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

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The superheavy element with atomic number $Z = 117$ was produced as an evaporation residue in the ^{48}Ca + ^{249}Bk 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 ^{294}117 and its decay products. A hitherto unknown α-decay branch in ^{270}Db ($Z = 105$) was observed, which populated the new isotope ^{266}Lr ($Z = 103$). The identification of the long-lived ($T_{1/2} = 1.0^{+1.9}_{-0.4}$ h) α-emitter ^{270}Db 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 $Z = 114$ and $N = 184$. Modern theoretical approaches [3,4] confirm this concept. Very long half-lives for these nuclei forming an "island of stability" are expected. In the past 50 years, a wealth of both theoretical and experimental work has been devoted to SHE [5,6] (and references therein). To date, nuclei associated with the "island of stability" can be accessed preferentially in ^{48}Ca-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 α-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 α-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, Δt, between two genuine chain members [5,6]. The longest half-lives detected for α-decaying nuclei of SHE are about 2 min, for ^{269}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 registering various nuclear decay modes. Also, detection of their ions stored in, e.g., Penning traps, has been suggested [24,25].

Here, we report on results of the $^{48}\mathrm{Ca} + {}^{249}\mathrm{Bk}$ 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, 3–4n) reactions came from a collaboration working at the DGFRS [20,21]. Recently, further data were reported, again from the DGFRS [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) $^{48}\mathrm{Ca}^{10+}$ beam was accelerated to energies of 268.3, 270.2, and 274.1 MeV. Targets in $^{249}\mathrm{Bk}_2\mathrm{O}_3$ form were electrodeposited [28] on 2.20(11) μm 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 4.7×10^{12} s^{-1}. The beam energies in the center of the target were estimated to be $E_{\mathrm{lab}} = 252.1(21)$, 254.0(21), and 258.0(21) MeV [30]. Beam doses of 2.3, 4.9, and 3.9×10^{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) $^{294}117$ and $^{293}117$ [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 multiwire 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 α 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 ≈ 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 μs-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 μs. 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 α source and α decays of implanted nuclei.

Different position and time correlation analyses between ER-, α-, and SF-like events were carried out. Promising candidates for decay chains were selected from results of analyses searching for ER-α-α ($\Delta t_{\mathrm{ER}\text{-}\alpha} < 1$ s, $\Delta t_{\alpha\text{-}\alpha} < 20$ s) and ER-α-α-SF ($\Delta t_{\mathrm{ER}\text{-}\alpha} < 20$ s, $\Delta t_{\alpha\text{-}\alpha} < 200$ s, $\Delta t_{\alpha\text{-SF}} < 500$ 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 α-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 $E_{\mathrm{lab}} = 254.0$ (chain 1) and $E_{\mathrm{lab}} = 258.0$ 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

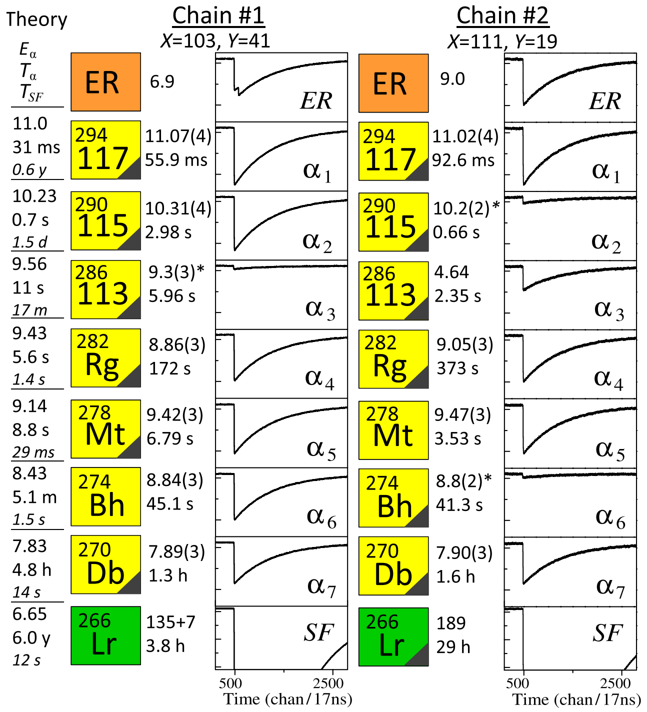

FIG. 1 (color online). Decay chains assigned to $^{294}117$. Experimental energies in MeV and Δt of all events together with their digitized traces are shown. Energies of reconstructed α particles are marked by an asterisk (*). Boxes with black triangles are corresponding to events observed during beam-off periods. The measured fission fragment energies are from stop + box detectors. Theoretically predicted [38] α-decay energies (in MeV), partial α, and unhindered SF half-lives are given on the left.

overall dominated by transfer products that have lower $B\rho$ than the ER, is significantly lower than the average over the whole stop detector [37].

Chain 1 was found in pixel $X = 103$ and $Y = 41$ (Fig. 1). Five α decays occurred during beam-off periods, where the counting rate of α-like events within the energy range of 6–12 MeV was $8 \times 10^{-5}\ \mathrm{s}^{-1}$ in this pixel. This number is based on seventy-four events detected in all beam-off periods during the complete 254.0 MeV run. A small stop detector signal of a fourth member (α_3) of the chain was stored only by the digital branch: an energy of 0.6 MeV was registered in strip $Y = 41$ with $\Delta t = 5.96$ s after α_2, in a beam-off period. A coincident signal was registered in a box detector and a full energy of 9.3(3) MeV was reconstructed. The counting rate for such events with reconstructed full energies of 6–12 MeV in strip $Y = 41$ with missing X signal was $4 \times 10^{-4}\ \mathrm{s}^{-1}$ during beam-off periods. One member, α_6, of chain 1 was detected during a beam-on period, where the counting rate was $1 \times 10^{-3}\ \mathrm{s}^{-1}$. Only one SF-like event was detected in pixel (103,41) during the entire 254.0 MeV run. The counting rate of ER-like events was $4 \times 10^{-3}\ \mathrm{s}^{-1}$. A probability of 5×10^{-15} for the observation of random sequences ER-α_{1-7}-SF similar to chain 1 was calculated, according to [39].

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 α-like events within the energy range of 6–12 MeV were detected, resulting in a counting rate of $7 \times 10^{-5}\ \mathrm{s}^{-1}$; 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 α decay. A signal with an energy $E_{\mathrm{stop}} = 4.64$ MeV was detected during a beam-off period in between the α_2 and α_4 members of the chain and attributed to the α_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 5×10^{-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 α particles and their correlation times point to an origin from heavy nuclei; however, the observed sequence ER-α_1-α_2 is not compatible with that of any known nucleus with $Z \leq 116$ [7,26]. On the other hand, high energetic α-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 α particles are recorded as single-peak traces, and the deduced energies from the peak amplitudes agree with those from the analog branch 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 [3,38] for the longest ever measured nuclear α-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 μs 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 α 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 α-decay properties of nuclei originating from $^{294}117$ have been given in [38], which satisfyingly agreed with results from the DGFRS [20–23]. The predictions from [38] are included in Fig. 1. Best agreement between the experimental and theoretical α-decay properties is observed for $^{294}117$, $^{290}115$, $^{286}113$, and ^{270}Db. The experimental T_{SF} of ^{266}Lr is about 3×10^3 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_{SF} 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_{SF} values are larger than the predicted T_α, which points to the dominance of α 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_{SF} and the experimental T_α.

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 α 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 Δt 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 $1.0^{+1.9}_{-0.4}$ and 11^{+21}_{-5} h, respectively, it appears that the last one of the DGFRS chains, where a significantly shorter Δt was recorded than in the three first ones, was indeed terminated by SF of ^{270}Db. In the other cases, the α 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].

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].

	E_α (MeV) $T_{1/2}$		Decay mode[a]	N_α/N_{SF}[b] $T_{1/2}$
Nuclei	This work	DGFRS [23]	Combined	
$^{294}117$	11.05(4)	10.81–10.97	α	5/-
	51^{+94}_{-20} ms	50^{+60}_{-18} ms	100%	51^{+41}_{-16} ms
$^{290}115$	10.31(4)	9.78–10.28	α	5/-
	$1.3^{+2.3}_{-0.5}$ s	$0.24^{+0.28}_{-0.09}$ s	100%	$0.75^{+0.61}_{-0.23}$ s
$^{286}113$	9.3(3)	9.61–9.75	α	6/-
	$2.9^{+5.3}_{-1.1}$ s	13^{+12}_{-4} s	100%	$7.9^{+5.5}_{-2.3}$ s
^{282}Rg	9.05(3), 8.86(3)	9.01(5)	α	6/-
	$3.1^{+5.7}_{-1.2}$ min	$1.0^{+1.0}_{-0.3}$ min	100%	$2.1^{+1.4}_{-0.6}$ min
^{278}Mt	9.45(3)	9.38–9.55	α	5/-
	$3.6^{+6.5}_{-1.4}$ s	$5.2^{+6.2}_{-1.8}$ s	100%	$4.4^{+3.6}_{-1.4}$ s
^{274}Bh	8.84(3)	8.76(5)	α	5/-
	30^{+54}_{-12} s	54^{+65}_{-19} s	100%	42^{+34}_{-13} s
^{270}Db	7.90(3)	SF	α/SF[c]	2/1[c]
	$1.0^{+1.9}_{-0.4}$ h	17^{+15}_{-6} h	83/17%	$1.1^{+1.5}_{-0.4}$ h
^{266}Lr	SF	(not observed)	SF	-/2
	11^{+21}_{-5} h		100%	11^{+21}_{-5} h

[a] Corresponding α and/or SF branching ratios are given.
[b] Number of α and SF events used for the calculations of $T_{1/2}$.
[c] One SF event out of four reported in [23], see text for discussion.

The partial SF half-life (6.5 h) of ^{270}Db is comparable to those of ^{267}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_α value of ^{270}Db.

The cross sections for the production of $^{294}117$ were evaluated to $0.7^{+1.6}_{-0.6}$ and $0.9^{+2.0}_{-0.7}$ pb at $E_{\mathrm{lab}} = 252.0$–256.1 and 255.9–260.0 MeV, respectively. These are consistent with values reported from the DGFRS. At $E_{\mathrm{lab}} = 250.0$–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 $^{48}\mathrm{Ca} + {}^{249}\mathrm{Bk}$ reaction. The nuclei $^{294}117$, $^{290}115$, $^{286}113$, ^{282}Rg, ^{278}Mt, ^{274}Bh, ^{270}Db were identified by their α 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 α 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 α-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 TASCA 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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[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.

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[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}Pa–^{218}Ac, where energies of both α'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.