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A new assessment of the alleged link between element 115 and element 117 decay chains



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ABSTRACT

A novel rigorous statistical treatment is applied to available data (May 9, 2016) from search and spectroscopy experiments on the elements with atomic numbers $Z = 115$ and $Z = 117$. The present analysis implies that the hitherto proposed cross-reaction link between α -decay chains associated with the isotopes $^{293}117$ and $^{289}115$ is highly improbable.

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During the last two decades, fusion-evaporation reactions of beams of ^{48}Ca impinging on various radioactive actinide target materials (Np, Pu, Am, Cm, Bk, Cf) have been used to study superheavy atomic nuclei with proton numbers up to element $Z = 118$ (see, for instance, Refs. [1,2] and references therein). Following their possible production and subsequent physical separation from the vast number of unwanted nuclear background reaction products, the anticipated superheavy evaporation residues are implanted in dedicated decay-spectroscopy stations. Here, position-, time-, and energy-correlation measurements between implantation and subsequent nuclear decays usually lead to the observation of sequences of α -particle decays, possibly including the coincident detection of γ rays, X rays, or conversion electrons.

Unfortunately, all hitherto observed α -decay chains produced by these ^{48}Ca -induced nuclear reactions with actinide targets proceeded through previously unknown nuclei and conclude with a spontaneous fission (SF) event. So far, none of these decay chains revealed a connection to well-known isotopes on the chart of the nuclides (see, e.g., Refs. [1,2]).

One indirect method to support that a new element was produced is via cross reactions or cross bombardments. The idea is that the same α -decay chain is entered by two or more different nuclear reactions. For example, the α -decay chains comprising the superheavy nuclei $^{289}115$, $^{285}113$ and ^{281}Rg are considered to have

been populated following two different fusion-evaporation reactions, namely via $^{243}\text{Am}(^{48}\text{Ca},2n)^{289}115$ and $^{249}\text{Bk}(^{48}\text{Ca},4n)^{293}117$. The isotope $^{293}117$ can decay with α -particle emission into its daughter nucleus $^{289}115$. With these interpretations of the observed decay chains, similar average decay characteristics of $^{289}115$, $^{285}113$, and ^{281}Rg produced in the two nuclear reactions are invoked to conclude a cross-reaction case, as detailed in Ref. [3]. Based on a novel, comprehensive statistical method and including all relevant decay data available to date (May 9, 2016), we show in the present study that a cross-reaction case as described in Ref. [3] is highly improbable.

Amongst an ensemble of by now more than one hundred decay chains associated with the production of element 115, four short recoil- α - α -SF decay chains, labelled D1–D4, were observed at relatively low beam energies in the reaction $^{48}\text{Ca} + ^{243}\text{Am}$ [4]. The individual correlation times of these short element 115 chains are shown in Fig. 1(a) as blue squares (D1, D2, D4) and red diamonds (D3). The violet line and its 1σ -uncertainty band [5] show the lifetime averages of chains D1–D4. The blue line and its 1σ -uncertainty band show the lifetime averages excluding D3, which exhibits exceptionally long correlation times compared with the other three short chains for all three decay steps. See Appendix A and Ref. [6] for more information on short element 115 chains. The dashed black line and its grey 1σ -uncertainty band represent the lifetime averages of ten out of sixteen chains associated with $^{293}117$ [7–10]. The selection of ten chains is inferred from Ref. [3]. For a compilation of decay data concerning all sixteen short element 117 chains, see Table 2 Appendix B.

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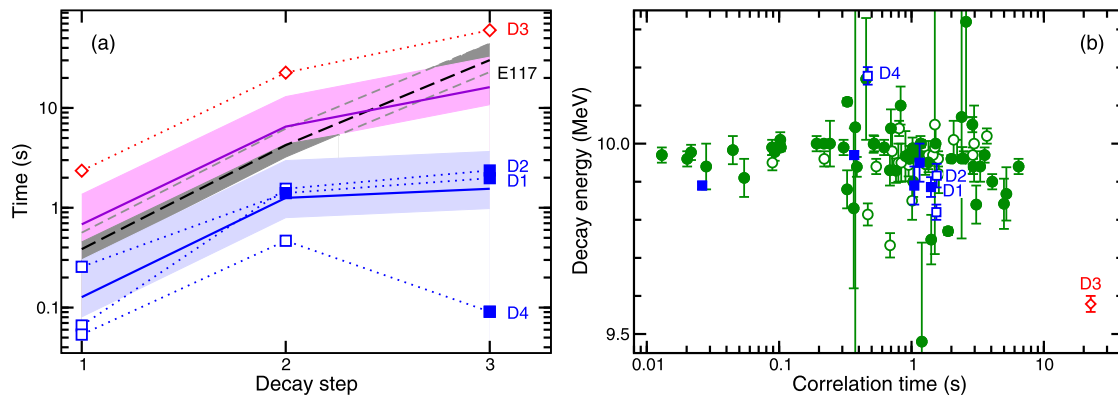


Fig. 1. (Colour online.) (a) Individual correlation times, Δt_i , of four short element 115 decay chains (D1–D4) and average lifetimes, τ , extracted for different data ensembles from element 115 and 117 decay chains (cf. Table 3 Appendix C). The lines are to guide the eye along decay step 1 (α decay from element 115) via decay step 2 (α decay from element 113) towards decay step 3 (SF or α decay from Rg, element 111). (b) Relation between measured decay energies and correlation times of the second decay step, $Z = 113 \rightarrow \text{Rg}$, for different data ensembles from element 115 decay chains. Data points from short element 115 chains are plotted as blue squares and a red diamond (D3). Green circles reflect data points from 96 long element 115 chains. In both panels, filled (open) symbols indicate detection during beam-off (beam-on) periods.

The lifetime averages of the four short element 115 (violet band) and the ten element 117 chains (grey band) are only consistent when the unusual chain D3 is included in the corresponding element 115 averaging procedure. This motivates a thorough statistical assessment of whether or not this chain has indeed the same radioactive origin as D1, D2, and D4.

In addition to chains D1–D4, two recoil- α -SF and one recoil- α - α -SF chains were observed in the reaction $^{48}\text{Ca} + ^{243}\text{Am}$ at Lawrence Berkeley National Laboratory (LBNL), United States of America, and are presented in the Supplemental Material of Ref. [11] (B1–B3). The derived average lifetimes of these three short decay chains are consistent with the lowest 1σ -uncertainty band in Fig. 1(a), i.e. they agree with the D1, D2, and D4 average. Two further recoil- α -SF and five additional recoil- α - α -SF chains were observed in the reaction $^{48}\text{Ca} + ^{243}\text{Am}$ at the GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany [6,12–14] (T1–T7). Adding these seven short decay chains into the analysis, the derived average lifetimes also converge within the lowest 1σ -uncertainty band in Fig. 1(a). Hence, all three correlation times in the D3 chain remain as the – by far – longest times observed in the complete set of fourteen short element 115 chains observed world wide. The call for excluding chain D3 from this element 115 data set due to obvious non-congruence is thus substantiated by data subsequently acquired at other laboratories. For more details and numbers, see Refs. [6,14] and Table 3 Appendix C.

Energy-time correlations of the $Z = 113 \rightarrow \text{Rg}$ decay stemming from decay chains associated with element 115 are shown in Fig. 1(b). This panel is similar to the more complete set of diagrams in Fig. 2 of Ref. [15]. Note the outstanding position of the D3 chain in the lower right corner of the diagram (red diamond). Any other data point from short element 115 chains (blue squares) is hardly distinguishable from the entries generated by 96 long chains commonly associated with $^{288}\text{115}$ (green circles) [4,11,16–19]. Only a forced average of the D3 decay energy with D1, D2, and D4 energies may provide a possible overlap in decay energy with the forced average of the decay energies of ten short element 117 chains (see Table 3 Appendix C).

One may inspect Fig. 2 of Ref. [18] for a possible alternative interpretation of the origin of short element 115 chains: Ignore the upper left part of the figure showing element 117 results. Then compare the decay characteristics of the short element 115 chain (in our work denoted D1) with the averages of 24 long element 115 chains known at the time: There is no apparent difference besides the decay mode, i.e. the colour of the Rg-square. Interestingly,

Table 1

Figure-of-Merit (FoM) [6,15,20] for different subsets of element 115 and element 117 data, and the corresponding 90% and 98% double-sided confidence intervals. The subset denoted D comprises all four short element 115 chains from Dubna experiments (D1–D4) [4], B consists of the three short chains from the Berkeley experiment (B1–B3) [11], and T consists of the seven short chains measured at TASCA, GSI (T1–T7) [6]. The ensemble D' consists of the three chains D1, D2 and D4, i.e. the D3 chain has been excluded. The ensemble E117 consists of the ten short element 117 chains that were considered in Ref. [3]. See also Table 2 Appendix B

Ensemble	FoM	90%-interval	98%-interval
D	0.114	[0.136,0.272]	[0.110,0.254]
D & B	0.120	[0.156,0.258]	[0.134,0.275]
D & B & T	0.162	[0.181,0.255]	[0.164,0.269]
D'	0.223	[0.124,0.248]	[0.099,0.265]
D' & B	0.223	[0.150,0.258]	[0.126,0.275]
D' & B & T	0.215	[0.178,0.256]	[0.162,0.269]
E117	0.165	[0.170,0.255]	[0.152,0.270]
D & E117	0.146	[0.181,0.253]	[0.165,0.266]
D' & E117	0.155	[0.179,0.253]	[0.163,0.266]
D3 & E117	0.160	[0.174,0.254]	[0.156,0.268]

on a one-by-one basis, all short element 115 chains besides D3 are also indistinguishable from the (average of the) 96 five- α -long chains commonly associated with $^{288}\text{115}$.

Elaborated statistical measures presented in Refs. [6,15,20] evidence that the world data set of the in total fourteen short element 115 chains is not congruent. In brief, the hypothesis that they can all be characterised by one half-life for each decay step is tested by comparing a Figure-of-Merit (FoM) for the experimental data set with the distribution of FoMs from generated data sets that do fulfill the hypothesis. 90% and 98% double-sided confidence intervals for FoMs for some relevant cases are presented in Table 1. If the experimental FoM for an ensemble is above the confidence interval, the spread in correlation times is too small, which indicates that the data may not originate from a radioactive decay. On the contrary, a too small FoM indicates that the spread in correlation times is too large, which indicates that the ensemble contains more than one radioactive source.

Already the evaluation of the congruence of the first four short element 115 chains (D1–D4) fails the hypothesis of a single radioactive source with $>95\%$ confidence level, as the obtained FoM is below the double-sided 90% confidence interval. Adding the three short chains from the LBNL experiment (B1–B3) and, further, the seven short chains from the GSI experiment (T1–T7) into the statistical analysis, the hypothesis of a common origin of all 14

chains can be rejected on a >99% confidence level. This is summarised in the upper part of Table 1. It can further be shown that all short chains but D3 are statistically consistent with each other, i.e. they can originate from one radioactive source [6]. This is illustrated, for instance, in Fig. 2 of Ref. [6].

Similarly, the hypothesis that the ten short element 117 chains form one common sequence must also be rejected on a >95% confidence level, as shown in the lower part of Table 1. Further, the ten element 117 chains together with the four element 115 chains from Dubna do not, on a close to 100 % confidence level, form a common ensemble either. This is in contradiction to the scenario presented in Ref. [3]. Adding the chains D1, D2, and D4, to the ten element 117 chains does not, with a confidence level of >99%, produce a congruent data set. Adding only chain D3 to the ten element 117 chains produces a data set that is not congruent on a >95% level. Table 3 Appendix C contains the respective average half-life values. Note that there are relevant cases in Table 3 that already fail the single-step Schmidt-test [21], of which the present congruence method is an extension.

To conclude, by employing a novel statistical method we have evidenced that the presently published world data does not result in the link between element 115 and 117 decay chains suggested previously. The analysis forming the basis for the hitherto proposed cross-reaction case in Ref. [3] is found to be insufficient. In turn, there are indications for possible links between element 115 and element 117 decay chains in the published data [23]. However, a corresponding careful assessment is beyond the scope of the present letter [24], while a scientifically clear case will most likely require more dedicated high-quality spectroscopic data.

Finally, we would like to point out that the present article does not put the underlying experimental results in question, but the new assessment is concerned with the interpretation and conclusions drawn from the original data.

Acknowledgements

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Appendix A. The element 115 basis

First data on four five- α -long decay chains associated with element 115 were published in 2004. The experiment was conducted 2003 at the Joint Institute for Nuclear Research (JINR) in Dubna, Russia [16]. A detailed description of that experiment followed one year later [17]. Additional nuclear α -decay spectroscopy experiments at Dubna in 2010 and 2011 led to a more comprehensive data set of in total 37 element 115 decay chains [18,4]. According to the interpretation in Ref. [4], two five- α ‘long chains’ stem from $^{287}\text{115}$ ($4n$ evaporation channel), 31 five- α long chains start from $^{288}\text{115}$ ($3n$ channel), and four two- α ‘short chains’ originate from $^{289}\text{115}$ ($2n$ channel). Tables II, III and IV in Ref. [4] compile all relevant “decay data for individual events” (cf. page 150 Ref. [3]) for those 37 decay chains.

A first independent experiment was conducted at the GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany, in 2012. Beyond plain detection of element 115 decay chains, high-resolution photon- α coincidence techniques were employed to search for Z -identification by means of characteristic X-ray measurements [22]. The decay data for individual events of 23 observed five- α long chains are available in Table I of the Supplemental Material of Ref. [19]. The detailed material on two recoil- α -SF and five recoil- α - α -SF element 115 decay chains from

Table 2 Compiled data used for the assessment of sixteen decay chains, which have been associated with the decay of $^{283}\text{117}$ in Refs. [7–10]. Decay chains marked with a symbol in the leftmost column are those ten element 117 chains used in Ref. [3] in subsets of three (S03, S04, S05; *) plus three (S06, S09, S10; †) plus four (S11–S14; #) chains (cf. Table 5 in Ref. [3] and Table 3 in the present manuscript). Excitation energies, E^* , of the compound nucleus and measured decay energies, $E(Z)$, along the chains as well as correlation times, $\Delta t(Z)$, are listed. Entries in bold were recorded during beam-off periods. Entries in italics relate to missed preceding α -decay events. Uncertainties in decay energies are here given as σ_E to the best of our knowledge. See text for details. N_{random} corresponds to the number of chains of a given type claimed to arise from random background as given in Refs. [8–10].

chain ID	Ref.	E^* (MeV)	$E(117)$ (MeV)	$\Delta t(117)$ (ms)	$E(115)$ (MeV)	$\Delta t(115)$ (s)	$E(113)$ (MeV)	$\Delta t(113)$ (s)	$E(111)$ (MeV)	$\Delta t(111)$ (s)	N_{random}
S01	[7,8]	372–41.4	10.99(4)	1701	missing		9.72(4)	16.17	SF	40.19	$6 \cdot 10^{-6}$
S02	[7,8]	372–41.4	11.14(4)	789	missing		9.52(43)	2.23	SF	4.25	$1 \cdot 10^{-3}$
S03*	[7,8]	372–41.4	11.08(8)	460	10.34(4)	0.0175	9.71(40)	1.17	SF	12.03	$1 \cdot 10^{-5}$
S04*	[7,8]	372–41.4	10.91(4)	530	10.25(4)	0.5118	9.79(4)	0.238	SF	31.66	$3 \cdot 10^{-11}$
S05*	[7,8]	372–41.4	11.00(4)	20.24	10.27(8)	0.4244	9.48(4)	13.49	SF	76.56	$3 \cdot 10^{-11}$
S06†	[9]	370–41.9	10.90(4)	7.525	10.37(28)	0.2665	9.857(17)	1.5155	SF	9.4192	$< 1 \cdot 10^{-5}$
S07	[9]	370–41.9	11.142(28)	3.305	10.310(28)	0.1719	missing		SF	7.4538	$< 5 \cdot 10^{-10}$
S08	[9]	370–41.9	11.114(38)	153.948	missing		9.631(29)	19.0456	SF	1.4809	$< 1 \cdot 10^{-5}$
S09†	[9]	370–41.9	10.914(29)	10.547	10.198(29)	1.4348	9.36(30)	1.3153	SF	103.406	$< 5 \cdot 10^{-10}$
S10†	[9]	370–41.9	10.598(21)	109.878	10.217(21)	0.1510	9.683(21)	18.3997	SF	42.1349	$< 1 \cdot 10^{-14}$
S11#	[10]	40.3–44.8	10.969(29)	0.043	10.69(33)	0.0136	9.902(29)	0.4281	SF	15.0551	$2 \cdot 10^{-11}$
S12#	[10]	40.3–44.8	11.183(20)	2.553	10.364(20)	0.9572	9.845(20)	1.3712	SF	4.7521	$3 \cdot 10^{-14}$
S13#	[10]	40.3–44.8	11.203(30)	8.173	10.279(30)	0.0565	9.867(30)	4.2130	SF	3.4642	$4 \cdot 10^{-17}$
S14#	[10]	43.8–48.3	11.059(21)	45.762	10.362(21)	0.0161	9.759(46)	0.2589	SF	2.9249	$3 \cdot 10^{-20}$
S15	[10]	43.8–48.3	missing		10.145(28)	0.0852	9.471(28)	0.2456	SF	21.8372	$1 \cdot 10^{-9}$
S16	[10]	43.8–48.3	11.190(30)	36.424	missing		missing		SF	4.4880	$2 \cdot 10^{-4}$

Table 3

This is a more comprehensive and corrected version of Table 5 (top) in Ref. [3]. The upper part of the Table compiles average [5] half-life values, $T_{1/2}$, for different subsets of data associated with element 117 (cf. Table 2) and element 115 [4,11,19]. The descriptors of the data sets follow Ref. [3] and Ref. [6], respectively. For the sake of completeness, the lower part of the Table presents average decay energies for the various subsets. E_w denotes the weighted mean including external and internal errors. E_m is the plain arithmetic mean with standard deviation, σ_m . The uncertainties of individual energy measurements presented in Refs. [4,7–10] have been treated according to the notes in Appendix B, i.e. converted from FWHM to σ_E when applicable.

Z	Reaction	$^{48}\text{Ca} + ^{249}\text{Bk}$				$^{48}\text{Ca} + ^{243}\text{Am}$				6 × short [4,11]	13 × short [46,11]	96 × long (3n) [4,11,19]
		Set 2010 3 × * [7,8]	Set 2012 3 × † [9]	Set 2013 4 × # [10]	average 10 × [7–10]	D1–D4 4 × [4]	D3 1 × [4]	D1, D2, D4 3 × [4,18]				
117	Observable	$18^{(25)}$	$30^{(41)}$	$10^{(10)}$ ^a	$18^{(8)}$ ^a	0.47 ⁽⁴⁷⁾	1.6	0.09 ⁽¹²⁾	0.12 ⁽⁸⁾	0.22 ⁽⁸⁾	0.17 ⁽²⁾	
115	$T_{1/2}$ (ms)	$0.23^{(32)}$	$0.43^{(59)}$	$0.18^{(6)}$	$0.27^{(12)}$	$4.5^{(45)}$	16	$0.9^{(2)}$	$0.70^{(26)}$	$0.61^{(24)}$	$0.97^{(12)}$	
113	$T_{1/2}$ (s)	$3.5^{(47)}$	$4.9^{(67)}$	$1.1^{(1)}$	$2.9^{(14)}$	$11^{(11)}$ ^a	42	$1.1^{(5)}$	$2.1^{(7)}$	$3.0^{(7)}$	$4.4^{(4)}$	
111	$T_{1/2}$ (s)	$28^{(38)}$	$36^{(13)}$	$4.6^{(15)}$	$21^{(5)}$	$11^{(4)}$						
115	E_w (MeV)	10.29(4)	10.21(11)	10.35(4)	10.30(7)	10.39(5)	10.37(2)	10.40(7)	10.42(13)	10.45(10)	10.44(9)	
	E_m (MeV)	10.29(4)	10.26(8)	10.42(16)	10.33(13)	10.40(9)	10.37(2)	10.40(10)	10.41(13)	10.44(11)	[10.28,10.59] ^b	
113	E_w (MeV)	9.64(15)	9.79(9)	9.85(4)	9.80(10)	9.87(24)	9.58(2)	10.02(14)	10.03(11)	9.95(8)	9.90(24)	
	E_m (MeV)	9.66(13)	9.63(21)	9.84(5)	9.73(17)	9.89(21)	9.58(2)	9.99(13)	10.01(12)	9.97(10)	[9.72,10.14] ^b	

^a For this decay step, the correlation times of this set of chains are not compatible with being of solely one origin: $\sigma_{E,\text{exp}} > \sigma_m$. See Ref. [21] and, related, Refs. [6,14] for details.

^b Energy interval comprising 90% of the yield of the distribution of energies (cf. black lines in Figs. 2(a) and (b) in Ref. [14]).

the same experiment has been available since early 2015 [12], and it has recently been published [6]. Shortly after the GSI experiment, a similar experiment was conducted at Lawrence Berkeley National Laboratory (LBNL), United States of America [11], late Spring 2013. The decay data for individual events of 43 observed five- α long chains, as well as two recoil- α -SF and one recoil- α - α -SF element 115 decay chains, are readily available in the Supplemental Material of Ref. [11]. Hence, the detailed decay data information on in total $37 + 30 + 46 = 113$ decay chains associated with the observation of element 115 was accessible since early 2015. This includes the $4 + 3 + 7 = 14$ short decay chains relevant for the element 115 and element 117 cross-reaction case.

Appendix B. Compilation of element 117 data

For convenience, published decay data on sixteen short element 117 chains are compiled in Table 2. This information is otherwise spread out over Refs. [7–10]. In Table 2 uncertainties in decay energies are given as σ_E , where the following procedure has been followed: full-width at half-maximum (FWHM) values for full energy (FWHM ≈ 60 –140 keV in Refs. [7,8] and FWHM ≈ 34 –73 keV in Refs. [9,10]) and reconstructed α events (FWHM ≈ 160 –230 keV in Refs. [7,8] and FWHM ≈ 83 –120 keV in Refs. [9,10]) are divided by 2.35, while $\sigma_E \approx 300$ –400 keV for side-detector α -decay events is kept [7–10]. For chains S01–S05 we assume average values for σ_E based on Table II in Ref. [8].

Appendix C. Revised cross-reaction data compilation

The numbers presented in Fig. 3 and Table 5 (top) of Ref. [3] are inconsistent with each other and with the original data. Table 3 in the present manuscript provides a revised compilation of relevant numbers for the previous and present assessments.

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