48Ca+249Bk Fusion Reaction Leading to Element Z=117: Long-Lived α-Decaying 270Db and Discovery of 266Lr


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The superheavy element with atomic number Z = 117 was produced as an evaporation residue in the 48Ca + 249Bk fusion reaction at the gas-filled recoil separator TASCA at GSI Darmstadt, Germany. The radioactive decay of evaporation residues and their α-decay products was studied using a detection setup that allowed measuring decays of single atomic nuclei with half-lives between sub-μs and a few days. Two decay chains comprising seven α decays and a spontaneous fission each were identified and are assigned to the isotope 294117 and its decay products. A hitherto unknown α-decay branch in 270Db (Z = 105) was observed, which populated the new isotope 269Lr (Z = 103). The identification of the long-lived (T1/2 = 1.0^{+1.9}_{−0.4} h) α-emitter 270Db marks an important step towards the observation of even more long-lived nuclei of superheavy elements located on an “island of stability.”

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The existence of superheavy elements (SHE) was predicted in the late 1960s as one of the first harvests of the macroscopic-microscopic theory of the atomic nucleus [1,2]. Shell effects were predicted to be pronounced at the macroscopic-microscopic theory of the atomic nucleus predicted in the late 1960s as one of the first harvests of stability (see therein). To date, nuclei associated with the “island of stability” can be accessed preferentially in 48Ca-induced fusion reactions with actinide targets [6]. Successful use of these reactions was pioneered at the Dubna Gas-Filled Recoil Separator (DGFRS) at the Flerov Laboratory of Nuclear Reactions in Dubna, Russia [6]. Synthesis of nuclei of SHE with Z up to 118 has been reported [7]. Identification was achieved by detecting their subsequent α-decay chain, which always ended with spontaneous fission (SF) [6,7]. Independent experiments at other laboratories have agreed with findings at the DGFRS [8–16]. Discovery of elements with Z = 114 and Z = 116 was approved by the IUPAC-IUPAP Joint Working Party [17], and recently they were named flerovium (Fl) and livermorium (Lv), respectively [18].
The identification and assignment of nuclear $\alpha$-decay chains of the odd-Z elements 115 [7,16,19] and 117 [20–23] is more difficult compared to that of the even-Z ones. They include more complex $\alpha$-decay patterns and often longer half-lives, $T_{1/2}$. Moreover, odd nucleons significantly hinder the SF decay of these nuclei, essentially leading to long partial SF half-lives. Thus, the reliability of the assignment of members of such chains is often reduced, because the probability to observe random correlations of unwanted background present in the data set increases with increasing the correlation search time, $\Delta t$, between two genuine chain members [5,6]. The longest half-lives detected for $\alpha$-decaying nuclei of SHE are about 2 min, for $^{260}$Sg [12] and $^{271}$Sg [7]. A successful closer approach towards the center of the “island of stability” depends on a reliable identification of single long-lived nuclei of SHE. An attractive option is to advance setups suitable for reliable identification of single long-lived nuclei of SHE.

Here, we report on results of the $^{48}$Ca + $^{249}$Bk fusion reaction leading to the SHE with $Z = 117$. First reports on the observation of two isotopes 293,294$^{117}$ produced in the $^{249}$Bk ($^{48}$Ca, 3–4$n$) reactions came from a collaboration working at the DFGRS [20,21]. Recently, further data were reported, again from the DFGRS [22,23]. This reaction is unique due to the short half-life of $^{249}$Bk ($T_{1/2} = 330(4)$ d, [26]), necessitating dedicated production at Oak Ridge National Laboratory [21] and safe handling of the highly radioactive material. This severely limits the number of independent experiments dedicated to the production of element $Z = 117$.

The experiment was performed at the gas-filled TransActinide Separator and Chemistry Apparatus (TASCA) at GSI [27]. A pulsed (5-ms-long pulses, 50 s$^{-1}$ repetition rate) $^{48}$Ca beam was accelerated to energies of 268.3, 270.2, and 274.1 MeV. Targets in $^{249}$Bk$_2$O$_3$ form were electrodeposited [28] on 2.20(11) $\mu$m thick Ti backings, which faced the beam. Four targets with an area of 6 cm$^2$ each were mounted on a wheel, which rotated synchronously with the beam macrostructure [29]. The $\approx 0.48$ mg/cm$^2$-thick target consisted of $^{249}$Bk (62%) and its decay product $^{249}$Cf (38%) at the beginning of the experiment. The average beam intensity was $4.7 \times 10^{12}$ s$^{-1}$. The beam energies in the center of the target were estimated to be $E_{lab} = 252.1(21)$, 254.0(21), and 258.0(21) MeV [30]. Beam doses of 2.3, 4.9, and $3.9 \times 10^{18}$ were accumulated during 6.3, 10.5, and 10.4 d, respectively. At these beam energies, the evaporation of three or four neutrons is expected from the 297$^{117}$ compound nuclei at excitation energies of $E^* = 37.8–41.3$, 39.4–42.9, and 42.7–46.2 MeV [31] populating the evaporation residues (ER) $^{290}$117 and $^{291}$117 [20–23], respectively.

Compared to the experiments on $^{288,289}$Fl performed in 2009 [11,13], TASCA was significantly upgraded for the current experiment [32]. According to the predicted average charge state ($\approx 6.8$) of $^{293,294}$117 ions moving in helium gas at 0.8 mbar pressure [33,34], the TASCA magnetic system was set to center ions with a magnetic rigidity ($B\rho$) of 2.20 Tm into the focal plane.

The detection system of TASCA consisted of a multi-wire proportional counter (MWPC) and a focal plane detector box (FPDB). The latter consisted of a double-sided silicon strip detector (DSSSD)-based implantation detector (hereafter: stop detector), with eight DSSSDs mounted perpendicular in the backward hemisphere of the stop detector to form a five-sided box configuration (box detectors). The FPDB detection efficiency for $\alpha$ particles emitted from implanted nuclei was estimated to be about 80% based on data from test experiments. The stop detector comprised 144 vertical (X) and 48 horizontal (Y) strips on the front and back sides, respectively. Two adjacent single-sided Si-strip detectors were mounted directly behind the stop detector to register particles passing through the stop detector (veto detector). The efficiency of TASCA to guide ER to the stop detector was estimated to be 47(5)% [35]. The average total triggering counting rate was $\approx 700$ s$^{-1}$, distributed over the almost 7000 pixels of the stop detector.

All preamplifier signals from the Y strips of the stop detector were digitized by 60 MHz-sampling ADCs and stored in 50 $\mu$s-long traces. All other signals, i.e., those from the X strips of the stop detector, from box and veto detectors, and from the MWPC were processed with analog electronics and peak-sensing ADCs with a dead time of about 35 $\mu$s. The stop detector’s analog and digital branches, which independently initialized the data storage, allowed determining the energy of events in two independent ways. Energy calibrations of stop and box detectors were made using an external $\alpha$ source and $\alpha$ decays of implanted nuclei.

Different position and time correlation analyses between ER-, $\alpha$-, and SF-like events were carried out. Promising candidates for decay chains were selected from results of analyses searching for ER-$\alpha$-$\alpha$ ($\Delta t_{ER,\alpha} < 1$ s, $\Delta t_{\alpha,\alpha} < 20$ s) and ER-$\alpha$-$\alpha$-SF ($\Delta t_{ER,\alpha} < 20$ s, $\Delta t_{\alpha,\alpha} < 200$ s, $\Delta t_{\alpha,\SF} < 500$ s) correlations. The search conditions were(i) ER-like events: energy of 3–20 MeV, coincident with a signal from the MWPC; (ii) first and second $\alpha$-like events: 8.5–12 and 8–11 MeV, respectively; (iii) SF-like events: energies > 50 MeV. Alpha and SF-like events were required to be without coincident MWPC signals.

In total, four chains of two different kinds, two long and two shorter ones with significantly different decay properties, all terminated by SF, were identified. All observed chains will be presented in detail in [36]. Here, we focus on the two long decay chains, which are shown in Fig. 1. The chains were detected at $E_{lab} = 254.0$ (chain 1) and $E_{lab} = 258.0$ MeV (chain 2) in strips $X = 103$ and 111, respectively, which are on the right half (higher $B\rho$) of the stop detector. In this area, the implantation rate, which is
Chain 2 occurred in pixel $X = 111$ and $Y = 19$ (Fig. 1). In all beam-off periods during the 258.0 MeV run, fifty-nine $\alpha$-like events within the energy range of 6–12 MeV were detected, resulting in a counting rate of $7 \times 10^{-5}$ s$^{-1}$; six of these are members of chain 2. Only one SF-like event was found in this pixel during the entire 258.0 MeV run; it occurred 29 h after the last $\alpha$ decay. A signal with an energy $E_{\text{stop}} = 4.64$ MeV was detected during a beam-off period in between the $\alpha_2$ and $\alpha_4$ members of the chain and attributed to the $\alpha_3$ member. It was likely emitted into the backward direction under a rather shallow angle, thus leaving a relatively large fraction of its energy in the stop detector, but missed detection in the box detector. In total, only twenty-nine events with energies from 3–6 MeV were registered during the beam-off periods in the entire 258.0 MeV run, supporting the nonrandom origin of this event. The probability to observe random sequences similar to chain 2 was estimated to be $5 \times 10^{-16}$.

The decay properties of the corresponding members of both chains are compatible with one common origin [39]. The high energies exceeding 10 MeV for the first and second $\alpha$ particles and their correlation times point to an origin from heavy nuclei; however, the observed sequence ER-$\alpha_1$-$\alpha_2$ is not compatible with that of any known nucleus with $Z \leq 116$ [7,26]. On the other hand, high energetic $\alpha$-like events, if registered solely with analog electronics, can result from the summing of overlapping low-energy signals [40]. The registered traces of all members of both chains are also presented in Fig. 1. All signals from $\alpha$ particles are recorded as single-peak traces, and the deduced energies from the peak amplitudes agree with those from the analog branches within the energy resolution. Our data allow excluding pile-up events as the source of any member of the two decay chains. Thus, these chains are assigned to originate from the radioactive decay of ER with $Z > 116$, produced in the fusion of $^{48}$Ca either with $^{249}$Bk or $^{249}$Cf, namely, $^{293,294}$117 or $^{293,294}$118. We attribute these chains to $^{294}$117, where the odd proton and neutron are responsible for the longest ever measured nuclear $\alpha$-decay chain originating from SHE [7–16,19].

As is visible in the trace of the ER of chain 1, a small signal with an energy of about 1.4 MeV was observed 1.5 $\mu$s after the ER implantation. The probability to observe a signal with an energy of 1–2 MeV presumably due to heavy charged particles like He-gas atoms or protons in the trace of an ER-like event is less than 1%. The absence of such a signal in the trace of the ER of chain 2, where the ER was implanted deeper into the stop detector, leads us to interpret this signal as not being due to a charged heavy particle from a true member of chain 1 (e.g., an escaped $\alpha$ particle). However, various scenarios leading to the observation of such a signal (e.g., an isomer decaying by cascades of conversion electrons) are being considered [36].
The theoretical calculation of decay properties of odd-odd nuclei is complicated by the scarce information on their structure [3]. However, reliable predictions of the $\alpha$-decay properties of nuclei originating from $^{294}$117 have been given in [38], which satisfyingly agree with results from the DGFRS [20–23]. The predictions from [38] are included in Fig. 1. Best agreement between the experimental and theoretical $\alpha$-decay properties is observed for $^{294}$117, $^{290}$115, $^{286}$113, and $^{270}$Db. The experimental $T_{1/2}$ of $^{266}$Lr is about $3 \times 10^3$ times longer than the theoretical prediction, which can be explained by the presence of odd nucleons. Using the same hindrance factor as a rough approximation, $T_{1/2}$ for $^{282}$Rg, $^{278}$Mt, $^{274}$Bh, and $^{270}$Db can be estimated to 1.2 h, 1.5 min, 1.3 h, and 12 h, respectively. These $T_{1/2}$ values are larger than the predicted $T_{1/2}$, which points to the dominance of $\alpha$ decay, in accordance with experimental observations. A possible SF branch of $\approx 8\%$ can be estimated for $^{270}$Db based on the estimated value for $T_{1/2}$ and the experimental $T_{1/2}$.

A summary of the present experimental data together with results reported from groups working at the DGFRS [20–23] is given in Table I. Comparing these data, there is good agreement in most cases. A new finding in our work is the identification of $\alpha$ decay in $^{270}$Db and the new nucleus $^{266}$Lr. In total four decay chains assigned to $^{294}$117 were reported from the DGFRS. All chains were assigned to be terminated by SF of $^{270}$Db. The measured $\Delta t$ were 33, 38, 24, and 1.2 h [20,22,23]. Respecting our data, which indicate the half-lives of $^{270}$Db and $^{266}$Lr to be $1.0_{-0.4}^{+1.9}$ and $11_{-5}^{+21}$ h, respectively, it appears that the last one of the DGFRS chains, where a significantly shorter $\Delta t$ was recorded than in the three first ones, was instead terminated by SF of $^{270}$Db. In the other cases, the $\alpha$ decay of $^{270}$Db may have remained unidentified (as considered in [20–22]), while the chains terminated by SF of $^{266}$Lr. If so, a branching ratio for SF, of about $1/6 = 17\%$ is deduced for $^{270}$Db. This is in agreement with the estimate given above within the uncertainty of low statistics [39].

The partial SF half-life (6.5 h) of $^{270}$Db is comparable to those of $^{265}$Db ($T_{1/2} \approx 1.4$ h, SF) [19] and $^{268}$Db ($T_{1/2} \approx 25$ h, SF) [19], which lie in the vicinity of the neutron shell closure at $N = 162$. The half-life of the new nucleus $^{266}$Lr with $N = 163$ is comparable to those of the heavier isotones $^{267}$Rf ($T_{1/2} \approx 1.3$ h, SF) [7] and $^{268}$Db. This suggests the stabilizing influence of the $N = 162$ shell closure to extend towards lower $Z$ at least down to $Z = 103$, which is also seen in the low $E_a$ value of $^{270}$Db.

The cross sections for the production of $^{294}$117 were evaluated to $0.7_{-0.6}^{+0.8}$ and $0.9_{-0.7}^{+0.6}$ pb at $E_{lab} = 252.0-256.1$ and $255.9-260.0$ MeV, respectively. These are consistent with values reported from the DGFRS. At $E_{lab} = 250.0-254.1$ MeV no events were observed, corresponding to an upper cross section limit of 2.9 pb [39].

In conclusion, we observed the nuclear decay of two atoms of element $Z = 117$ and its daughter products, synthesized in the $^{48}$Ca + $^{249}$Bk reaction. The nuclei $^{294}$117, $^{290}$115, $^{286}$113, $^{282}$Rg, $^{278}$Mt, $^{274}$Bh, $^{270}$Db were identified by their $\alpha$ decays and $^{266}$Lr by its SF decay, which terminated the decay chains. Results of the present work confirm previously reported data [20–23] on the decay chains assigned to $^{294}$117. In addition, we report a previously unknown $\alpha$ branch in $^{270}$Db, which populated the new SF unstable nucleus $^{266}$Lr. Our data confirm the perseverance of the $N = 162$ shell closure towards lighter $Z$, at least down to $Z = 103$. $^{270}$Db is the most long-lived $\alpha$-decaying nucleus above No ($Z = 102$). The decay chain members from $^{290}$115 to $^{266}$Lr all decay with $T_{1/2} \gtrsim 1$ s, which opens prospects for their chemical investigation and off-line studies. Our experimental data show the sensitivity of TASSCA for the identification of the radioactive decay of single nuclei with half-lives from sub-µs up to about 1 d. Future studies of SHE designed to allow a closer approach towards the center of the “island of stability” may require the safe measurement of even longer half-lives.

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### Table I. Decay properties of the nuclei originating from $^{294}$117 observed in this work, reported from DGFRS [23] and from the combined data sets. Half-life uncertainties corresponding to the 68% confidence level are given, according to [39].

<table>
<thead>
<tr>
<th>Nuclei</th>
<th>$E_a$ (MeV)</th>
<th>$T_{1/2}$</th>
<th>Decay mode</th>
<th>$N_a/N_{SF}$</th>
<th>$T_{1/2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{294}$117</td>
<td>11.05(4)</td>
<td>10.81–10.97</td>
<td>$\alpha$</td>
<td>5/–</td>
<td>Combined</td>
</tr>
<tr>
<td></td>
<td>51.94(30) ms</td>
<td>50.60(18) ms</td>
<td>100% $\alpha$</td>
<td>51.41(16) ms</td>
<td></td>
</tr>
<tr>
<td>$^{290}$115</td>
<td>10.31(4)</td>
<td>9.78–10.28</td>
<td>$\alpha$</td>
<td>5/–</td>
<td></td>
</tr>
<tr>
<td>$^{286}$113</td>
<td>9.3(3)</td>
<td>9.61–9.75</td>
<td>$\alpha$</td>
<td>6/–</td>
<td></td>
</tr>
<tr>
<td>$^{282}$Rg</td>
<td>9.05(3)</td>
<td>8.86(3)</td>
<td>$\alpha$</td>
<td>6/–</td>
<td></td>
</tr>
<tr>
<td></td>
<td>9.01(5)</td>
<td>9.01(5)</td>
<td>100% $\alpha$</td>
<td>2.10(1) min</td>
<td></td>
</tr>
<tr>
<td>$^{278}$Mt</td>
<td>9.45(3)</td>
<td>9.38–9.55</td>
<td>$\alpha$</td>
<td>5/–</td>
<td></td>
</tr>
<tr>
<td>$^{274}$Bh</td>
<td>8.84(3)</td>
<td>8.76(5)</td>
<td>$\alpha$</td>
<td>5/–</td>
<td></td>
</tr>
<tr>
<td>$^{270}$Db</td>
<td>7.90(3)</td>
<td>SF/$\alpha$</td>
<td>2/1</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.0$\pm$0.2 h</td>
<td>17.1$\pm$0.6 h</td>
<td>83/17%</td>
<td>1.1$\pm$0.4 h</td>
<td></td>
</tr>
<tr>
<td>$^{266}$Lr</td>
<td>SF (not observed)</td>
<td>SF</td>
<td>0/2</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>11$\pm$5 h</td>
<td>100%</td>
<td>11$\pm$21 h</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Corresponding $\alpha$ and/or SF branching ratios are given.
*Number of $\alpha$ and SF events used for the calculations of $T_{1/2}$.
*One SF event out of four reported in [23], see text for discussion.
(VH-NG-723), the Australian and Swedish Research Councils, the U.S. Department of Energy by LLNL (DE-AC52-07NA27344), the Laboratory Directed Research and Development Program at LLNL (11-ERD-011) and the Helmholtz Institute Mainz. This work was co-sponsored by the Office of Science, U.S. Department of Energy, and supported under U.S. DOE Grant No. DE-AC05-00OR22725, and at Vanderbilt DOE Grant No. DE-FG05-88ER40407, and the UK Science and Technology Funding Council (STFC). J. M. G., K. E. G., and H. N. were supported by the U.S. Department of Energy, Office of Science, Nuclear Physics, Low Energy Physics Program, through the Lawrence Berkeley National Laboratory under Contract No. DE-AC02-05CH11231.

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[37] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.112.172501, Fig. 1, showing the distribution of implantation counting rates along the X strips of the stop detector.
[40] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.112.172501, Fig. 2, showing an example for a spectrum containing events from the decay-chain $^{222}\text{Pa} \rightarrow ^{218}\text{Ac}$, where energies of both $\alpha$’s are summed up due to the short $T_{1/2} = 1 \mu s$ of the daughter. Only an analysis of digitized data, as they are simultaneously recorded at TASCA, allows resolving pile-up events and producing clear spectra for mother and daughter. While the analog spectrum contains a large number of entries within wide energy range 7–18 MeV, the spectra produced from the digitized data clearly show peaks.