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Synthesis of a New Element with Atomic Number $Z = 117$

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The discovery of a new chemical element with atomic number $Z = 117$ is reported. The isotopes $^{293}117$ and $^{294}117$ were produced in fusion reactions between ^{48}Ca and ^{249}Bk. Decay chains involving 11 new nuclei were identified by means of the Dubna gas-filled recoil separator. The measured decay properties show a strong rise of stability for heavier isotopes with $Z \geq 111$, validating the concept of the long sought island of enhanced stability for superheavy nuclei.

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The existence of the heaviest atomic nuclei bound against immediate disintegration depends on the detailed properties of proton and neutron quantum states; see, e.g., [1,2] and references therein. Therefore, studies aimed at the identification of new superheavy elements contribute to the fundamental knowledge of nuclear potentials and the resulting nuclear structure. The concept of an ''island of stability'' existing near the next spherical doubly magic nucleus heavier than ^{208}Pb arises in every advanced model of nuclear structure. Reactions involving doubly magic ^{208}Pb and singly magic ^{209}Bi target nuclei and stable neutron-rich projectiles as heavy as ^{64}Ni or ^{70}Zn have been used for the synthesis of new heavy elements. These reactions, termed cold fusion, led to the observation of isotopes with $Z \leq 113$ and $N \leq 165$ [3,4], stabilized by the $Z = 108$ and $N = 162$ shell gaps occurring for deformed shapes. The dramatic drop of the production cross section with increasing Z practically excludes the continuation of such experiments for heavier elements.

A new method of synthesizing superheavy elements, with $Z \geq 112$ and neutron numbers closer to the predicted spherical shell closure at $N = 184$, was pioneered at the Flerov Laboratory of Nuclear Reactions (FLNR) of Joint Institute for Nuclear Research (JINR) about a decade ago. Four new isotopes of element $Z = 112$ and 14 new isotopes of new elements with $Z = 113$–116 and 118 were identified [1] among the products of heavy-ion fusion reactions employing doubly magic ^{48}Ca projectiles and actinide radioactive targets of U-Cm and Cf, respectively. The sequential α decays of the heaviest even-Z nuclei were found to be terminated by spontaneous fission (SF) of the descendant even-even or even-odd nuclei with $Z = 114$, 112, or 110 ($T_\alpha \geq T_{SF}$), with total decay times in the range of about 0.1 s to 1 min depending on neutron number [1]. The probabilities of formation and the decay properties of these 18 new nuclei provide evidence of a considerable increase in nuclear stability with increasing neutron number in the nucleus. The production cross sections, the identification, as well as the decay properties of the $Z = 112$ and $Z = 114$ isotopes obtained at Dubna [1] were recently confirmed in several independent experiments [5–8].

We present here the experimental evidence for synthesis of a new chemical element with $Z = 117$; see Fig. 1. The identified $^{293}117$ and $^{294}117$ isotopes were produced in the fusion reaction between ^{48}Ca projectiles and radioactive ^{249}Bk target nuclei followed by the emission of four and three neutrons, respectively. The decay properties of the resulting 11 new neutron-rich nuclides offer additional experimental support for the nuclear shell model predicting the existence of the island of stability for heaviest nuclei. The ^{249}Bk was produced at Oak Ridge National Laboratory (ORNL) through intense neutron irradiation of Cm and Am targets for approximately 250 d in the High Flux Isotope Reactor. The Bk chemical fraction, separated and purified at the Radiochemical Engineering Development Center at ORNL, contained 22.2 mg of ^{249}Bk, only 1.7 ng of ^{252}Cf, and no other detectable impurities. Six arc-shaped targets, each with an area of 6.0 cm^2, were made at the Research Institute of Atomic Reactors (Dimitrovgrad, RF) by depositing BkO_2 onto 0.74 mg/cm^2 Ti foils to a thickness of 0.31 mg/cm^2 of ^{249}Bk. The targets were mounted on the perimeter of a disk that was rotated at 1700 rpm perpendicular to the beam direction. The experiments were performed employing the Dubna gas-filled recoil separator [10] and the heavy-ion cyclotron U-400 at JINR. A detailed description of experiment will be given in a forthcoming

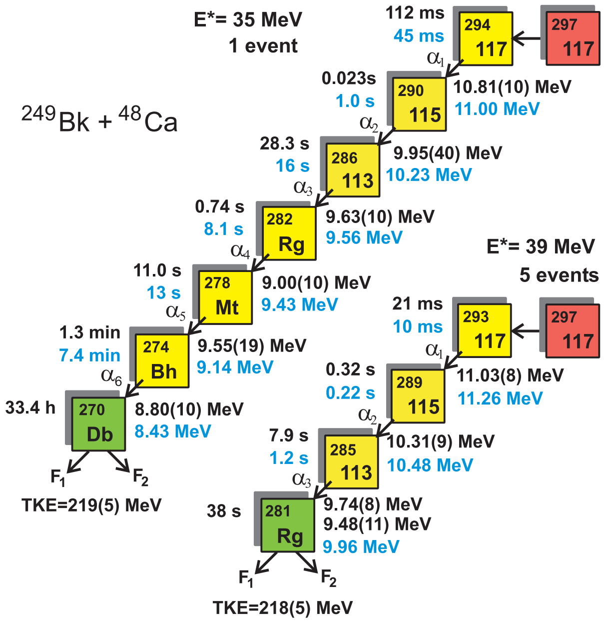

FIG. 1 (color). Observed decay chains interpreted as originating from the isotopes $A = 294$ (single event) and $A = 293$ (average of five events) of the new element $Z = 117$. The deduced and predicted [9] lifetimes ($\tau = T_{1/2}/\ln 2$) and α-particle energies are shown in black and blue, respectively.

paper [11]; here we present the basic features. Evaporation residues (ER) passing through the separator with an overall transmission about 35% were registered by a time-of-flight system with a detection efficiency of 99.9%, and were implanted in a 4 cm $\times$ 12 cm Si-detector array with 12 vertical position-sensitive strips surrounded by eight 4 cm $\times$ 4 cm side detectors. The position-averaged detection efficiency for α particles emitted from implanted nuclei was 87% of 4π. The energy resolution for α particles implanted in the focal-plane detector measured as a full width at half maximum (FWHM) was 60–140 keV, depending on the strip and the position within the strip. Alpha-escape signals detected in the side detectors had an energy resolution of 160–230 keV. If an α particle was detected only by a side detector (its position was lost), the total energy was estimated as a sum of the energy measured by the side detector and half of the threshold energy ($\approx$0.5 MeV), with the uncertainty in the total energy increased to $\approx$ 0.4 MeV. The total kinetic energy (TKE) released in the SF of nuclei with $Z \geq 102$ was determined from the sum $E_{\rm tot}$ + 23 MeV, where $E_{\rm tot}$ is the observed energy signal (with a systematic uncertainty of about 5 MeV when both fission fragments were detected) and 23 MeV is the correction related to the pulse height effect and energy loss in the dead layer detector as determined from a ^{252}No measurement. The position resolution (FWHM) of the strip detector in registering correlated decay chains of the ER-α_1-α_2-α_3-SF type was $\leq$1.2 mm. 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 parameters of implantation energy expected for $Z = 117$ ERs, followed by an α-like signal with an energy between 10.7 and 11.4 MeV, in the same strip, within a 2.2 mm wide position window. For the ^{48}Ca projectiles at 252 MeV energy in the middle of the ^{249}Bk target, the excitation energy of the compound nucleus ^{297}117 is estimated to be $E^* = 39$ MeV, near the expected maximum for the total ER cross section (sum of 3 n and 4 n evaporation channels [1]). The intensity of the ^{48}Ca-ion beam was 7×10^{12} ions/s at the target. Irradiation at this beam energy was performed for 70 d between July 27 and October 23, 2009, with a total beam dose of 2.4×10^{19}. The beam was interrupted for a total beam-off time of 79 h. The energy spectra of the α-like signals registered by the front detector during all 1680 h of the irradiation and those registered only in the beam-off intervals are presented in Fig. 2(a). The background in the beam-off spectrum is due to the decay of Po isotopes that are daughters of the heavier nuclei produced in transfer reactions. We observed five position-correlated decay chains in the 252-MeV ^{48}Ca irradiation; in each case, two or three α decays were observed between the time of arrival of the ER and the detection of SF (see Fig. 1 for the averaged decay properties assigned to ^{293}117 isotope). All

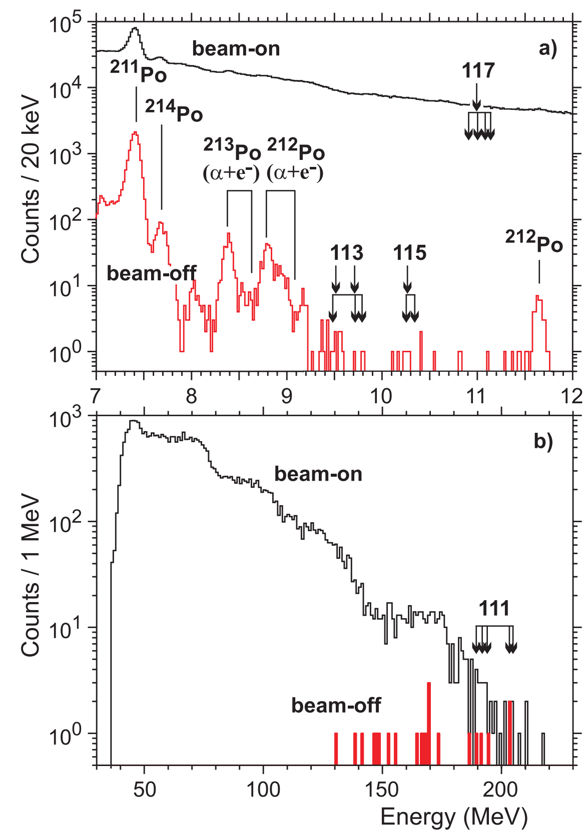

FIG. 2 (color). Energy spectra recorded during the 252 MeV ^{48}Ca + ^{249}Bk run ($E_* = 39$ MeV). (a) Total energy spectra of beam-on α-like signals and beam-off α particles. (b) Total fission-fragment energy spectra, both beam on and beam off. The arrows show the energies of events observed in the correlated decay chains; see Fig. 1.

five of the first α particles emitted after the implantation of the recoils have the same energies (within the energy resolution of the focal-plane detector), yielding average values for the α energy $E_{\alpha_1} = 11.03 \pm 0.08$ MeV and $T_{\alpha_1} = 14(+11, -4)$ ms. The energies of the α particles emitted by the daughter nuclei and detected in three out of five chains were the same within the accuracy of the measurements, resulting in $E_{\alpha_2} = 10.31 \pm 0.09$ MeV and $T_{\alpha_2} = 0.22(+0.26, -0.08)$ s. The third α transition was observed as having $E_{\alpha_3} = 9.74 \pm 0.08$ MeV and $E_{\alpha_3} = 9.48 \pm 0.11$ MeV, and $T_{\alpha_3} = 5.5(+5.0, -1.8)$ s. In all five cases the decay chains ended with the spontaneous fission with $T_{SF} = 26(+25, -8)$ s.

At the $E^* = 39$ MeV excitation energy, the maximum cross section is expected for the 4 n evaporation channel; therefore we assign the observed decay chains as originating from the isotope $^{293}117$. This conclusion is supported by the systematics of the cross sections $\sigma_{xn}(E^*)$ measured previously for production of isotopes of superheavy nuclei with $Z = 108$, 112–116, and 118 in ^{48}Ca-induced reactions [1], by calculations made directly for the evaporation residues of the reaction ^{249}Bk + ^{48}Ca [12–14] as well as by the result of the ^{249}Bk + ^{48}Ca experiment performed at lower beam energy (see below). In the E_α energy range between 8.8 and 11.3 MeV, where we expect α particles of the first five transitions $117 \rightarrow 115 \rightarrow 113 \rightarrow 111 \rightarrow 109 \rightarrow 107$, the counting rate was 0.17/s (with beam on) and 10^{-3}/s (beam off) for the whole area of the front detector. Similar spectra of fission fragmentlike signals measured under the same conditions are shown in Fig. 2(b). In the energy range $E_{SF} \geq 135$ MeV, the SF counting rate in the front detector was 1.2×10^{-4}/s (beam on) and 7×10^{-5}/s (beam off). We have calculated the total numbers of random sequences [15] imitating each of the observed five decay chains, by using extended intervals of time ($\Delta t \geq 5T_{1/2}$), α-particle energy and position (both exceeding 4.7 standard deviations), to be 6×10^{-6}, 10^{-3}, 10^{-5}, 3×10^{-11}, and 3×10^{-11}.

The experiment was continued at a ^{48}Ca energy of 247 MeV for 70 d with a total beam dose of 2×10^{19}. The resulting excitation energy of the compound nucleus $^{297}117$ was about 35 MeV, favoring the 3 n reaction channel. A new decay chain was detected involving six consecutive α decays and ending in SF; see Fig. 1. In this chain, the great-granddaughter nucleus with $Z = 111$ did not undergo SF, but instead emitted an α particle with $E_{\alpha_4} = 9.00$ MeV. It was followed by at least two more α transitions and then, after about 33 h, the fission event was recorded. The total number for random sequences [15] imitating the observed decay chain amounts to 6×10^{-11}. Therefore, we assign this chain to the decay of the neighboring odd-odd nucleus $^{294}117$. Note that this decay chain was registered when about 30% of ^{249}Bk decayed to ^{249}Cf. Attributing it to the decay of $^{294}(118)$ nucleus is unlikely due to the small production yield and significantly different decay properties.

The decay properties of the neighboring isotopes $^{293}117$ and $^{294}117$, their daughters $^{289}115$ and $^{290}115$, as well as granddaughters $^{285}113$ and $^{286}113$, do not display substantial differences. These decay properties change significantly for the great-granddaughter nuclei. Despite the strong hindrance resulting in the relatively long half-life, SF is a principal decay mode of the odd-even nucleus $^{281}111$ (see Fig. 1). On the other hand, the heavier isotope $^{282}111$ undergoes α decay. The SF decay of $^{281}111$ can be explained by comparing the results of the present experiment with the properties of the neighboring even-Z nuclei. In the $T_{SF}(N)$ systematics, the decrease in the half-life with increasing neutron number in the region of nuclei with $N > 162$ changes to a strong increase in stability as N approaches the spherical shell at $N = 184$ [16]. Minimum values of T_{SF} are characteristic of the transition region $N = 168$–170 where the effect of nuclear shells is at a minimum. Indeed, the $Z = 110$ darmstadtium isotopes with $N = 169$ and $N = 171$, as well as the $Z = 112$, $N = 170$, and $N = 172$ copernicium isotopes, undergo SF rather than α decay [1]. For the odd-Z nuclei produced in the reactions ^{237}Np + ^{48}Ca and ^{243}Am + ^{48}Ca, the high hindrance of SF for nuclei with an odd number of protons and the relatively low T_α for the isotopes of elements 113 and 115 with $N = 169$–173 result in a preference for α decay [17,18]. Spontaneous fission is observed only in the isotopes of element 105, where the α decay half-life exceeds 10^5 s for ^{268}Db. In the reaction ^{249}Bk + ^{48}Ca, the daughter nuclei that originate from the evaporation residues $^{293}117$ and $^{294}117$ have one or two extra neutrons compared with those produced in the lower-Z reactions. A closer approach to the shell at $N = 184$ should result in a decrease in their decay energy Q_α and an increase in T_α with respect to the neighboring lighter isotopes at the same Z. This regularity is clearly observed experimentally for all the isotopes with $Z \geq 111$; see Fig. 3. In analogy with the neighboring even-Z isotopes, all the nuclei in the decay chains of $^{293}117$ and $^{294}117$ with $Z > 111$ and $N \geq 172$ are expected to undergo α decay. The nucleus $^{281}111$ ($N = 170$) lies in the "critical" region, and may avoid SF only because of the hindrance resulting from an odd proton. Despite a hindrance of 3×10^4 with respect to its even-even neighbor $^{282}112$ [1], the isotope $^{281}111$ undergoes SF with a probability $b_{SF} \geq 83\%$. Accordingly, even the high hindrance caused by the odd proton does not "save" the nucleus from SF because of the weakening of the stabilizing effect of neutron shells $N = 162$ and $N = 184$. The presence of an extra and unpaired neutron in the neighboring isotope $^{282}111$ further hinders SF relative to the α decay of this nucleus. In Figs. 3(a) and 3(b), the experimental values of decay energies Q_α and half-lives T_α are presented for isotopes with $Z = 111$, 113, 115, and 117. Increasing the neutron number in the heaviest nuclides results in a decrease of Q_α and a considerable increase in T_α. An especially strong growth of $T_\alpha(N)$ is observed for the isotopes of elements 111 and 113. Except for $^{281}111$, all the nuclides presented in Fig. 3 are α emitters; for them T_α

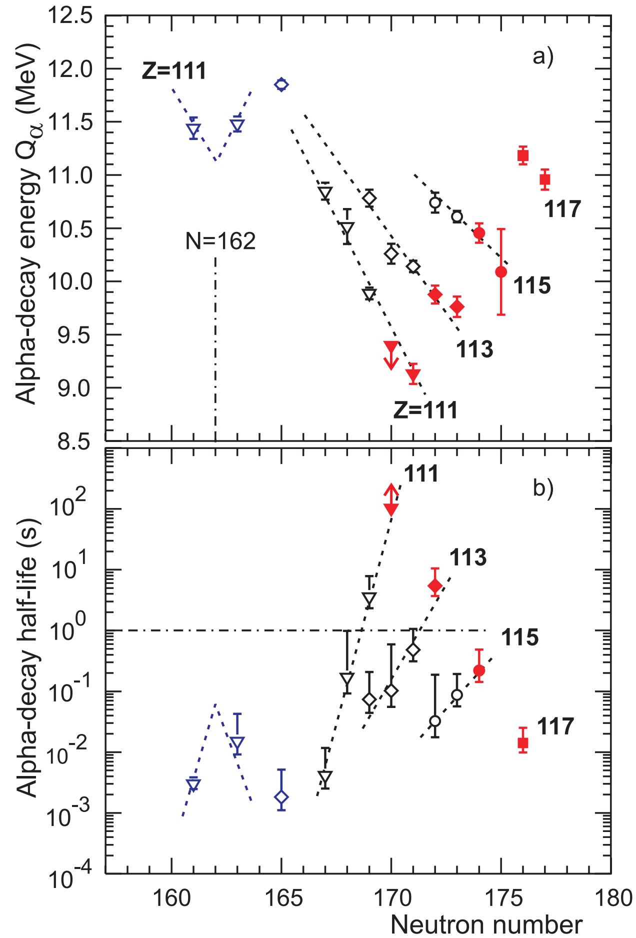

FIG. 3 (color). (a) α-decay energy and (b) half-lives versus neutron number for the isotopes of elements with $Z = 111$–117 (new results in red). All the nuclides with $N > 165$ have been produced in ^{48}Ca induced reactions. Our T_α (exp) values are given for the nuclei belonging to the ^{293}117 decay chain (5 events). The limit for $T_\alpha(^{281}\mathrm{Rg})$ was estimated from the measured half-life and number of observed nuclei.

is smaller than T_{SF}. This is another indication of the high stability of the superheavy nuclei with respect to SF. From the experimental and theoretical α-particle energies given in Fig. 1, it is obvious that for all the nuclei in the decay chains of the isotopes of element 117, the macroscopic-microscopic calculations of the masses of the superheavy nuclei [9] are in a good agreement with our experiment. The cross sections for producing the nuclei of element 117 in the reaction ^{249}Bk + ^{48}Ca are $\sigma = 0.5(+1.1, -0.4)$ pb and $\sigma = 1.3(+1.5, -0.6)$ pb at $E^* = 35$ MeV and $E^* = 39$ MeV, respectively. These values are similar to the results of previous experiments where cross sections for the reactions of ^{233,238}U, ^{237}Np, ^{242,244}Pu, ^{243}Am, ^{245,248}Cm, and ^{249}Cf targets with ^{48}Ca beams have been measured [1].

In conclusion, a new chemical element with atomic number 117 has been synthesized in the fusion of ^{249}Bk and ^{48}Ca. The data are consistent with the observation of two isotopes of element 117, with atomic masses 293 and 294. These isotopes undergo α decay with $E_\alpha = 11.03(8)$ and 10.81(10) MeV and half-lives 14(+11, −4) and 78(+370, −36) ms, respectively, giving rise to sequential α-decay chains ending in spontaneous fission of ^{281}Rg ($T_{\mathrm{SF}} \sim 26$ s) and ^{270}Db ($T_{\mathrm{SF}} \sim 1$ d), respectively. The decays of 11 identified isotopes substantially expand our knowledge of the properties of odd-Z nuclei in the region of the most neutron-rich isotopes of elements 105–117. These nuclei generally display a trend of increased stability with larger neutron number N. The longer half-lives offer the potential for investigation of the chemistry of superheavy elements and establishing their location in the periodic table. The new isotopes, together with superheavy nuclides previously synthesized in reactions with ^{48}Ca, present a consistent picture of nuclear properties in the area of heaviest nuclei. They demonstrate the critical role of nuclear shells and represent an experimental verification for the existence of the predicted island of stability for superheavy elements.

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