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Spectroscopy of Element 115 Decay Chains

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Spectroscopy of Element 115 Decay Chains

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A high-resolution α , x-ray, and γ -ray coincidence spectroscopy experiment was conducted at the GSI Helmholtzzentrum für Schwerionenforschung. Thirty correlated α -decay chains were detected following the fusion-evaporation reaction $^{48}\text{Ca} + ^{243}\text{Am}$. The observations are consistent with previous assignments of similar decay chains to originate from element $Z = 115$. For the first time, precise spectroscopy allows the derivation of excitation schemes of isotopes along the decay chains starting with elements $Z > 112$. Comprehensive Monte Carlo simulations accompany the data analysis. Nuclear structure models provide a first level interpretation.

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Citius, altius, fortius.—The Olympic motto translated to the nuclear landscape calls for the quest of new isotopes at its outskirts, which is particularly true at its upper end: here, the first observation of a new isotope may imply the specifically rewarding first observation of a new chemical element.

During the past decade, a number of correlated α -decay chains, which all terminate by spontaneous fission, have been observed in several experiments using ^{48}Ca -induced fusion-evaporation reactions on actinide targets [1,2]. The collaborations working at the Flerov Laboratory of Nuclear Reactions (FLNR) in Dubna, Russia, interpret these to originate from the production of isotopes with proton numbers $Z = 112$ –118 [3].

In recent years, decay chains consistent with even- Z data from FLNR were independently observed [4–10]. In 2011, the discovery of elements $Z = 114$ (flerovium) and $Z = 116$ (livermorium) was acknowledged [11]. However, assignments of Z and mass numbers A of these superheavy elements rely on indirect techniques, such as measurements of excitation functions, cross bombardments, or α -decay systematics, which all depend largely on nuclear mass models. Additional evidence—eventually direct or independent—for Z and A is being requested for claims on elements $Z = 113$, 115, 117, and 118 [11].

Alpha decay of odd- A or odd-odd nuclei typically proceeds to excited states in the daughter nucleus, because unpaired nucleons tend to remain in the same single-particle orbitals as in the α -decay parent [12]. Observation of electromagnetic decays from these excited states can thus elucidate the low-lying nuclear structure of the daughter [13]. Electromagnetic decay into lower-lying states can also proceed by internal conversion, and following the emission of the conversion electron from the atom, x-ray emission is by far the most likely process in the atomic deexcitation. The energy of characteristic ($K_{\alpha 1}$, $K_{\alpha 2}$, and K_{β}) x rays is one direct measure of Z [14]. These energies are predicted with high accuracy (e.g., Refs. [15–18]) and are consistent with experimental data existing up to Db ($Z = 105$) [19]. The atomic number of rutherfordium ($Z = 104$) was determined by x-ray spectroscopy in the early 1970s [20].

Here we report on the first such nuclear spectroscopy experiment and x-ray fingerprinting attempt along α -decay chains anticipated to start from element $Z = 115$.

The Universal Linear Accelerator at the GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany, provided typically 6×10^{12} $^{48}\text{Ca}^{8+}$ ions per sec (time averaged over its pulsed structure, 5 ms beam on and 15 ms beam off) at energies of 5.400 and 5.462 MeV/ u .

The beam impinged on a rotating target wheel, which comprised four target segments averaged $0.83(1) \text{ mg/cm}^2$ $^{243}\text{Am}_2\text{O}_3$. At Mainz University, the ^{243}Am material from Oak Ridge was electroplated onto $2.20(5) \mu\text{m}$ thick titanium backing foils. These foils faced the beam, which implies midtarget beam energies of 242.1 and 245.0 MeV. Beam integrals of $2.13(12)$ and $3.89(23) \times 10^{18}$ ^{48}Ca ions were collected, respectively.

The target wheel was positioned at the entrance of the recoil separator TASCA [9,21,22], filled with He gas at a pressure of $p_{\text{He}} = 0.8 \text{ mbar}$ [23]. For the major portion of the experiment, TASCA was set to center ions in the focal plane with a magnetic rigidity of $B\rho = 2.21 \text{ Tm}$. These ions entered the multicoincidence spectroscopy setup TASISpec [24]. Based on earlier in-beam calibrations and simulations, the efficiency for element $Z = 115$ fusion-evaporation residues was estimated to $30(3)\%$ [25,26].

TASISpec comprises a 32×32 -strip double-sided silicon strip detector (DSSSD), $6 \times 6 \text{ cm}^2$ in area and 0.52 mm thick. A cubelike detector arrangement is formed by four additional 16×16 -strip DSSSDs of the same size and placed upstream. The 96 preamplified signals from the n -doped sides of the DSSSDs were processed with standard electronics [24], while $70\text{-}\mu\text{s}$ long traces of the preamplified signals of all p -doped sides, which are most relevant for high-resolution spectroscopy, were digitized by 60 MHz, 12-bit sampling ADCs. Five composite germanium detectors were placed closely behind each of the five DSSSDs. Their signals were handled by 100-MHz, 16-bit sampling ADCs. Baseline and flat-top energies and time were recorded, eventually flagged with pileup recognition.

List mode data were trigger generated by a coincident signal from a p -side and n -side strip of the implantation detector, the latter limiting the trigger threshold to some 500 keV energy deposited. Trigger rates were typically 100–120 events per sec. Beam on-off status and irradiated target segment number were recorded. Upon detection of an 8.5–11.0 MeV particle in the implantation DSSSD during beam-off periods, the ^{48}Ca beam was chopped within $<20 \mu\text{s}$ for a period Δt_1 . If within Δt_1 another event occurred in the same detector strip at time t_2 , the beam-off period was extended for a period Δt_2 starting at $t = t_2$. Pairs of $(\Delta t_1, \Delta t_2) = (5, 30 \text{ s}), (20, 40 \text{ s}),$ and $(20, 60 \text{ s})$ were used.

During the offline data analysis, emphasis has been put on spectroscopic precision. To name but a few, this includes individually optimized preamplifier decay constants for the moving-window algorithm of the sampled DSSSD pulses, self-consistent pixel-by-pixel determination of DSSSD dead layer thicknesses based on electron and α source data as well as known α -decay energies and Q_α values of background radiation from transfer reaction products. More details can be found in Ref. [27].

A search for time- and position-correlated recoil- α - and recoil- α -fission sequences was conducted using

- (i) $12.0 < E_{\text{rec}} < 18.0 \text{ MeV}$, beam on;
- (ii) $9.0 < E_{\alpha 1} < 12.0 \text{ MeV}$, $\Delta t_{\text{rec}-\alpha 1} = 5 \text{ s}$, beam off, or $10.0 < E_{\alpha 1} < 12.0 \text{ MeV}$, $\Delta t_{\text{rec}-\alpha 1} = 1 \text{ s}$, beam on;
- (iii) $9.0 < E_{\alpha 2} < 11.0 \text{ MeV}$, $\Delta t_{\alpha 1-\alpha 2} = 20 \text{ s}$, beam off, or $9.5 < E_{\alpha 2} < 11.0 \text{ MeV}$, $\Delta t_{\alpha 1-\alpha 2} = 5 \text{ s}$, beam on;
- (iv) $E_{\text{SF}} > 120 \text{ MeV}$, $\Delta t_{\alpha 1-\text{SF}} = 30 \text{ s}$, beam off.

During beam-off and background measurement periods 64 fission events occurred. Most of these were correlated with one of the thirty α -decay chains found via the search algorithm, or associated with short-lived recoil-fission events of transfer reaction products such as ^{242m}Am .

The thirty identified chains contain five recoil- α -fission and two recoil- α -fission events, which are compatible with decay chains proposed to originate from either $^{289}\text{115}$ or $^{288}\text{115}$ [27,28]. One five- α long chain displayed as chain 23 on the right-hand side of Fig. 1 is clearly compatible with the characteristics of the decay chain attributed to start from the isotope $^{287}\text{115}$ [28,29]. None of these chains had Ge-detector events in prompt coincidence with α decays. The applied two-dimensional prompt coincidence window is shown on the upper left of Fig. 2.

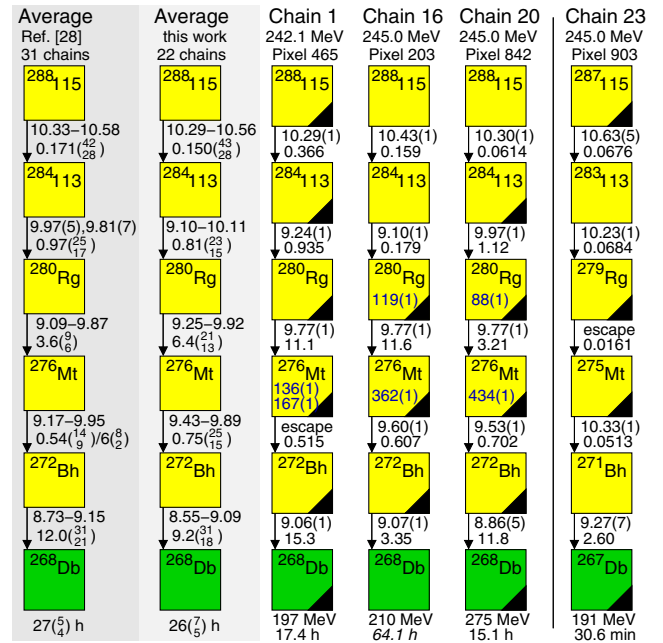


FIG. 1 (color online). Correlated decay chains of $^{287,288}\text{115}$ of the present work compared with Ref. [28]. Individual chains are labeled with an identification number, beam energy at midtarget, and a pixel identifier of the implantation detector. Decay energies are in MeV and times in seconds. Uncertainties of 0.01 MeV for single measurements imply full-energy detection in the implantation DSSSD, otherwise the energy was reconstructed from coincidences in the implantation and one box DSSSD. Cases marked “escape” denote events with low-energy detection solely in the implantation DSSSD. Numbers with uncertainties inside the yellow squares relate to photon energies detected in prompt coincidence with the respective α -decay mother. Black triangles indicate detection during beam-off periods.

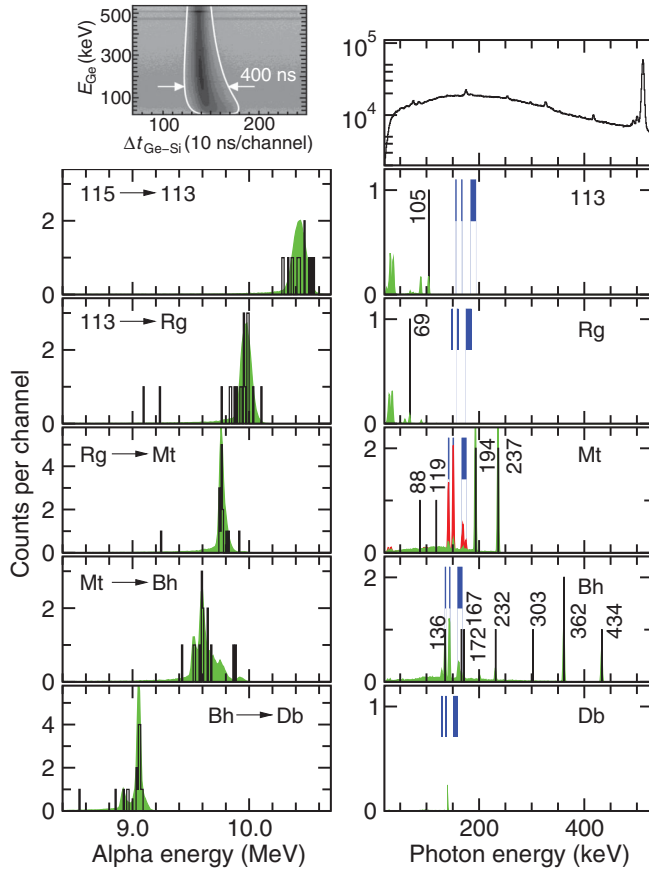


FIG. 2 (color). Top row: correlation time between events in the implantation DSSSD (left) and any of the Ge detectors as a function of Ge-detector energy, and the total projection of Ge-detector events in prompt coincidence with events in the implantation DSSSD (right), both accumulated during beam-off periods. The prompt coincidence window is indicated by the white line in the two-dimensional plot. The remaining five rows show energy spectra of the DSSSD Si cube, and Ge-detector spectra in prompt coincidence with the DSSSD events for each α -decay step starting from $^{288}\text{115}$ (black histograms). The blue vertical bars in the photon spectra indicate predicted $K_{\alpha 2}$, $K_{\alpha 1}$, and K_{β} energies of the corresponding element. Peak labels mark photon energies in keV. The smooth curves (green and red) show scaled Monte-Carlo simulations of 10^5 $^{288}\text{115}$ ions implanted into a virtual detector setup [32] using decay schemes as those in Fig. 3.

The remaining 22 chains are fully or in significant portions compatible with the 31 chains previously assigned to the decay of $^{288}\text{115}$ [28,29]. Figure 1 compares averaged values reported from FLNR [28] with our values, shown together with three selected chains, which are considered most relevant for the construction of decay schemes and x-ray fingerprinting. A table with detailed information on our results is available as Supplemental Material [30]. For a comprehensive assessment, see Ref. [27].

The left column of Fig. 2 provides five energy spectra taken with the DSSSD cube in the energy range of interest.

They comprise both full-energy and reconstructed events (cf. caption Fig. 1) of the respective step in the α -decay chains, starting with $^{288}\text{115}$.

The topmost spectrum of the right column shows the total number of counts in all Ge-detector crystals during beam-off periods ($t_{\text{beam-off}} = 1.10 \times 10^6$ s) taken in prompt coincidence with events at any energy detected above threshold in the implantation DSSSD. It has $N_{\gamma} = 8.0 \times 10^6$ data points in the displayed energy range, dominated by the 511-keV e^+e^- annihilation peak. The remaining five spectra in the right column provide Ge-detector spectra taken in prompt coincidence with the corresponding, firmly established α decays along the chains. These spectra have 1, 1, 6, 8, and 0 counts, respectively. For a given pixel, e.g., No. 465 from chain 1 (cf. Fig. 1), and escape-like energies, $0.5 < E_{\text{Si}} < 2.0$ MeV, the corresponding prompt Ge-detector spectrum has 5620 events, i.e., a coincidence rate of about 1 in 200 s during beam-off periods. Selecting detected full-energy and reconstructed α particles instead, $8.0 < E_{\text{Si}} < 12.0$ MeV, these numbers reduce to four counts, hence 1 such coincidence per 75 h. The expected low random rate is nicely corroborated by the absence of 511-keV events in any of the five spectra of interest. Nevertheless, there is a finite probability of 10%–15% for random α -photon coincidences [27], corresponding to about 2 of the 16 photon events in Fig. 2. Another random photon may arise from an unrelated escape-like event possibly being assigned as a true member to any of the present 22 chains (cf. the Supplemental Material [27,30]).

Only recently, realistic GEANT4 Monte Carlo simulations have been introduced to support conclusions drawn from experimental data on heavy nuclei [31]. To start with, the simulations provide proper implantation profiles of the ions of interest, followed by tracking recoil and decay energies. They account for transition probabilities of nuclear and atomic deexcitations, including emission and detection of α particles, γ rays, conversion electrons, x rays, and Auger electrons. In particular, possible energy summing of interactions of different types of decay radiation within a given active detector element of the setup is handled properly.

Assuming production and decay of $^{288}\text{115}$, the chains of 10^5 ions implanted into a virtual TASI Spec [32] were simulated. The decay schemes of the isotopes along the chain are the physics input (cf. Fig. 3). The output is organized such that it produces list mode data, i.e., energy and time of hits in a given TASI Spec detector element. This data set was sent through the same offline analysis codes as used for the calibrated experimental data. The simulated energy spectra are displayed by smooth curves in the lower five rows of Fig. 2. One common normalization factor is used for all simulated energy spectra, such that the ratio between the number of counts in simulated and measured α -decay spectra is on average 2.

continues to act as a spectator in the decay to ^{272}Bh . Here, the up-sloping neutron orbitals $\nu[606]11/2$ and $\nu[604]9/2$ stemming from the $g_{9/2}$ and $i_{11/2}$ shells, are close to the $\nu[613]5/2$ orbital linked to the $d_{5/2}$ - $g_{7/2}$ shells. Transitions between these orbitals can explain the proposed γ -decay scheme of ^{272}Bh .

In conclusion, thirty correlated α -decay chains were observed following the reaction $^{48}\text{Ca} + ^{243}\text{Am}$. Decay schemes arising from high-resolution spectroscopic coincidence data, in conjunction with comprehensive Monte Carlo simulations, open the door for direct nuclear structure insights of these heaviest man-made atomic nuclei. Previous assignments linking the majority of the decay chains to the decay of $^{287,288}115$ [28,29] are confirmed. This includes first candidates for Z fingerprinting the decay of Mt by means of characteristic K -x ray detection. There is clearly potential for direct determination of the atomic number of the descendants of superheavy elements.

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TABLE I: (Supplemental material) Energies of the implanted recoils E_{rec} , α energies E_{α} measured with the silicon detectors, photon – i.e., γ -ray or X -ray – energies E_{ph} detected in the Ge-detector crystals in prompt coincidence with particle decays, and spontaneous fission energies E_{SF} , together with the associated correlation times of decay chains attributed to the decay of $^{288}\text{115}$ (Nos. 1 to 22) and $^{287}\text{115}$ (No. 23) at the two mid-target laboratory-frame beam energies of 242.1 MeV (Nos. 1 to 8) and 245.0 MeV (Nos. 9 to 23). Entries in bold were recorded during beam-off periods. Entries in italic relate to tentative or unsecure assignments, often in connection to a missing α -decay event in a chain. These entries are thus not part of the spectra displayed in Fig. 1. Uncertainties of individual energy measurements are 10 keV or better at typical α -decay energies of 10 MeV in the implantation DSSSD, worsened for reconstructed events including DSSSD box detectors due to the energy straggling in the deadlayers of the Si detectors. The full width at half maximum (FWHM) for α -decay lines are FWHM = 25 keV at 7.5 MeV and 33 keV at 11.8 MeV for the total DSSSD spectrum. The corresponding numbers are 1 keV and a FWHM = 2 keV for Ge-detector entries in the relevant energy range up to 500 keV.

No.	E_{rec} (MeV)	$^{288}\text{115}$ $E_{\alpha 1}$ (MeV)	$^{284}\text{113}$ $E_{\alpha 2}$ (MeV)	^{280}Rg $E_{\alpha 3}$ (MeV)	^{276}Mt $E_{\alpha 4}$ (MeV)	^{272}Bh $E_{\alpha 5}$ (MeV)	^{268}Db E_{SF} (MeV)
	pixel (x,y)	$\Delta t_{\alpha 1}$ (s) E_{ph} (keV)	$\Delta t_{\alpha 2}$ (s) E_{ph} (keV)	$\Delta t_{\alpha 3}$ (s) E_{ph} (keV)	$\Delta t_{\alpha 4}$ (s) E_{ph} (keV)	$\Delta t_{\alpha 5}$ (s) E_{ph} (keV)	Δt_{SF} (h) γ -ray fold
1	13.4 465 (14,17)	10.29(1) 0.366	9.24(1) 0.935	9.77(1) 11.1	0.825(3)^a 0.515 136(1), 167(1)	9.06(1) 15.3	197^b 17.4
2	13.9 748 (23,12)	10.48(1) 0.177	10.00(1) 1.18	missing and 4.80 136(1)	0.936(3)^a 0.600	9.07(1) 8.23	232 25.5 2
3	16.3 557 (17,13)	missing	9.99(1) 0.240	9.77(1) 18.0 237(1)	9.60(1) 0.600	9.04(1) 2.70	233^b 39.7 4
4	16.1 716 (22,12)	10.37(4)^c 0.212	9.99(1) 1.50	9.5(3)^d 26.4	9.65(1) 8.95	9.05(4)^c 17.8	236 26.6 > 9
5	16.4 331 (10,11)	10.44(4)^c 0.363	10.00(6)^c 0.242	9.76(1) 11.3	9.65(1) 0.539 232(1)	8.55(1)^e 29.8	230 48.9 4
6	15.4 621 (19,13)	> 3^f 0.653	9.85(5) ^c 1.01	several options ^g (see Ref. [27])		9.06(1) 52.6	189^h 79.7 1
7	14.1 368 (11,16)	10.48(4)^c 0.0663 105(1)	9.94(4)^c 3.30	1.38(1)^a 8.21	9.60(1) 0.0550	9.06(5)^c 8.95	182 1.98 > 8
8	12.5 200 (6,8)	0.609(3)^a 0.545	9.96(1) 2.40	9.75(1) 19.1 194(1)	several options ⁱ (see Ref. [27])		212 15.5 > 5
9	15.9 38 (1,6)	10.38(1) 0.152	9.96(1) 2.00		several options ^j (see Ref. [27]) 350(1) with α_3 or α_4		153 0.905 4
10	14.5 1007 (31,15)	1.66(1)^a 0.0702	9.84(5)^c 3.08	1.28(1)^a 1.56	9.68(4)^c 0.371 172(1)	8.97(5)^c 24.5	189 95.6 2
11	16.5 90 (2,26)	10.34(1) 0.0404	9.99(1) 0.103	9.81(1) 6.81 194(1)	9.54(5)^c 0.242	9.08(1) 39.9	238 12.3 7
12	15.1 44 (1,12)	10.56(1) 0.293	9.93(4)^c 0.687	9.77(2) 14.2	0.903^a 0.232 362(1)	8.93(1) 19.0	195 22.7 > 8
13	15.3 210 (6,18)	3.68(1) ^{a,k} 0.0611	9.88(5)^c 0.326	9.78(2) 0.561	9.43(2) 0.652	9.04(1) 5.11	201 10.2 > 6
14	14.9 465 (14,17)	10.52(4) ^c 0.203	10.11(1) 0.327 69(1)	several options ^l (see Ref. [27])		9.09(4)^c 47.1	252 25.3 5
15	17.2 180 (5,20)	10.42(1) 0.437	9.96(1) 0.965	9.75(1) 11.4	9.87(1) 1.69	9.07(1) 4.05	161 134.4 > 7

16	12.6	10.43(1)	9.10(1)	9.77(1)	9.60(1)	9.07(1)	210 ^b
	203	0.159	0.179	11.6	0.607	3.35	64.1
	(6,11)			119(1)	362(1)		> 7
17	16.3	0.467(3) ^a	10.04(5) ^c	9.78(1)	several options ^m		207
	784	0.0649	0.699	4.04	(see Ref. [27])		66.6
	(24,16)						> 4
18	14.4	6.37(1) ⁿ	9.77(1)	9.25(5) ^c	9.58(1)	8.96(4) ^c	216
	236	0.104	1.89	6.59	0.419	0.702	42.3
	(7,12)				303(1)		3
19	14.9	10.51(4) ^c	10.00(1)	9.75(1)	9.89(1)	9.06(4) ^c	224 ^b
	623	0.0400	0.218	3.13	0.273	15.7	27.8
	(19,15)			237(1)			7
20	15.0	10.30(1)	9.97(1)	9.77(1)	9.53(1)	8.86(5) ^c	275 ^b
	842	0.0614	1.12	3.21	0.702	11.8	15.1
	(26,10)			88(1)	434(1)		2
21	15.1	10.54(1)	missing ^o	9.92(1)	9.61(1)	several	176
	79	0.114		9.18	1.12	options ^p	12.0
	(2,15)					(see Ref. [27])	> 3
22	14.5	0.301(3) ^{a,q}	9.90(4) ^c	9.83(1)	9.61(1)	9.07(1)	220 ^b
	536	0.252	1.03	0.752	0.295	6.14	45.9
	(16,24)						> 4
No.		²⁸⁷ 115	²⁸³ 113	²⁷⁹ Rg	²⁷⁵ Mt	²⁷¹ Bh	²⁶⁷ Db
23	13.6	10.63(5) ^c	10.23(1)	1.33(1) ^a	10.33(1)	9.27(7)	191
	903	0.0676	0.0684	0.0161	0.0513	2.60	0.510
	(28,7)						> 7

^aEscaped α particle registered solely by the implantation detector. Statistical assessments imply that it is likely that one out of the ten assigned escape-like events during beam-off periods is random [22].

^bFission event registered by both implantation and box detector.

^c α particle registered by both implantation and box detector.

^d α particle registered by both implantation and box detector, but pixel in box detector not uniquely defined.

^eHigh-energy implant consistent with transfer reaction product 2.45 s prior to this α decay.

^f α particle registered by both implantation and box detector, but into corner pixel and/or guard ring of box detector.

^gDuring beam-on periods, a total of five possible decay events are registered in the pixel between α_2 and α_5 , two with apparent full energy and three escape-like.

^hFission event registered about two days after completion of the in-beam part of the experiment.

ⁱA total of four escape-like events are registered in pixel 200 within 60 s after α_3 , two during beam-on and two during beam-off periods.

^jFour candidates exist for remaining decays in the chain within 10 s after α_2 : One beam-off escape-like and one beam-off reconstructed event, as well as one beam-on reconstructed and one beam-on full-energy event.

^kAssessments based on detector rates imply that this in-beam escape-like event has a random probability of up to 50% [22].

^lA total of four escape-like events are registered in pixel 465 between α_2 and α_5 , two during beam-on and two during beam-off periods.

^mA total of five escape-like events are registered in pixel 784 during beam-on periods within 30 s after α_3 .

ⁿEscaped α particle registered solely by the p-side of the implantation detector while triggered by another event in the implantation detector 10.7 μ s earlier.

^oA 237(1)-keV γ -ray is observed in prompt coincidence with an escape-like event seen during a beam-off period 0.304 s after the 9.92(1)-MeV full energy α -event assigned to ²⁸⁰Rg.

^pA total of three escape-like events are registered in pixel 79 during beam-off periods within 4 s after α_4 .

^qA second escape-like event is observed during a beam-off period 0.152 s after the first one.