2,\beta}(^{186}\mathrm{Bi}) = 2.1$ s, calculated within the QRPA framework of [36], a single value of $b_\beta(^{186}\mathrm{Bi}^{m1,m2}) \sim 0.6\%$ was derived.

Then, by using the ratio of $\frac{N_f}{N_\alpha}(^{186}\mathrm{Bi}) = 2.2(13) \times 10^{-4}$, and assuming presently an equal population of two isomers in ^{186}Bi, an estimate of $P_{\beta\mathrm{DF}}(^{186}\mathrm{Bi}^{m1,m2}) \sim 7.6\%$ was derived for both isomers, with an uncertainty of a factor of ∼5. In the above estimate we assumed that all three observed fission events originate from one isomer only. Clearly, provided that both isomers indeed undergo βDF, the actual division of fission events between the two isomers and the account of their realistic population ratio will change the above-deduced $P_{\beta\mathrm{DF}}$ estimate. However, this change is expected to be within the quoted uncertainty.

C. Comparison of $P_{\beta\mathrm{DF}}$ values for the uranium and lead region

Even though only order of magnitude estimates could be given for the $P_{\beta\mathrm{DF}}$ values for ^{186,188}Bi, a few tentative conclusions can be drawn from the above discussion. Within the applied assumptions and large uncertainties associated with the estimates, a trend of increasing βDF probabilities can be seen by moving from ^{188}Bi to ^{186}Bi. This is indeed what one expects based on a larger *calculated* difference $Q_{\mathrm{EC}}(^{186}\mathrm{Bi}) - B_f(^{186}\mathrm{Pb}) = +2.08$ MeV in comparison with $Q_{\mathrm{EC}}(^{188}\mathrm{Bi}) - B_f(^{188}\mathrm{Pb}) = +0.51$ MeV, if one uses FRDM masses [25] and FRLDM barriers [14] (see Fig. 1).

Even within these large uncertainties, the estimated $P_{\beta\mathrm{DF}}$ value for ^{186}Bi seems to be consistently higher than the βDF values in the uranium and transuranium regions, for which

typical values in the range of 10^{-4}%–10^{0}% [15] were reported. To our knowledge, only in the βDF of ^{246}Md ($Q_{\mathrm{EC}} - B_f = +0.14$ MeV [14]) has a similarly high value of $P_{\beta\mathrm{DF}} \geqslant 10\%$ been proposed [4].

Finally, similarly large βDF values (as estimated here for ^{186}Bi), in the range of 7%–35%, were derived in our recent βDF study of ^{192}At ($Q_{\mathrm{EC}} - B_f = +2.08$ MeV) [3]. It is interesting to note the practically identical positive $Q_{\mathrm{EC}} - B_f$ differences for ^{186}Bi and ^{192}At. Due to this, in both cases the population of excited states well above the top of the fission barrier can be expected, whereby the fission might be facilitated.

In this respect, the rather high $P_{\beta\mathrm{DF}}$ estimate for ^{246}Md from [4] looks somewhat surprising, as in this case one expects the predominant population of excited states below the top of the fission barrier (so-called sub-barrier fission), which is determined by the strongly energy dependant penetrability through the fission barrier.

V. CONCLUSIONS

In this work we identified for the first time βDF of ^{186}Bi and unambiguously confirmed the occurrence of βDF in ^{188}Bi. An indication for the large βDF probabilities for both nuclei was qualitatively deduced.

Clearly, dedicated follow-up experiments are required to determine relevant nuclear spectroscopic properties, in particular, β-branching ratios for each of the long-lived isomers in these nuclei. These experiments should also determine whether both or only one isomer in each nucleus undergoes βDF. In the case of relatively long-lived ^{188}Bi, this could possibly be performed with the application of highly selective laser ionization techniques, e.g., with the Resonance Ionization Laser Ion Source (RILIS) of the ISOLDE mass separator [17,38]. In this case, by applying isomer-selective ionization with RILIS, one can expect a clean separation of two isomeric states and thus measurements of branching ratios and of the origin of the fission events.

The shorter-lived ^{186}Bi, however, is most probably not yet accessible at such ISOL systems, mostly due to the long extraction time from the ion source. Instead, as demonstrated by this work, this isotope could be studied at recoil separators, especially if the recently developed technique of In Gas Laser Ionization and Spectroscopy (IGLIS) [39] is further applied in such experiments.

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