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

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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 $\alpha$-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 radioactive target 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 $\alpha$-particle decays, possibly including the coincident detection of $\gamma$ rays, X rays, or conversion electrons.

Unfortunately, all hitherto observed $\alpha$-decay chains produced by these $^{48}$Ca-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. This idea is that the same $\alpha$-decay chain is entered by two or more different nuclear reactions. For example, the $\alpha$-decay chains comprising the superheavy nuclei $^{285}115$, $^{285}113$ and $^{281}Rg$ are considered to have been populated following two different fusion-evaporation reactions, namely via $^{243}Am(^{48}Ca,2n)^{289}115$ and $^{243}Bk(^{48}Ca,4n)^{293}117$. The isotope $^{293}117$ can decay with $\alpha$-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-$\alpha$-$\alpha$-SF decay chains, labelled D1–D4, were observed at relatively low beam energies in the reaction $^{48}Ca + ^{243}Am$ [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 $1\sigma$-uncertainty band [5] show the lifetime averages of chains D1–D4. The blue line and its $1\sigma$-uncertainty band show the lifetime averages excluding D3, which experiences 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 $1\sigma$-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.
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 $^{208}$Pb (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-α-long chains commonly associated with $^{208}$Pb.

Elaborated statistical measures presented in Refs. [6,15,20] evidence that the world data set of the in total fourteen short element 115 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.

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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 in 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\(^{115}\) (4n evaporation channel), 31 five-α long chains start from 288\(^{115}\) (3n evaporation channel), and four two-α ‘short chains’ originate from 289\(^{115}\) (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 1 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
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 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 $\sigma_E \approx 300–400$ keV for side-detector $\alpha$-decay events is kept [7–10]. For chains 501–505 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