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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 293117 and 289115 is highly improbable.

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During the last two decades, fusion-evaporation reactions of beams of 48Ca 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 radioactive 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 48Ca-induced nuclear reactions with actinide targets proceed 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-decays comprising the superheavy nuclei 285115, 285113 and 281Rg are considered to have been populated following two different fusion-evaporation reactions, namely via 243Am(48Ca,2n)289115 and 249Bk(48Ca,4n)293117. The isotope 293117 can decay with α-particle emission into its daughter nucleus 289115. With these interpretations of the observed decay chains, similar average decay characteristics of 289115, 285113, and 281Rg 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 48Ca + 243Am [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 293117 [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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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-$\alpha$-SF and one recoil-$\alpha$-$\alpha$-SF chains were observed in the reaction $^{48}$Ca + $^{243}$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-$\alpha$-SF and five additional recoil-$\alpha$-$\alpha$-SF chains were observed in the reaction $^{48}$Ca + $^{243}$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 worldwide. 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 $Z = 113 \rightarrow$ 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}$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, on a one-by-one basis, all short element 115 chains besides D3 are also indistinguishable from the (average of the) 96 five-$\alpha$-long chains commonly associated with $^{288}$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 is still not supported. The experimental FoM is below the double-sided 90% confidence interval. Thus, the radioactive sources of the element 115 chains are different from each other.
Supplemental data on two recoil-α and five recoil-α+γ element 115 decay chains from 2011. The detailed material of Ref. [19] is available in Table 5 in Ref. [3] and Table 3 in the present manuscript. Excitation energies, E°, of the compound nucleus and measured decay energies, E(1), along the chains as well as correlation times, Δt, 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 given as σE to the best of our knowledge. See text for details. Nrandom corresponds to the number of chains of a given type claimed to arise from random background as given in Refs. [8–10].

Table 2

<table>
<thead>
<tr>
<th>Chain ID</th>
<th>Ref.</th>
<th>E° (MeV)</th>
<th>E(117) (MeV)</th>
<th>Δt(117) (ns)</th>
<th>Δt(115) (s)</th>
<th>E(115) (MeV)</th>
<th>Δt(113) (s)</th>
<th>E(113) (MeV)</th>
<th>Δt(111) (s)</th>
<th>Nrandom</th>
</tr>
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<tr>
<td>S01</td>
<td>[7,8]</td>
<td>372±4.14</td>
<td>1099(4)</td>
<td>17.01</td>
<td>missing</td>
<td>9.72(4)</td>
<td>16.17</td>
<td>SF</td>
<td>40.19</td>
<td>6.10^-6</td>
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<tr>
<td>S02</td>
<td>[7,8]</td>
<td>372±4.14</td>
<td>1114(4)</td>
<td>7.89</td>
<td>missing</td>
<td>9.52±4.32</td>
<td>22.3</td>
<td>SF</td>
<td>4.25</td>
<td>1.10^-3</td>
</tr>
<tr>
<td>S03+</td>
<td>[7,8]</td>
<td>372±4.14</td>
<td>1108(4)</td>
<td>4.60</td>
<td>10.3±4.41</td>
<td>9.71±4.1</td>
<td>11.7</td>
<td>SF</td>
<td>12.0±3</td>
<td>1.10^-5</td>
</tr>
<tr>
<td>S04+</td>
<td>[7,8]</td>
<td>372±4.14</td>
<td>1091(4)</td>
<td>5.30</td>
<td>10.2±4.01</td>
<td>9.79±4.0</td>
<td>0.238</td>
<td>SF</td>
<td>3.16±6</td>
<td>3.10^-11</td>
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<tr>
<td>S05+</td>
<td>[7,8]</td>
<td>372±4.14</td>
<td>1090(4)</td>
<td>2.024</td>
<td>10.27±8.0</td>
<td>9.46±8.4</td>
<td>1.349</td>
<td>SF</td>
<td>7.65±6</td>
<td>3.10^-11</td>
</tr>
<tr>
<td>S07</td>
<td>[9]</td>
<td>370±4.19</td>
<td>1114(28)</td>
<td>3.305</td>
<td>10.31±28(10)</td>
<td>0.1719</td>
<td>missing</td>
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<td>7.45±38</td>
<td>&lt;5.10^-10</td>
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<tr>
<td>S11+</td>
<td>[10]</td>
<td>403±4.48</td>
<td>1096±29</td>
<td>0.043</td>
<td>10.6±99.33</td>
<td>9.90±29.2</td>
<td>0.4281</td>
<td>SF</td>
<td>15.0±561</td>
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</tr>
<tr>
<td>S13+</td>
<td>[10]</td>
<td>403±4.88</td>
<td>1102±30</td>
<td>8.173</td>
<td>10.27±90.19</td>
<td>9.86±70.3</td>
<td>4.21±30</td>
<td>9.3±46.3</td>
<td>3.46±26</td>
<td>4.10^-17</td>
</tr>
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<td>S14+</td>
<td>[10]</td>
<td>438±4.83</td>
<td>1105±21</td>
<td>45.762</td>
<td>10.36±21</td>
<td>9.76±96.4</td>
<td>0.2589</td>
<td>9.2±80.21</td>
<td>2.92±49</td>
<td>3.10^-20</td>
</tr>
<tr>
<td>S15</td>
<td>[10]</td>
<td>438±4.83</td>
<td>1119(30)</td>
<td>36.424</td>
<td>missing</td>
<td>4.71±28</td>
<td>0.2456</td>
<td>SF</td>
<td>4.88±50</td>
<td>2.10^-4</td>
</tr>
<tr>
<td>S16</td>
<td>[10]</td>
<td>438±4.83</td>
<td>1119(30)</td>
<td>36.424</td>
<td>missing</td>
<td>4.71±28</td>
<td>0.2456</td>
<td>SF</td>
<td>4.88±50</td>
<td>2.10^-4</td>
</tr>
</tbody>
</table>
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 Σ = 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 provided in Table 2. This information is otherwise spread over Refs. [7–10]. In Table 2 uncertainties in decay energies are given as $\sigma_E$, where the following procedure has been followed: full-width at half-maximum (FWHM) values for full-energy (FWHM $\approx$ 60–140 keV in Refs. [7,8] and FWHM $\approx$ 34–73 keV in Refs. [9,10]) and reconstructed $\alpha$ events (FWHM $\approx$ 160–230 keV in Refs. [7,8]) and FWHM $\approx$ 83–120 keV in Refs. [9,10]) are divided by 2.35, while $\sigma_E$ for side-detector $\alpha$-decay events is kept [7–10]. For chains S01–S05 we assume average values for $\sigma_E$ 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.

References