A Novel Method for the Measurement of Half-Lives and Decay Branching Ratios of Exotic Nuclei

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Abstract. A novel method for simultaneous measurement of masses, Q-values, isomer excitation energies, half-lives and decay branching ratios of exotic nuclei has been demonstrated. The method includes first use of a stopping cell as an ion trap, combining containment of precursors and decay-recoils for variable durations in a cryogenic stopping cell (CSC), and afterwards the identification and counting of them by a multiple-reflection time-of-flight mass spectrometer (MR-TOF-MS). Feasibility has been established by recording the decay and growth of $^{216}$Po and $^{212}$Pb (alpha decay) and of $^{119}$Sb ($t_{1/2} = 850 \pm 90$ ms) and $^{119}$Sb (isomer transition), obtaining half-lives and branching ratios consistent with literature values. Hardly any non-nuclear-decay losses have been observed in the CSC for up to $\sim$10 seconds, which exhibits its extraordinary cleanliness. For $^{119}$Sb, this is the first direct measurement of the ground and second isomeric state masses, resolving the discrepancies in previous excitation energy data. These results pave the way for the measurement of branching ratios of exotic nuclei with multiple decay channels.

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1 Introduction

Half-lives and branching ratios to various decay channels are important properties of unstable exotic nuclei, as they provide vital input to the verification and extension of nuclear structure models throughout the nuclear chart, and to astrophysical nucleosynthesis models [1]. Exotic nuclei, especially far from the stability valley, likely have numerous decay channels, which may include $\alpha$ and $\beta$ decays, spontaneous fission, direct single- or multi-nucleon emission, and $\beta$-delayed fission and nucleon emission. For a review on $\beta$-delayed decays and their importance to nuclear physics and astrophysics, see Ref. [2].

Half-lives and decay branching ratios are mainly measured by the detection of the relevant light particles involved in the decay ($\alpha$, $\beta$, nucleons) as a function of time, and the identification of the recoil nuclei via their $\gamma$ lines, as they are usually created in excited states [3-5]. Penning traps are powerful for purification of precursor beams, whose decay can be researched via several post-trap spectroscopy techniques with minimal background [6].

Other methods use in-trap nuclear decay, and include direct mass measurements of precursors and/or recoils via a Penning trap [7] or a multi-reflection time-of-flight mass-spectrometer (MR-TOF MS) [8]. In another approach, different decay branches are identified and counted by mea-
suring the momentum of the recoil nucleus via time of flight, after decay inside a Paul trap [9]. It has further been proposed to measure decay branching ratios with storage rings [10] and by a series of gas-filled stopping cells and MR-TOF-MSs [11].

In this paper we describe a novel method for measuring half-lives and decay branching ratios of exotic nuclei. We demonstrate for the first time use of a stopping cell as an ion trap. Containment times up to ~10 seconds are shown, indicating the high cleanliness level of this cell. The method is universal, and can be used for nuclides that decay via one or more of any of the channels that are listed above.

We require only very general prior knowledge of nuclear properties, since all are measured during the experiment. This is accentuated in the current article, where while demonstrating our method, we measured directly for the first time the mass of the ground and the second isomeric state \(^{(119mSb)}_{119}\), \(t_{1/2} = 850 \pm 90\) ms of \(^{119}\)Sb, offering a solution to a long-existing conflict regarding its excitation energy and spin assignment [12,13,14,15].

The paper is organised as follows: in Section 2 we describe our novel method for measuring half-lives and branching ratios in the FRS-IC; we present the results of our feasibility study in Section 3; Section 4 discusses ongoing developments of the method, and lists future possible measurements with it. We conclude in Section 5.

2 Outline of methodology

Our method is implemented in the Fragment Separator (FRS) Ion Catcher (IC) [16] at the FRS [17], which comprises a gas-filled cryogenic stopping cell (CSC) [18], an RFQ-based mass filter and beam line [19], and an MR-TOF-MS [20].

The general concept is outlined in Fig. 1. A primary beam is accelerated to relativistic energy by the GSI SIS-18 synchrotron. A pure ensemble of precursors is produced and separated in flight via the FRS. The precursors are contained in the CSC for a controllable duration, while they decay. Due to the high CSC gas density, almost all recoil isotopes are contained. The precursor and recoil isotopes are consequently extracted to the MR-TOF-MS, where they are identified by their mass-to-charge ratio and counted. The precursor decay branching-ratios are determined from the ratios between the number of recoils.

In addition to branching ratios, we simultaneously measure precursor and recoil masses (and thus the precursor Q-values), and the precursor half-life. To reduce systematic errors, half-lives and branching ratios are studied as a function of CSC containment time. Such a method is uniquely-possible with the Ion Catcher at the FRS, because the high energy of the FRS fragments enables the stopping of a clean sample of the ions of interest in the CSC.

2.1 Obtaining clean precursor samples in the CSC

A major requirement of our method is to have a pure sample of precursor ions stopped in the CSC. Our main concern are neighboring nuclides that are either the eventual recoil isotopes of the decay of interest, or nuclides that decay into the precursor or its recoil isotopes. Such effects might mask the investigated precursor and recoils at a level that will render the method inadequate. A visualization of masking for the case of \(\beta\)-delayed neutron emission is given in Fig. 2, indicating which are the nuclides that should not be stopped in the CSC directly from the fragment beam. Similar identification of unwanted contaminant nuclides can be deduced for any other nuclear decay process.

The FRS can provide an isotope beam with the required cleanliness due to the high energy of its fragments. This can be achieved by either using an achromatic mode and closing the slits [17], or using a monochromatic mode and taking advantage of the stopping thickness in the CSC (a few mg/cm\(^2\)), which will stop only the precursor of interest. LISE++ [21] and MOCADI [22] simulations that were performed for several cases indicate maximal masking of investigated precursors and recoils at a level of \(10^{-4}\), setting this value as a lower limit on measurable branching ratios with this method.

Another potential source for contaminants is generated by neutron removal reactions of the incoming beam upstream of the CSC. Such reactions generate neutron-deficient isotopes of the investigated precursor. However, the majority of these contamination isotopes will not be stopped in the CSC. The upper limit of this contribution is estimated to be \(10^{-4}\), for neutron-rich nuclides.

We thus conclude that for \(\beta\)-delayed neutron emission, branching ratios as low as \(10^{-4}\) are detectable by this method. This lower limit will vary for different decay channels, depending on the relative positions of the precursor and recoils in the nuclide chart.

2.2 Decay recoils in the CSC

The ranges of recoil nuclei in relevant decay processes in He buffer gas was studied with the stopping and range of ions in matter Monte Carlo program, SRIM [23], using a typical density of 30 \(\mu\)g/cm\(^2\) (namely 3.17 mg/cm\(^2\) for the 105 cm long CSC, which was the areal density for the measurements with beam-generated isotopes in this work). For \(\gamma\) decay, the recoil kinetic energy is a few tens of eV, corresponding to ranges of a few \(10^{-2}\) mm. In \(\beta\) decay, the recoil kinetic energy is a few hundred eV, resulting in a range of 0.05-0.20 mm. When there is also emission of \(\beta\)-delayed nucleons (or for direct nucleon emission), the recoil kinetic energy rises to tens of keV, increasing the ranges to 0.5-2.0 mm. In \(\alpha\) decay, the recoil energy is in the 100–200 keV range, leading to ranges of about 10-12 mm for typical \(\alpha\) recoil isotopes.

The CSC inner dimensions are 105 cm in length and 25 cm in diameter, so as long as the isotopes of interest are
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Fig. 1. Scheme of the measurement method. A single decay event inside the CSC and the accumulated mass spectrum in the MR-TOF-MS after the extraction of precursors and recoils are depicted. The internal $^{228}\text{Th}$ $\alpha$-recoil source inside the CSC, used for the offline measurements in this work, is also shown.

Fig. 2. Visualization of the masking effect of contamination nuclides on the branching ratio measurement in the case of $\beta$-delayed neutron emission. The green nuclide is the precursor of interest, and the yellow ones are the recoils of interest from the possible branches ($\beta$, $\beta n$, $\beta 2n$), indicated by dashed arrows. Beam contamination of these recoils will obviously interfere with the measurement. In addition, contamination of the red nuclides, which might decay by $\beta$, $\beta n$ and $\beta 2n$ to the recoils of interest (either directly or in a chain), indicated by dotted-dashed arrows, will mask those that result from the precursor of interest. Contamination by white nuclides will not affect the results and can thus be tolerated.

However, for $\beta$ decay and $\beta$-delayed and direct nucleon emission, the recoil range is in the same order of magnitude as the distance from the RF carpet, rendering a systematic effect that is very difficult to estimate. In Section 4.1 we describe a solution to this problem, by containment of the isotopes of interest in the bulk for the vast majority of the containment time.

2.3 Mass measurements and isotope counting

Mass measurements and isotope counting at the FRS-IC are performed with an MR-TOF-MS, which entails a unique combination of performance parameters - fast ($\sim$ ms), accurate (relative mass accuracy of $< 10^{-6}$) and non-scanning [20].

The development and validation of the MR-TOF-MS data-analysis procedure is presented in a separate publication [24]. Mass values and amounts of the measured isotopes and their uncertainties are obtained after fitting the mass peaks to a Hyper-EMG function [25]. The procedure allows accurate mass determination even for the most challenging conditions, including very low numbers of events and overlapping mass distributions. In the mass measurements reported in Ref. [24], a total relative uncertainty down to $6 \cdot 10^{-8}$ was achieved.

2.4 Half-life and branching ratio measurement

Half-lives of exotic isotopes are measured directly in the FRS-IC by varying the isotopes’ containment time in the CSC. The containment time is varied from its minimal extraction time, tens of milliseconds [16] up to $\sim$10 seconds (see Section 3.2), setting the half-life range that can be measured with this method. This is suitable for a very wide range of exotic isotopes and isomeric states.

Decay branching ratios are derived by calculating the ratios between the amount of the recoil isotope resulting from a specific decay channel and the total amount of recoil isotopes from all decay channels.

contained and decay in the bulk, there is no risk to lose recoils on the inner walls. However, in the experiments that are described in this paper, containment of the isotopes took place on the CSC RF carpet close to the exit nozzle (see Fig. 1). As described in detail in [18], the pitch of the RF carpet rings is 0.25 mm, and the isotopes are contained in the buffer gas at about half that distance above the RF carpet.

For $\gamma$-decay there is no effect, since the recoil range is much below $\sim$0.1 mm. On the other hand, for $\alpha$ decay all recoils in the hemisphere towards the RF carpet will be lost, so one merely needs to double the number of recoils counted in the MR-TOF-MS to accommodate for this. These are the decay types that were investigated within the current work.
For the measurements with beam-generated isotopes described herein, the spill structure of the heavy-ion synchrotron SIS-18 is adjusted to allow for short (few millisecond) bunches, separated by long (~10 second) breaks. This separation is chosen to match the longest anticipated storing time inside the CSC. As a result, short spills of ions are injected into the CSC.

Ion storage in the CSC is achieved by increasing the potential on the extraction nozzle to block extraction for an adjustable duration, and lowering it for a few milliseconds to enable extraction. The containment time is defined as the time between the temporal mid-point of the beam-spill and the start of the low-potential pulse at the extraction nozzle.

For isotopes generated by internal radioactive recoil sources, ‘spills’ are created by pulsing the potential of the source. It is normally negative with respect to the surrounding electrodes, thereby preventing ions from leaving the vicinity (~1 mm) of the source, and is pulsed periodically to positive values for a few milliseconds.

After extraction from the CSC, the isotopes are identified and counted by analyzing the MR-TOF-MS spectra via the procedure from [24]. The precursor’s half-life is extracted by fitting the precursor \( (P(t)) \) and recoils \( (R_i(t)) \) counts to the well-known solutions of the Bateman equations [26]:

\[
P(t) = A_p \cdot e^{-\lambda_P \cdot t}
\]

\[
R_i(t) = y_{0i} \cdot e^{-\lambda_{Ri} \cdot t} \cdot \frac{\lambda_P}{\lambda_{Ri} - \lambda_P} \cdot n_{ri} \cdot A_p \cdot (e^{-\lambda_P \cdot t} - e^{-\lambda_{Ri} \cdot t})
\]

where \( A_p \) is the initial amount of the precursor, \( \lambda_P \) is the precursor’s decay constant, related to its half-life by \( \lambda_P = \frac{\ln(2)}{t_{1/2}^P} \), \( \lambda_{Ri} \) is the \( i \)’th recoil decay constant, \( y_{0i} \) is a constant contamination of the \( i \)’th recoil isotope, and \( n_{ri} \) is the \( i \)’th branching ratio. \( A_p \cdot \lambda_{Ri} \) is the amount of the \( i \)’th recoil isotope at infinite time (practically, a time much longer than the precursor half-life). The sum of \( n_{ri} \) is equal to unity.

In the current work the half-lives of the recoils are much longer than the precursor’s, as well as than the containment times, so for our analysis we assume \( \lambda_R \rightarrow 0 \) and use the simplified expression:

\[
R_i(t) = y_{0i} \cdot n_{ri} \cdot A_p \cdot (1 - e^{-\lambda_P \cdot t})
\]

Note that in the investigation of \( \alpha, \beta \) and direct and \( \beta \)-delayed nucleon emission, the FRS will provide an essentially contaminant-free ion sample (Section 2.1). However, in the case of \( \gamma \)-decay of isomeric states, there will always be a non-zero constant contamination, since there is no way to separate isomeric states from ground states in the FRS (Section 2.1). The amount of constant contamination \( (y_{0i}) \) can be obtained directly by a measurement of all recoil candidates with a containment time much shorter than the precursor half-life.

The precursor half-life and decay branching ratios are obtained via a global fit of the precursor and all recoil candidates, thereby providing an improved uncertainty, both statistically and systematically. In particular, the statistics in such a measurement remain constant as a function of containment time, whereas when measuring only the precursor, the statistics decrease as less and less precursors are detected.

By ‘global’ we refer to a unified fit of the data sets of both the precursor and all recoil candidates, using Eq. 1 for the precursor data and Eq. 2 or Eq. 3 for the recoil candidates data. We apply the following constraints to the global fit: the same decay constant parameter \( (\lambda_P) \) for the precursor decay and recoil growth, all branching ratios \( (n_{ri}) \), constant contaminations \( (y_{0i}) \) and the precursor initial amount \( (A_p) \) are non-negative, and the sum of \( A_p \) and all \( y_{0i} \) is unity, as well as the sum of all \( n_{ri} \).

The precursor’s half-life and decay branching ratios can be also extracted from a measurement at only one CSC containment time, provided that the recoil constant contaminations are negligible or are known (and can be subtracted from the counts), and the recoils half-lives are relatively long. The precursor half-life is obtained by division of Eq. 1 by Eq. 3 (setting \( y_{0i} = 0 \)) and subsequent algebraic manipulation:

\[
t_{1/2} = \ln 2 \cdot t_c \cdot \left[ \frac{\ln(1 + D)}{D} \right]^{-1}
\]

where \( D \equiv \frac{P(t_c)}{\Sigma R_i(t_c)} \), and \( t_c \) is the selected CSC containment time. The branching ratio of the \( i \)’th recoil is obtained by

\[
n_{ri} = \frac{R_i(t_c)}{\Sigma R_i(t_c)}
\]

The statistical uncertainty in the measured half-lives and branching ratios depend on the statistical uncertainty of the isotope counts for each containment time, which are evaluated by the analysis procedure [24].

A possible source for systematic uncertainties might be loss of particles due to chemical reactions (e.g., charge exchange, molecular formation) in the CSC buffer gas. The results in this paper (Section 3) indicate that this is a minute effect.

In particular, branching ratio results might be affected systematically by element dependence of these chemical reactions. However, it has been established in previous FRS-IC experiments that the CSC survival and extraction efficiency is essentially element independent, including comparison of a noble element (Rn), and one of the most reactive ones (Th) [27]. In case of doubt or for newly-investigated elements, we will measure the temporal behavior of stable or near-stable isotopes of the element under investigation.

3 Results and discussion

In the following sub-Sectios we present results from both off-line \( (\alpha\text{-recoil source}) \) and on-line (fragment beam) ex-
periments. In all these measurements, the DC pushing field in the CSC was 17 V/cm and the RF carpet voltage was 97 V_p-p at a frequency of 5.92 MHz.

Offline measurements were performed with the $^{228}\text{Th}$ $\alpha$-recoil source that is installed inside the CSC (Fig. 1). During these measurements, the He gas pressure in the CSC was 33 mbar and its temperature was 94 K, corresponding to an areal density of 1.8 mg/cm$^2$. The decay scheme of this source is shown in Fig. 3.

Online measurements were performed with a 300 MeV/u $^{238}\text{U}$ primary beam from the heavy-ion synchrotron SIS-18 with an intensity of up to $2.5 \times 10^8$ ions per spill. A typical spill length of 200 ms was used, which impinged on a beryllium target with an areal density of 0.270 g/cm$^2$ to generate fragments. Due to the low primary beam energy the matter in the FRS beam-line was minimized. The monochromatic degrader at the mid-focal plane of the FRS had an areal density of 737.1 mg/cm$^2$. During these measurements, the He pressure in the CSC was 64.3 mbar and the temperature was 103 K, corresponding to an areal density of 3.17 mg/cm$^2$. A consequence of the low primary beam energy and intensity was that fragment separation was not optimal, generating systematic effects on our data, and signal statistics were generally low, increasing statistical uncertainties.

### 3.1 Mass spectra

In Fig. 4 we show a measured mass spectrum of $^{216}\text{Po}$ and $^{212}\text{Pb}$ ions, generated by the $^{228}\text{Th}$ recoil ion source that is installed inside the CSC. The fit curves in Fig. 4 are the result of the procedure described in [24]. Similar spectra and their analysis were used at various containment times to measure directly the half-life of $^{216}\text{Po}$ and to exhibit the simultaneous decay and growth of the precursor and recoil isotopes in the $\alpha$ decay of $^{216}\text{Po}$ (Section 3.3).

In Fig. 5 we show a measured mass spectrum of $^{119m2}\text{Sb}$, $^{119}\text{Sb}$, $^{119}\text{Te}$ and $^{119}\text{Sn}$, resulting from the fragment beam described above. Note that $^{119}\text{Te}$ and $^{119}\text{Sn}$ both have long-lived isomeric states (200.96 keV, 4.70 days and 89.531 keV, 293.1 days, respectively [28]). Based on [24], the excitation energy of the $^{119}\text{Sn}$ isomeric state is below the FRS-IC resolving power, and although that of $^{119}\text{Te}$ is in principle resolvable, it was not possible to distinguish it with the low statistics and primary beam conditions of this work. Therefore, the mass spectra are fitted to a single peak, and the results are compared to the ground state literature values.

As in Fig. 4 the fit curves are from the procedure described in [24]. One can observe the separate peak fits for the distinguishable overlapping peaks ($^{119m2}\text{Sb}$ and $^{119}\text{Sn}$), which give the mass values and the number of counts for each isotope. The mass of $^{119m2}\text{Sb}$ and the excitation energy of $^{119m2}\text{Sb}$ are measured directly for the first time. Our results and several measurement details are given in Table 1. The measured masses of $^{119m2}\text{Sb}$, $^{119}\text{Sn}$ and $^{119}\text{Te}$ are consistent with the literature values. In Section 5.2 we compare our $^{119m2}\text{Sb}$ excitation energy result to literature and discuss its impact.

### 3.2 Insights from $^{119m2}\text{Sb}$ mass measurement

The second isomeric level of $^{119}\text{Sn}$, namely $^{119m2}\text{Sb}$ ($I_{1/2} = 850 \pm 90$ ms), was first identified in 1979 [12]. Its excitation energy and spin assignment could not be firmly established so far in spite of multiple attempts [12,13,14,15]. This was because of the methodological problems due to correlations and/or multiple $\gamma$ rays present in the spectra at almost the same energy, as well as the existence of another isomeric state at 2553.6 keV. The adopted values for $^{119m2}\text{Sb}$ in the Atlas of Nuclear Isomers [29] and ENSDF [30] are an excitation energy of 2841.7$\pm 0.4 + x$
keV, where x is to be determined, and spin and parity of $27/2^+$. The adopted assignment for the 2841.7 keV level is $I^\pi = 21/2^+$ [30].

Our result for the $^{119m}Sb$ isomeric state excitation energy, 2799±30 keV (Table 1), is the first experimental data that is based on a direct mass measurement, and is 1.4 $\sigma$ below 2841.7±0.4 keV. It suggests that the above-mentioned parameter x is consistent with 0, and the 2841.7 keV level itself is the long-lived isomeric state, as was presented originally in Ref. [13] (fusion reaction experiment) and adopted in Ref. [14] (induced fission experiment).

The experiments of Refs. [13,14] were based on $\gamma$ spectroscopy, and both designated $I^\pi = 25/2^+$ to the 2841.7 keV level, due to the conversion coefficient value of the 288.2 keV transition depopulating the isomer.

We thus conclude that our result confirms the $^{119m}Sb$ isomeric state designation of Refs. [13,14], in contradiction to the adapted assignment [20,30].

### 3.3 Long-term containment in the CSC

We investigated long-term containment in the CSC with the stable and relatively long-living isotopes of the $^{228}$Th decay chain from the CSC internal radioactive $\alpha$-recoil source. Our goal was to demonstrate isotope containment in the CSC without non-nuclear-decay losses (e.g., charge exchange, molecular formation).

We measured the amount of a combination of $^{208}$Pb (stable) and $^{208}$Tl ($t_{1/2} = 3.053$ minutes) as a function of CSC containment time, which we varied from 0.2 seconds up to 9 seconds. The MR-TOF-MS was set to low mass resolving power, so these two isotopes were not resolved. As can be seen from the red data points in Fig. 4 which are fitted to a constant (red solid line), there are no non-nuclear-decay losses for Pb and Tl ions over these long containment times.

$^{220}$Rn ($t_{1/2} = 55.6$ seconds) ions are also detected in the same measurement. As can be seen from the black data points in Fig. 6 which are fitted to an exponential decay curve with the literature time constants. The $^{208}$Pb and $^{208}$Tl counts are consistent with the expected stable behavior and the $^{220}$Rn counts are consistent with the literature value $t_{1/2} = 55.6$ seconds.

### 3.4 Half-life and branching ratio measurement

We demonstrated simultaneous precursor decay and recoil growth via the decay of $^{210}$Po (($t_{1/2} = 145\pm2$ ms) to $^{212}$Pb ($t_{1/2} = 10.64$ hours), both part of the decay chain of the internal $^{228}$Th radioactive recoil source in the CSC.

During data analysis, we realized that in the experiment there was an error in the CSC RF carpet settings, which resulted in unusually long transport of the isotopes along the RF carpet, ~170 ms instead of a few ms. This meant that in these measurements, all isotopes were contained within ~ 0.1 mm from the RF carpet for an unknown duration ranging between 0 and ~170 ms (namely, ~85±85 ms), after the 70 ms drift from the $^{228}$Th source situated at the upstream edge of the CSC. We were able to correct the data for that error, albeit inducing a large systematic uncertainty on the number of $^{210}$Po counts, especially for containment times below ~200 ms. A dead-time correction was also applied to the data, further increasing the systematic uncertainty.

In the error correction procedure we used the literature half-life of $^{210}$Po, in order to account for possible decays...
during the containment of the isotopes near the RF carpet. This was done only for the counts with containment times below \( \sim 200 \text{ ms} \). The effect of including the literature half-life on the final half-life result was small with respect to the main uncertainty component, which is the \( \pm 85 \text{ ms} \) uncertainty in the transport time of the isotopes near the RF carpet.

We plotted the precursor and recoil counts normalized to their sum and performed a global fit on all data points to Eqs. 1 and 3 according to the procedure described in Section 2.4. The relatively long half-life of \(^{212}\text{Pb}\) justified using Eq. 3 instead of Eq. 2. No a-priori assumptions are taken, since we include in the normalization all isotopes whose decay from the investigated precursor have a positive Q-value. For \(^{216}\text{Po}\), only \( \alpha \) decay to \(^{212}\text{Pb}\) meets this criterion.

In Fig. 7, we depict \( \alpha \) decay of \(^{216}\text{Po}\) to \(^{212}\text{Pb}\) in the self-normalizing presentation described above. The effect of the CSC setup error discussed above is visible in the increased uncertainties of both the \(^{216}\text{Po}\) and \(^{212}\text{Pb}\) counts, especially at containment times below \( \sim 200 \text{ ms} \), as expected. The \(^{212}\text{Pb}\) counts were further corrected for the 50\% loss at the RF carpet vicinity due to their long range during the containment of the isotopes near the RF carpet.

### Table 1
| Nuclide    | Half-life / ms  | \( E_{\text{exc, FRS-IC}} \) / keV | \( E_{\text{exc, AME16}} \) / keV | \( E_{\text{exc, FRS-IC}} - E_{\text{exc, AME16}} \) / keV | Number of events |
|------------|-----------------|------------------------------------|------------------------------------|-------------------------------------------------|-----------------|
| \(^{119}\text{Sb}\) | 38.19 ± 0.22 | -89482 ± 17 | -89474 ± 8 | -8 ± 19 | 857 |
| \(^{119m2}\text{Sb}\) | 850 ± 90    | 2799 ± 30 | 2841.7 ± 0.4 + x | -43 ± 30 | 467 |
| \(^{119}\text{Sn}\) | 776 ± 181 ms | -90100 ± 36 | -90065.0 ± 0.7 | -35 ± 36 | 237 ** |
| \(^{119}\text{Te}\) | Stable      | -87310 ± 177 | -87181 ± 8 | -129 ± 177 | 25** |

Table 1. Results of direct mass measurements performed at the FRS-IC. The uncertainties shown correspond to the total experimental uncertainty. Literature values are from [28][29][30][31]. In the MR-TOF-MS, the total time-of-flight was 18.9 ms, and the number of turns was 560. The reference ion in all these measurements was \(^{12}\text{C}\) \(^{18}\text{F}\) (A=119). * Literature and measured value given. ** Total number of events for the unresolved ground and isomeric states. The literature values of \(^{119}\text{Sn}\) and \(^{119}\text{Te}\) are for their ground states.

![Fig. 7](image)

**Fig. 7.** \(^{216}\text{Po}\) and \(^{212}\text{Pb}\) counts, normalized to their sum, as a function of CSC containment time, up to about 1 second. The curves represent a global fit to both data sets to Eqs. 1 and 3.

### Table 2
| \(^{216}\text{Po}\) | \( A_r \) | 1.00 ± 0.10 |
|-------------------|----------|-------------|
| \(^{212}\text{Pb}\) | \( t_{1/2} \) / [ms] | 145 ± 11 |
| \(^{212}\text{Pb}\) | \( g_{0-1} \) | 0.00 ± 0.10 |

Table 2. \(^{216}\text{Po}\) and \(^{212}\text{Pb}\) global fit results

We further used our method to measure the half-life and decay branching ratios of \(^{119m2}\text{Sb}\), obtained from the fragment beam. For \(^{119m2}\text{Sb}\) it is energetically possible to decay to \(^{119}\text{Sn}\) (\( \beta^+ \) decay) and \(^{119}\text{Te}\) (\( \beta^- \) decay), in addition to the \( \gamma \)-decay to \(^{119}\text{Sb}\). We plotted the normalized counts of the precursor \(^{119m2}\text{Sb}\), and its energetically possible decay products. Such a presentation is independent of absolute normalization of the incoming fragment beam from the FRS. Because of the limited primary beam energy, we obtained data with limited statistics and significant beam-induced contamination, including \(^{119}\text{Sn}\) and \(^{119}\text{Te}\).

The results are shown in Fig. 8. The counts are normalized to the sum of the precursor and all decay recoil...
4 Further developments and future measurements

The method presented in this article is well-suited for the measurement of decay branching ratios of nuclei, which decay by more than one channel.
Snapshots from a simulation via ITSIM [32,33] of the isotope trajectories during the above process are given in Fig. 9. The simulation conditions were: He gas at 100 µg/cm³, 155 mbar, and 80 K, RF carpet voltage of 120 V peak-to-peak at 6 MHz, with a radial DC gradient of 4 V/cm, DC longitudinal field of 10 V/cm in step 1 and -0.5 V/cm in step 2. Ions with mass-to-charge ratio of 219 u/e were used, with a macroscopic collision model, which implies that ion motion through the gas is governed by their mobility value, set to 17.5 cm²/Vs.

The simulation shows that the isotopes are kept at a safe distance from the CSC inner walls for most of their duty cycle. If longer containment times are needed, Step 1 and 2 may be cycled. It is important to emphasize that this scheme is possible only because of the small diffusion and low temperature, which enables a long containment time.

4.2 Half-lives and decay branching ratio measurements

The novel measurement method presented here is a powerful mean to investigate masses, half-lives and decay branching ratios of numerous exotic nuclides, in ground and isomeric states, with half-lives ranging from a few tens of milliseconds to a few seconds. In the future CSC for the Low-Energy Branch of the Super-FRS [34], shorter-lived isotopes will be reachable, since its extraction time will be improved to ~5 ms [35].

Furthermore, branching ratios to spontaneous and β-delayed fission could be investigated in the future CSC. This is because its density will be as high as 200 µg/cm³ [35], enabling almost full fission fragment stopping and containment (the ~100 MeV fission fragments' range will be ~12 cm ). This will enable unique research opportunities in the field of fission, because our MR-TOF-MS will provide isotope yield distributions and isomer yield ratios, not only mass distributions.

As our method relies on nuclide identification by direct mass measurements and not by gamma de-excitation lines, it may be the method of choice for nuclides far away from stability, whose excited energy levels are not well-known, or not known at all. It can also provide independent and complementary data for exotic nuclides whose properties were measured via gamma de-excitation, but their values (or their physical interpretation) is under debate, e.g. the excitation energy of the isomeric state 119m2Sb discussed in this work.

According to the Atlas of Nuclear Isomers [29], there are at least 20 more isomeric states that could be investigated with our method at the FRS-IC at GSI (ranging from Co to Ra isotopes, with half-lives in the above-mentioned region and resolvable excitation energies), whose excitation energy and quantum numbers are still ambiguous. Specific examples include 178Ta with two such isomeric levels at 1467.82±x keV (15±, 58 ms) and 2902.2±x keV (21±, 290 ms), and 188Bi and 199Rn for whom also the branching ratio between α and β⁺ decays is yet unmeasured.

A particularly interesting isomeric state for investigation with our method is the 6490±500 keV isomeric state of 94Ag (21⁺, 400 ms), which attracted substantial attention as the so-far only nuclear state to decay by both one-proton and two-proton direct emission [3], in addition to regular β⁺ decay, and β-delayed one- and two-proton emission. This rich variety of decay channels, and the fact that its direct two-proton direct emission channel is still under scrutiny [28], makes this nuclide especially attractive for investigation with a new independent method. An experiment that will investigate N=Z nuclei (including 94Ag) in the FRS Ion Catcher is foreseen in the near future [36].

β-delayed nucleon emission may be a field where our method could make a significant impact. For very neutron-deficient nuclides, the direct identification of the recoils can serve to differentiate between direct and β-delayed proton emission, especially for cases where the precursor and recoil candidates are not sufficiently well known to rule out one of the channels due to negative Q-value.

β-delayed single- and multi-neutron emission is of particular interest to nuclear structure and r-process nucleosynthesis, and is more challenging experimentally than β-delayed proton emission due to the relative complexity of quantitative neutron detection. There are thus numerous ongoing and near-future campaigns to measure β-delayed single- and multi-neutron emission branching ratios [39, 10, 11, 37], including a planned experiment utilizing our method at GSI [38].

This new paradigm, of measuring nuclear processes by mass measurements of the outgoing nuclides in an Ion Catcher, may be expanded from decays to reactions. Identifying and counting the outgoing nuclides can be used to infer simultaneously total reaction cross sections of all existing channels for particular projectile-target combinations. A first experiment in this direction is planned for investigating multi-nucleon transfer reactions of 238U on several medium- and heavy-ion targets that will be installed in the CSC of the FRS-IC [39].

5 Conclusions

We described a new method to measure simultaneously masses, isomer excitation energies, Q-values, half-lives and branching ratios, which was developed and demonstrated in the FRS-IC at GSI. The high fragment energy in the FRS makes it the only facility where this method can be implemented. The method includes a first use of a stopping cell as an ion trapping device, for controllable duration from a few tens of ms up to ~10 seconds.

We demonstrated the method’s feasibility with isotopes from an internal α-recoil radioactive source in the stopping cell, and from the FRS fragment beam. For both cases, the expected temporal behavior of precursors and recoils was shown, and half-lives and branching ratios that are consistent with the literature were obtained.

The method requires only very general prior knowledge of the investigated isotopes. This was shown nicely in this work, where while demonstrating the method, we...
performed the first direct mass measurement of the ground state and second isomeric state of $^{119}$Sb, whose excitation energy and quantum numbers are still not firmly established. Our result for the $^{119m2}$Sb excitation energy suggests that the isomeric state is the energy level 2841.7 keV, with $I^\pi = 25/2^+$ (based on previous $\gamma$-ray spectroscopy experiments [13,14]), in contradiction to the adopted assignment [29,30]. We also confirmed positively that $\gamma$ decay is the only branch of the depopulation of this isomeric state.

We have developed a dynamic storage procedure that will ensure isotope containment mainly in the CSC bulk, enabling research of all possible decay channels, including spontaneous and $\beta$-delayed fission. This method opens the door for a vast range of decay and reaction processes, in an independent and complementary way with respect to existing measurement methods.

6 Authors contributions

All the authors were involved in the preparation of the manuscript. All the authors have read and approved the final manuscript.

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Fig. 9. ITSIM simulation results for dynamic storage in the CSC. Each panel depicts the CSC downstream section. The RF carpet is marked as a hash. The nozzle diameter is not to scale. The vertical axis ($z$) is from -40 to 0 cm (top), and the horizontal axis ($x$) is from -10 to +10 cm. The nozzle is at center-top, $z = -0.5$ cm. The CSC is 105 cm long ($z$) and 25 cm in diameter ($x$). Left: from a late part of Step 1, to 8 ms into Step 2, when the isotopes are $\sim$1 cm away from the nozzle. Middle: Time extended to 100 ms into Step 2. Right: Isotope trajectories from 100 to 600 ms into Step 2 (bulk containment). Incoming and outgoing trajectories are $x$-separated for clarity. Arrows indicate isotopes general direction.

References

1. C. Iliadis, Nuclear Physics of Stars (Wiley-VCH Verlag GmbH & Co, 2015), ISBN 978-3-527-33648-7
2. M.J.G. Borge, Physica Scripta 2013, 014013 (2013)
3. I. Dillmann, A. Tarifeno-Saldivia, Nuclear Physics News 28, 28 (2018)
4. G. Lorusso, A. Becerril, A. Amthor, T. Baumann, D. Bazin, J.S. Berryman, B.A. Brown, R.H. Cyburt, H.L. Crawford, A. Estrade et al., Phys. Rev. C 86, 014313 (2012)
5. I. Mukha, E. Roeckl, L. Batist, A. Blazhev, J. Döring, H. Grawe, L. Grigorenko, M. Huyse, Z. Janas, R. Kirchner et al., Nature 439, 298 (2006)
6. T. Eronen, A. Kankainen, J. Aystö, Progress in Particle and Nuclear Physics 91, 259 (2016)
7. A. Herlert, S. Van Gorp, D. Beck, K. Blaum, M. Breitfeldt, R.B. Cakirli, S. George, U. Hager, F. Herfurth, A. Kellerbauer et al., The European Physical Journal A 48, 97 (2012)
8. R. Wolf, D. Atanasov, K. Blaum, S. Kreim, D. Lunney, V. Manea, M. Rosenbusch, L. Schweikhard, A. Welker, F. Wienholtz et al., Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 376, 275 (2016), proceedings of the XVIth International Conference on Electromagnetic Isotope Separators and Related Topics (EMIS2015), Grand Rapids, MI, U.S.A., 11-15 May 2015
9. R.M. Yee, N.D. Scielzo, P.F. Bertone, F. Buchinger, S. Caldwell, J.A. Clark, C.M. Deibel, J. Fallis, J.P. Greene, S. Gulick et al., Phys. Rev. Lett. 110, 092501 (2013)
10. A. Evdokimov, I. Dillmann, M. Marta, F. Bosch, A. Dolinski, C. Kozhuharov, Y. Litvinov, F. Nolden, M. Steck, H. Weick et al., Proceedings of Science (NIC XII), 115 (2012)
11. H. Miyatake, M. Wada, X.Y. Watanabe, Y. Hirayama, P. Sehury, M. Ahmed, H. Ishiyama, S.C. Jeong, Y. Kakiguchi, S. Kimura et al., AIP Conference Proceedings 1947, 020018 (2018), https://aip.scitation.org/doi/pdf/10.1063/1.5030822
