Production and Decay of the Heaviest Nuclei $^{293,294}117$ and $^{294}118$

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(Received 16 August 2012; published 15 October 2012)

Two years after the discovery of element 117, we undertook a second campaign using the ^{249}Bk + ^{48}Ca reaction for further investigations of the production and decay properties of the isotopes of element 117 on a larger number of events. The experiments were started in the end of April 2012 and are still under way. This Letter presents the results obtained in 1200 hours of an experimental run with the beam dose of ^{48}Ca of about 1.5×10^{19} particles. The ^{249}Bk target was irradiated at two energies of ^{48}Ca that correspond to the maximum probability of the reaction channels with evaporation of three and four neutrons from the excited ^{297}117. In this experiment, two decay chains of ^{294}117 ($3n$) and five decay chains of ^{293}117 ($4n$) were detected. In the course of the long-term work, ^{249}Cf—the product of decay of ^{249}Bk (330 d)—is being accumulated in the target. Consequently, in the present experiment, we also detected a single decay of the known isotope ^{294}118 that was produced during 2002–2005 in the reaction ^{249}Cf(^{48}Ca, $3n$)^{294}118. The obtained results are compared with the data from previous experiments. The experiments are carried out in the Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, using the heavy-ion cyclotron U400.

DOI: 10.1103/PhysRevLett.109.162501 PACS numbers: 27.90.+b, 23.60.+e, 25.70.Gh

The production and spectroscopic studies of the heaviest elements is one of the most rewarding and challenging investigations in nuclear physics. The discoveries of new superheavy atoms expand simultaneously the periodic table of elements and the Segrè chart of nuclei. New data on decay properties are helping to verify the predicted existence of the so-called ''island of stability'' of superheavy elements. The stabilizing effect of the expected $N = 184$ neutron shell closure found previously for even-Z nuclei [1] is supported by the experimental observation of a decrease of the decay constant for α emission with increasing neutron number. This observation was found to apply to the decays of odd-Z isotopes created during the discovery of elements $Z = 115$ and 117 [2–5]. Longer lifetimes of nuclei located closer to $N = 184$ can facilitate the measurements of chemical properties when the heaviest atoms are produced in sufficient amounts.

However, since the cross sections to create superheavy nuclei are at the picobarn and subpicobarn level, experiments utilizing the current technology last several months and usually only result in few identified nuclei. Therefore, verification of low-statistics data and investigation of new isotopes are of utmost importance. The continuation of experiments results in optimization of the production methods through the determination of excitation functions, as demonstrated in recent studies of the ^{243}Am + ^{48}Ca reaction [4,5].

These goals motivated our new search for $Z = 117$ isotopes among the ^{249}Bk + ^{48}Ca reaction products. The discovery of element 117 in the complete-fusion reaction ^{249}Bk + ^{48}Ca was reported in 2010 [3]. In experiments performed between July 27, 2009, and March 1, 2010, at two projectile energies of 252 and 247 MeV in the middle of the target, corresponding to the excitation energies (E^*) of the compound nucleus ^{297}117 of 39 and 35 MeV, respectively, two isotopes of element 117 were synthesized for the first time.

The excitation energy $E^* = 39$ MeV of the ^{297}117 complete-fusion nucleus is close to the maximum yield of the $4n$ evaporation channel, as expected from the comparison with other fusion-evaporation reactions with ^{48}Ca [1]. Indeed, we synthesized five decay chains of the odd-even isotope ^{293}117 at an excitation energy of 39 MeV. An α decay of ^{293}117 was followed by two consecutive α decays of previously unknown ^{289}115 and ^{285}113, and was terminated by the spontaneous fission (SF) of ^{281}Rg; the decay sequence spanned an interval of about 1 min.

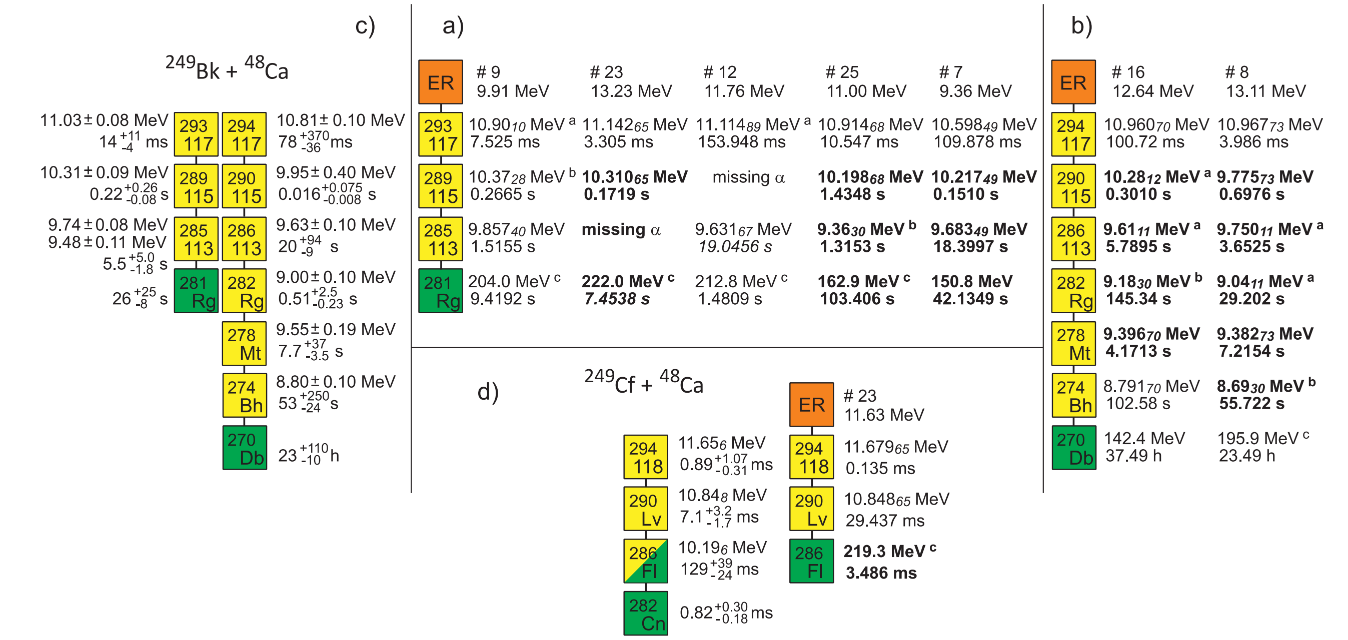

FIG. 1 (color). (a) Decay properties of the isotope $^{293}117$ and its decay products observed in this work. The first rows show ER energies and detector strip numbers. Subsequent rows provide α-particle and SF fragment energies and the time intervals between events. Bold events were registered during a beam-off period. The α-particle energy errors are shown by smaller italic numbers. Time intervals for events following a "missing α" were measured from preceding registered events and are shown in italics (superscript a, α particle registered by both focal-plane and side detectors; superscript b, escaped α particle registered by side detector only; superscript c, fission event registered by both focal-plane and side detectors). (b) The same for isotope $^{294}117$. (c) Average decay properties ($T_{1/2}$ and E_α) of $^{293}117$-^{281}Rg and $^{294}117$-^{270}Db observed in [3]. (d) Average decay properties ($T_{1/2}$ and E_α) of $^{294}118$-^{282}Cn determined in the reaction ^{249}Cf + ^{48}Ca [1,6] (left-hand panel) and properties of $^{294}118$-^{286}Fl measured in this Letter (right-hand panel).

From the well-established behavior of the excitation functions measured for numerous reactions, it followed that a reduction of the projectile energy should result in a decrease of the cross section for the $4n$ channel but permit observation of a heavier isotope with lower α-particle energy and longer lifetime, the product of the $3n$ channel. In an odd-odd nucleus, fission is suppressed compared to α emission because of the unpaired nucleons, resulting in a longer α decay chain. Indeed, at the 35 MeV excitation energy, we produced one longer decay chain of the odd-odd isotope $^{294}117$. In this chain, the great-granddaughter nucleus with $Z = 111$ did not undergo SF, but instead emitted an α particle. It was followed by two more α transitions and then, after about 33 h, a spontaneous fission event was recorded.

For more precise measurement of the radioactive properties of $^{293,294}117$ and their descendant nuclei, we started a new series of experiments. Here, we present results of the two runs performed at ^{48}Ca beam energies of 247 and 252 MeV.

As in the previous experiments from 2009–2010 [3], the ^{249}Bk was produced at Oak Ridge National Laboratory (ORNL) through the intense neutron irradiation of Cm and Am seed material. In 2010–2011, two irradiations were performed at the High Flux Isotope Reactor. The first irradiation of four targets lasted for approximately 250 days. The second irradiation of five targets lasted for only one month (August 2011) but created about half of the total ^{249}Bk material. The Bk fraction was chemically separated from all irradiated targets and was purified at the Radiochemical Engineering Development Center at ORNL; see Ref. [3] for a description of irradiations and chemical procedures. Half of the total ^{249}Bk material, 12.7 mg, was shipped in early March 2012 to the Joint Institute for Nuclear Research (JINR), Dubna. It contained 0.51 mg of ^{249}Cf (β^- decay product of ^{249}Bk) and only 0.45 ng of ^{252}Cf, and no other detectable impurities. Six arc-shaped targets, each with an area of 5.4 cm^2, were made at the Research Institute of Atomic Reactors (Dimitrovgrad, Russian Federation) by depositing BkO_2 onto 0.72 mg/cm^2 Ti foils to a thickness of 0.33 mg/cm^2 of ^{249}Bk. The targets were mounted on the perimeter of a disk 12 cm in diameter that was rotated at 1700 rpm perpendicular to the beam direction. The experiments were performed employing the Dubna gas-filled recoil separator and the heavy-ion cyclotron U400 at JINR.

Based on results of previous experiments [3] in which the ion charge of element 117 evaporation residues (ER) was measured to be $6.1^{+0.3}_{-0.2}$, we used a magnetic rigidity of the separator of 2.395 T $\cdot$ m during the whole run. This is close to the magnetic rigidity of about 2.36 T $\cdot$ m used in the experiments on the synthesis of element 118 [6].

The detection system was modified to increase the position granularity of the detectors, which reduces the probability of observing sequences of random events that mimic decay chains of synthesized nuclei. The new focal-plane detectors consisted of two $6 \times 6\ \mathrm{cm}^2$ silicon detectors each having 16 strips with position sensitivity in the vertical direction. These detectors were surrounded by six similar $6 \times 6\ \mathrm{cm}^2$ side detectors without position sensitivity. Behind the focal-plane detectors, which had a thickness of 0.3 mm, a pair of veto detectors similar to the side detectors was mounted for the detection and rejection of signals from high-energy long-range charged particles (α's, protons, etc., produced in direct reactions of projectiles with the Dubna gas-filled recoil separator media) which can pass through the separator without being detected by the time-off-light system. The FWHM energy resolution of the implantation detector was 34 to 73 keV, while the summed signals recorded by the side and implantation detectors had an energy resolution of about 83 to 117 keV. The FWHM position resolutions of the implantation detector were 1.1–1.8 mm for ER-α and 0.5–1.2 mm for ER-SF signals; compare with [3]. Other experimental conditions, including the method of calibration of the detectors, were the same as in [3]. In order to reduce the background rate in the detector, the beam was switched off for at least 3 min after a recoil signal was detected with $E_{\mathrm{ER}} = 7$–18 MeV, followed by an α-like signal in the focal-plane detector within energy intervals of 10.7–11.3 and 10.0–10.7 MeV and time intervals of 0.4 or 2.0 s, respectively, in the same strip, within a 3.2 mm wide position window.

The experiment was performed from April 23 to July 13, 2012, at two ^{48}Ca projectile energies of 252 and 247 MeV (midtarget) with total beam doses of 1.2×10^{19} particles and 3.4×10^{18} particles, correspondingly. With the energy spread of the incident cyclotron beam, the small variation of the beam energy during irradiation, and the energy losses in the target (3.0 MeV), we expected the resulting $^{297}117$ compound nuclei to have excitation energies of 37.0–41.9 and 32.8–37.5 MeV, respectively. Excitation energies of the compound nuclei are calculated using the masses in [7,8]. The beam energy losses in the separator's entrance window ($0.71\ \mathrm{mg/cm}^2$ Ti foil), target backing, and target layer were calculated using the nuclear data tables in [9,10].

The decay properties of four nuclei originating from the $4n$ evaporation reaction product, $^{293}117$, measured in the five similar decay chains observed in the first run at $E^* = 39$ MeV and seven nuclei in two decay chains originating from the heavier isotope $^{294}117$ observed at $E^* = 35$ MeV, are shown in Figs. 1(a) and 1(b), respectively. In two cases [chains 1 and 3 in Fig. 1(a)], the α particles of the parent nucleus $^{293}117$ were detected by both the focal-plane and side detectors with $E_{f-p} = 3.457$ MeV, $E_s = 7.443$ MeV ($E_{f-p} + E_s = 10.900$ MeV) and $E_{f-p} = 1.376$ MeV, $E_s = 9.738$ MeV ($E_{f-p} + E_s = 11.114$ MeV), respectively. In the first chain, a side-only event with $E_\alpha = 9.99$ MeV was found between α particles with energies 10.90 and 9.86 MeV; in the third chain, this α particle was not detected. Thus, in both of these cases, the beam was not switched off. In the three other cases, the daughter products of $^{293}117$ ($^{289}115$, $^{285}113$, and ^{281}Rg) were observed when the beam was off and thus associated with a very low counting rate of background events.

Despite the detection of two decay chains completely during beam-on intervals, the expected total number of random events ER-$\alpha_{1,\mathrm{on}}$-$\alpha_{3,\mathrm{on}}$-SF$_{\mathrm{on}}$ is lower than 10^{-5}. The expected numbers of random sequences of the types ER-$\alpha_{1,\mathrm{on}}$-$\alpha_{2,\mathrm{off}}$-SF$_{\mathrm{off}}$ (2nd and 4th decay chains) and ER-$\alpha_{1,\mathrm{on}}$-$\alpha_{2,\mathrm{off}}$-$\alpha_{3,\mathrm{off}}$-SF$_{\mathrm{off}}$ (5th chain) are lower than 5×10^{-10} and 10^{-14}, respectively [11]. Note that only eight α particles with $E_\alpha = 9.4$–10.6 MeV and five SF events were registered by the whole focal-plane detector during the total beam-off time interval ($t = 2 \times 10^5$ s); among them, five α particles and three SF events belong to the decay chains of $^{293}117$.

The loss of two α particles [marked "missing α" in Fig. 1(a)] in five decay chains, consisting of three α decays each, is in agreement with the 87% efficiency of the detector array for observing full-energy α particles. The probability of the random appearance of a beam-on signal in one of the six side detectors with $E = 9.6$–10.3 MeV and within a 1 s time interval of a triggering event in strip 9 was about 8%. We tentatively assigned the side-only signal in chain 1 to $^{289}115$; its total energy was estimated to be the sum of the energy measured by the side detector and half of the threshold energy of the focal-plane detector (0.77 MeV for strip 9) with the uncertainty in determining the total energy increased to ≈ 0.3 MeV (68% confidence limit). In the fourth decay chain, the third α decay, $^{285}113$, was also registered by the side detector only but during very low background conditions. The probability of its random origin is about 1%.

The other nine α particles were detected solely by the focal-plane detector. The position deviations between ER signals and two α particles detected by both the focal-plane and side detectors, nine α decays registered entirely by the focal-plane detector as well as five SF events, were in full agreement with the position resolutions of detectors for consecutive ER-α and ER-SF signals.

In two of the decay chains of $^{294}117$, the $3n$ evaporation product, all 12 α decays were registered [see Fig. 1(b)]. Six α particles were absorbed by only the focal-plane detector, four α particles were detected by both the focal-plane and side detectors, and two α particles were detected by the side detector only. The last α decay (^{274}Bh) in the first decay chain was detected when the beam was back on ($\Delta t_{\alpha 1-\alpha 6} > 3$ min). But the probability of this event as a random particle plus two beam-off α particles detected by the side detectors only (without signal in the focal-plane detector) is 0.01–0.03. Within a limited event count of $^{294}117$, for which only a single event was observed in [3], the properties of nuclei in the new decay chains point to the same activities arising from $^{294}117$ detected in two experiments using a ^{294}Bk target. Note that, as it was discussed in [3], in both the new decay chains we observed longer lifetimes for $^{290}115$ and ^{282}Rg compared with the values detected in the first experiment. The summary of radioactive properties of nuclei observed in the reaction $^{249}\text{Bk}(^{48}\text{Ca}, 3n)^{294}117$ [3] is shown in Fig. 1(c).

The radioactive properties of nuclei observed in the reaction $^{249}\text{Bk}(^{48}\text{Ca}, 4n)^{293}117$ in 2010 [3] and this work, as well as in the reaction $^{243}\text{Am}(^{48}\text{Ca}, 2n)^{289}115$ [4,5], are shown in Table I. The radioactive decay properties of $^{293}117$ and all descendant nuclei discovered in 2010 [3] were completely confirmed by registration of five new decay chains in this new series of experiments. One can see in Fig. 1 that the results of the five events in the first experiment are in good agreement with the data of this Letter. Moreover, the α-particle energies and decay times of the isotopes $^{289}115$, $^{285}113$, and ^{281}Rg registered after the α decay of the parent nucleus $^{293}117$ in the reaction $^{249}\text{Bk} + {}^{48}\text{Ca}$ and synthesized directly in the reaction $^{243}\text{Am} + {}^{48}\text{Ca}$ [4,5] are comparable. Therefore, the isotope $^{289}115$ was produced in two reactions with target nuclei ^{243}Am and ^{249}Bk as a $2n$ evaporation product in the first case and as a daughter nucleus after the α decay of the heavier parent nuclide $^{293}117$ in the second case. The observed spread of α energies, often clearly exceeding the energy resolution of detectors, is most likely related to fine structure in α decays. However, the experiments with better energy resolution, better statistics, and the observation of α-γ correlations are needed to corroborate this interpretation.

The cross sections for the $3n$ and $4n$ evaporation channels at $E^* = 35$ and $E^* = 39$ MeV were measured to be $\sigma_{3n} = 3.6^{+6.1}_{-2.5}$ pb and $\sigma_{4n} = 2.0^{+2.2}_{-1.0}$ pb in this work, which are larger but within experimental uncertainties when compared with the previous results of $\sigma_{3n} = 0.5^{+1.1}_{-0.4}$ pb and $\sigma_{4n} = 1.3^{+1.5}_{-0.6}$ pb [3]. These cross section values are consistent with the results of previous experiments where cross sections for the reactions of ^{238}U, ^{237}Np, ^{242,244}Pu, ^{243}Am, ^{245,248}Cm, and ^{249}Cf targets with ^{48}Ca beams have been measured [1–6].

One also should note that the target isotope $^{249}\text{Bk}(Z = 97)$ decays into $^{249}\text{Cf}(Z = 98)$ with a half-life of $T_\beta = 330$ d. During a long experiment, the percentage of ^{249}Bk in the target decreases and the quantity of ^{249}Cf becomes larger. This creates an opportunity to produce $Z = 118$ isotopes during the same experiment in the $^{249}\text{Cf} + {}^{48}\text{Ca}$ reaction [6] after sufficient ^{249}Cf material accumulates in the target layer.

The isotope $^{294}118$ of the new element 118 was produced for the first time in the reaction $^{249}\text{Cf} + {}^{48}\text{Ca}$ [6]. With 245 MeV ^{48}Ca projectiles, one decay chain of $^{294}118$ was observed. An increase of the ^{48}Ca energy to 251 MeV

TABLE I. Decay properties of nuclei originating from $^{294}118$, $^{294}117$, and $^{293}117$.

Z	N	A	Number observed[a]	Decay mode, branch (%)[b]	Half-life[c]	E_α (MeV)[d]
118	176	294	4(4/4)	α	$0.69^{+0.64}_{-0.22}$ ms	11.66 ± 0.06
117	177	294	3(3/3)	α	50^{+60}_{-18} ms	10.81–10.97
	176	293	10(10/10)	α	27^{+12}_{-6} ms	10.60–11.14
116	174	290	11(11/11)	α	$8.3^{+3.5}_{-1.9}$ ms	10.85 ± 0.07
115	175	290	3(3/3)	α	$0.24^{+0.28}_{-0.09}$ s	9.78–10.28
	174	289	11(11/11)	α	$0.38^{+0.18}_{-0.10}$ s	10.20–10.54
114	172	286[e]	25(20/12)	α/SF:50/50	$0.12^{+0.04}_{-0.02}$ s	10.19 ± 0.06
113	173	286	3(3/3)	α	$8.7^{+10.4}_{-3.1}$ s	9.61–9.75
	172	285	14(12/12)	α	$5.6^{+2.2}_{-1.2}$ s	9.48–10.18
112	170	282[e]	12(12/−)	SF	$0.82^{+0.30}_{-0.18}$ ms	
111	171	282	3(3/3)	α	40^{+49}_{-14} s	9.00–9.18
	170	281	14(12/−)	SF	22^{+8}_{-5} s	
109	169	278	3(3/3)	α	$5.2^{+6.2}_{-1.8}$ s	9.38–9.55
107	167	274	3(3/3)	α	54^{+65}_{-19} s	8.69–8.80
105	165	270	3(3/−)	SF	22^{+26}_{-8} h	

[a]Number of observed decays and number of events used for calculations of half-lives and α-particle energies, respectively.
[b]The branching ratio is not shown if only one decay mode was observed.
[c]Error bars correspond to 68% confidence level.
[d]For odd-Z nuclei, the energy range of α particles detected by the focal-plane detector or both the focal-plane and side detectors is shown.
[e]Decay properties of these nuclei are in agreement with those measured in two decay chains in [12,13].

resulted in an increase of the cross section of the $3n$ evaporation channel and two additional $^{294}118$ atoms were synthesized. In two cases, $^{294}118$ underwent two consecutive α decays terminated by the spontaneous fission of ^{286}Fl. In the third decay chain, three α decays of $^{294}118$, ^{290}Lv, and ^{286}Fl were detected that were followed by the SF of ^{282}Cn. Such a decay pattern of $^{294}118$ is consistent with the radioactive properties of ^{286}Fl (a 50% fission branch was determined for its decay in [14,15]) and ^{282}Cn [12–15].

In addition to two nuclei $^{294}117$ at 247 MeV ^{48}Ca, we observed one more decay chain in strip 23. This decay chain consists of an evaporation residue ($E_{\mathrm{ER}} = 11.63$ MeV), two α decays registered during a beam-on interval by the focal-plane detector, and a spontaneous fission within a beam-off pause caused by the ER-α_2 pair [see Fig. 1(d)]. The decay properties of the nuclei in this chain agree well with those determined for $^{294}118$ and its descendant nuclei ^{290}Lv and ^{286}Fl (fission branch of 50%) [1,6,14,15].

The ingrowth of ^{249}Cf in the ^{249}Bk target material at the end of this run was 28%. According to the previously obtained cross section data on the production of the nuclide $^{294}118$ [6], as well as that of the other superheavy nuclides produced in the ^{48}Ca-induced reactions, $^{294}118$ can be observed in the reaction $^{249}\mathrm{Cf}(^{48}\mathrm{Ca}, 3n)$ over the whole energy range from $E^* = 26.6$ MeV [6] to about 40 MeV [1–6,12–15]. Taking into account the buildup of ^{249}Cf in the preceding [3] and present experiments, where $^{294}118$ was not registered, and the total beam dose of ^{48}Ca of 5.9×10^{19} (effective beam dose is about 1.6×10^{19} for a 0.3 mg/cm^2 target), the detected decay chain of $^{294}118$ corresponds to $\sigma_{3n} = 0.3^{+0.7}_{-0.26}$ pb for the total excitation-energy interval of 26.6–37.5 MeV for $^{297}118$. This value is in good agreement with cross sections measured for this reaction at 245 MeV ($E^* = 26.6$–31.7 MeV, $\sigma_{3n} = 0.3^{+1.0}_{-0.27}$ pb) and 251 MeV ($E^* = 32.1$–36.6 MeV, $\sigma_{3n} = 0.5^{+1.6}_{-0.3}$ pb) [6].

In conclusion, the discovery of a new chemical element with atomic number 117 that was reported for the first time in 2010 [3] has now been corroborated through the observation of five additional decay chains originating from the $4n$ evaporation product, $^{293}117$, and two decay chains of the $3n$ channel, $^{294}117$, of the reaction $^{249}\mathrm{Bk} + ^{48}\mathrm{Ca}$. The radioactive decay properties of the 11 isotopes $^{293,294}117$, $^{289,290}115$, $^{285,286}113$, ^{281,282}Rg, ^{278}Mt, ^{274}Bh, and ^{270}Db measured in this work are in full agreement with the results of the first experiment [3] as well as with the decay data determined for $^{289}115$, $^{285}113$, and ^{281}Rg measured in the cross-bombardment reaction $^{243}\mathrm{Am}(^{48}\mathrm{Ca}, 2n)^{289}115$ [4,5]. The average cross sections for the production of $^{293}117$ and $^{294}117$ nuclei in the $^{249}\mathrm{Bk} + ^{48}\mathrm{Ca}$ reaction at $E^* = 39$ and 35 MeV determined from the observation of 10 and 3 events amount to $1.5^{+1.1}_{-0.5}$ pb and $1.1^{+1.2}_{-0.6}$ pb, respectively. The earlier reported discovery of the heaviest known element 118 [6] was confirmed by the observation of one more decay chain of $^{294}118$ and its daughter nuclei ^{290}Lv and ^{286}Fl. The decay properties of the heaviest nuclei were determined more accurately and demonstrate once more the decisive role of nuclear shell effects in the stability of superheavy nuclei.

We are grateful to the JINR Directorate and U400 cyclotron and ion source crews for their continuous support of the experiment. We acknowledge the support of the Russian Foundation for Basic Research Grants No. 11-02-12050 and No. 11-02-12066. Research at ORNL was supported by the U.S. DOE Office of Nuclear Physics under DOE Contract No. DE-AC05-00OR22725 with UT-Battelle, LLC. Research at Lawrence Livermore National Laboratory was supported by LDRD Program Project No. 08-ERD-030, under DOE Contract No. DEAC52-07NA27344 with Lawrence Livermore National Security, LLC. This work was also supported by the U.S. DOE through Grant No. DE-FG-05-88ER40407 (Vanderbilt University). These studies were performed in the framework of the Russian Federation/U.S. Joint Coordinating Committee for Research on Fundamental Properties of Matter.

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