

Synthesis of the isotope $^{282}113$ in the $^{237}\text{Np} + ^{48}\text{Ca}$ fusion reaction

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The decay properties of the new isotope $^{282}113$ and its daughter nuclei have been measured in the $^{237}\text{Np}(^{48}\text{Ca}, 3n)^{282}113$ reaction. During an irradiation with a beam dose of 1.1×10^{19} 244-MeV ^{48}Ca projectiles, two decay chains originating from the odd-odd isotope $^{282}113$ ($E_\alpha = 10.63 \pm 0.08$ MeV, $T_\alpha = 73_{-29}^{+134}$ ms) were produced in the complete fusion reaction with a cross section of $0.9_{-0.6}^{+1.6}$ pb; these properties are all in agreement with expectations based on the results of previous experiments.

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This experiment was aimed at the synthesis of the new isotopes of element 113 produced as evaporation residues in the reaction $^{237}\text{Np} + ^{48}\text{Ca}$. In previous experiments, 17 new heavy nuclides with $Z = 112$ – 116 and 118 have been produced directly in the complete-fusion reactions of ^{238}U , $^{242,244}\text{Pu}$, ^{243}Am , $^{245,248}\text{Cm}$, and ^{249}Cf targets with ^{48}Ca beams (see Refs. [1–4] and references therein); we have also observed daughter nuclides arising in their radioactive decays. For three of the even- Z elements, four neighboring isotopes of each element were observed ($^{282-285}112$, $^{286-289}114$, and $^{290-293}116$). For the odd- Z elements, only two isotopes, $^{287,288}115$, $^{283,284}113$, and their decay products leading to $^{267,268}\text{Db}$, were synthesized in the reaction $^{243}\text{Am} + ^{48}\text{Ca}$ [3].

The synthesis of lighter isotopes of element 113 produced via the $3n$ - and $4n$ -reaction channels, $^{281,282}113$ ($N = 168, 169$) and their α -decay daughters, would provide information on the influence of closed neutron shells at $N = 162$ and $N = 184$ on the decay properties of nuclei in the intermediate region between these shells. According to different theoretical predictions (see, e.g., Refs. [5–8]) and the α -decay properties of lighter and heavier isotopes originating from the decays of $^{278}113$ [9] and $^{283,284}113$ [3], one would expect an increase in the α -particle energies of the isotopes $^{281,282}113$, $^{277,278}\text{Rg}$, and $^{273,274}\text{Mt}$ in comparison with heavier isotopes of these elements, and similar or somewhat lower α -decay energies for the isotopes $^{269,270}\text{Bh}$ compared to ^{272}Bh . The expected decay chains of $^{281,282}113$ should consist of at least three or four consecutive α decays followed by the spontaneous fission (SF) of the unknown daughter nuclei $^{265,266}\text{Db}$. However, one cannot exclude *a priori* an α -decay branch for $^{265,266}\text{Db}$ and $^{261,262}\text{Lr}$ [10,11], as well as their electron capture/ β^+ decay (ϵ) leading to the even- Z isotopes $^{265,266}\text{Rf}$ and $^{261,262}\text{No}$ [11] (see below). In the case of the α decay of isotopes $^{265,266}\text{Db}$ and $^{261,262}\text{Lr}$, their chains will lead to long-lived ^{257}Md ($b_\epsilon = 85\%$) \rightarrow ^{257}Fm ($T_{1/2} = 100$ d) or ^{258}Md ($T_{1/2} = 52$ d) [12,13]. In any case, the lifetimes of $^{265,266}\text{Db}$ ($N = 160, 161$) could be comparable with those of $^{267,268}\text{Db}$ ($N = 162, 163$), and would be much

longer than preceding α emitters, similar to ^{268}Db , which was observed as a descendant nucleus in the decay chain of the parent isotope $^{288}115$ [3,14].

The compound nucleus $^{285}113$ ($N = 172$), which is the product of the complete-fusion reaction $^{237}\text{Np} + ^{48}\text{Ca}$, possesses an intermediate number of neutrons in comparison with nuclei produced in the reactions $^{233}\text{U} + ^{48}\text{Ca}$ ($^{281}112$, $N = 169$), and $^{238}\text{U} + ^{48}\text{Ca}$ ($^{286}112$, $N = 174$). In the latter case, the cross section for the $3n$ -evaporation channel was found to be about 2 pb and only an upper limit of 0.6 pb was established for the cross sections of the $2-4n$ -reaction with the more neutron deficient target nucleus $^{233}\text{U} + ^{48}\text{Ca}$ [2]. There is a dependence of the fusion-evaporation reaction cross sections on the number of neutrons in the compound system (see Fig. 7 in Ref. [2]), so one would expect a maximum cross-section value of about 1 pb for the $3n$ -evaporation channel at an excitation energy of $^{285}113 E^* = 39$ MeV. According to theoretical estimates [15], the yields of the $2n$ - and $4n$ -reaction products are predicted to be noticeably lower.

Irradiation of the ^{237}Np target with ^{48}Ca projectiles was performed in June–July 2006. The experimental setup was the same as that used in our previous experiments [1–4]. The ^{48}Ca -ion beam was accelerated by the U400 cyclotron. The typical beam intensity at the target was 1.1 pμA. In a 31.5-d experiment a total beam dose of 1.1×10^{19} ^{48}Ca ions was accumulated. The beam energy was determined and controlled by employing a time-of-flight system with a systematic uncertainty of 1 MeV. The target consisted of the enriched isotope ^{237}Np ($> 99\%$, 0.37 mg/cm²) deposited as an oxide onto 1.5-μm Ti foils. The lab-frame projectile energy in the middle of the target layer was 244 MeV. An excitation-energy of the compound nuclei of 39.1 ± 2.2 MeV was calculated using the masses of [6,16] and taking the thickness of the target and the energy spread of the incident cyclotron beam into account.

The evaporation residues (ER) recoiling from the target were spatially separated in flight from ^{48}Ca beam ions, scattered particles and transfer-reaction products by the Dubna

Gas-Filled Recoil Separator [17] with an estimated transmission efficiency of approximately 40% for $Z = 113$ nuclei. ERs passed through a time-of-flight system (TOF) and were implanted in a 4-cm \times 12-cm semiconductor detector array with 12 vertical position-sensitive strips surrounded by eight 4-cm \times 4-cm side detectors. The position-averaged detection efficiency for full-energy α particles emitted in the decays of implanted nuclei was 87%. The energy resolutions were 60–120 keV (depending on the strip) for α particles absorbed in the focal-plane detector, 260–380 keV for α particles that escaped this detector with a low energy release and were subsequently registered by a side detector, and ≈ 1 MeV for α particles detected only by a side detector (without a focal-plane position signal). The measured fission-fragment energies were not corrected for the pulse-height defect of the detectors or for energy losses of escaping fragments in the detectors and the pentane gas filling the detection system. For most of the strips, the FWHM position resolutions of the signals of correlated decays of nuclei implanted in the detectors were 0.8–1.3 mm (2.4–3.8 mm for strips 6, 8, 11, 12) for ER- α signals and 0.5–1.1 mm (1.4–3.6 mm for strips 6, 8, 11, 12) for ER-SF signals.

For optimum detection of expected sequential decays of the daughter nuclides in the absence of a beam-associated background, the beam was switched off after a recoil signal was detected with the following parameters: implantation energy $E_{ER} = 7$ –18 MeV, expected for complete-fusion evaporation residues, followed by an α -like signal with an energy of $9.9 \text{ MeV} \leq E_{\alpha 1} \leq 11.4 \text{ MeV}$, in the same strip, within a 2.8-mm wide position window and a time interval of $\Delta t \leq 1.5$ s. If, during the first 10-s beam-off time interval, an α particle with $E_{\alpha 2} = 9.9$ –11.4 MeV was registered in any position of the same strip, the beam-off interval was automatically extended to 10 min. In the 10-min period, if other α particles with energies expected for heavy nuclei were observed, we could prolong the beam-off pause right up to the detection of an SF signal in the same strip.

In the $^{237}\text{Np} + ^{48}\text{Ca}$ experiment, approximately 1570 beam-off intervals occurred, for a total of 4.7 h. The spectrum of α -like signals (all events without a registered TOF signal) in all strips in the energy range $7 \leq E_{\alpha} \leq 11$ MeV is shown in Fig. 1. This figure also shows the α -particle spectrum detected during beam-off time intervals. In the high-energy part of the α -particle spectrum, where the decays of the daughter nuclei from the isotopes $^{281-283}113$, $^{277-279}\text{Rg}$, $^{273-275}\text{Mt}$, and $^{269-271}\text{Bh}$ ($E_{\alpha} = 8$ –11 MeV), are expected, 90 events were detected. Three of them, as will be shown below, belong to the decays of the daughter isotopes of $^{282}113$.

The measured parameters of the members of the two decay chains observed in this irradiation are presented in Fig. 2. In the first decay chain, implantation of a 13.99-MeV recoil in the center of strip 6 (24.1 mm from the top of the strip) of the focal-plane detector was followed 88.9 ms later by an α particle with $E_{\alpha 1} = 10.62$ MeV at a position of 25.3 mm. Detection of this sequence caused the beam to switch off, and all subsequent decays were detected in the absence of a beam-associated background. The second α particle was detected with an energy $E_{\alpha 2} = 10.69$ MeV 6.2 ms later. This event automatically prolonged the beam-off interval from

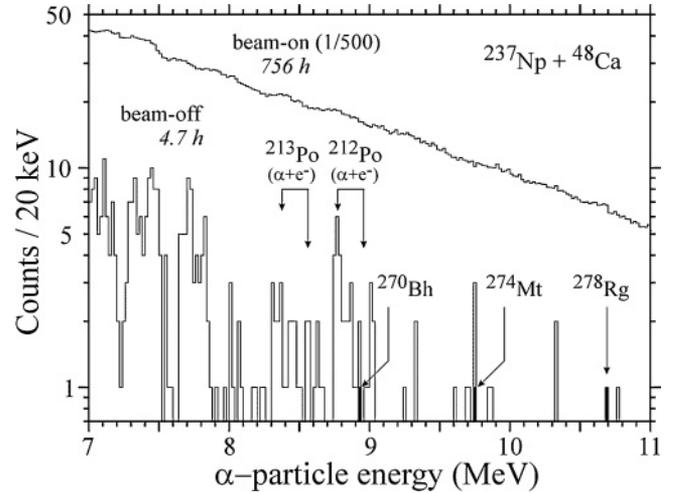


FIG. 1. Total beam-on and beam-off α -particle energy spectra of events registered by the focal-plane detector in the $^{237}\text{Np} + ^{48}\text{Ca}$ reaction. In the beam-off α -particle spectrum we observe the peaks originating from isotopes of Po, the decay products of long-lived isotopes of Ra-Th produced in transfer reactions, and ^{211}Po , the descendant nucleus of ^{219}Th produced in calibration reactions with a ^{nat}Yb target. The energies of events observed during beam-off periods in the correlated decay chains are shown by arrows (see Fig. 2).

10 s to 10 min. Then, in another 473 ms, the third α particle with $E_{\alpha 3} = 10.02 \pm 1.08$ MeV was registered by a side detector without an associated signal in the focal-plane detector. 87.98 s later, the last α decay with $E_{\alpha 4} = 8.93$ MeV was observed in the focal-plane detector. The beam-off interval was then prolonged further after this sequence of decays. The SF decay of the final nucleus in this chain was detected 31.74 min after the last α decay by both the focal-plane and side detectors with energies $E_{F1} = 136$ MeV (this signal corresponds to the total energy of one SF fragment moving deeper into the detector plus part of the energy of the second fragment escaping the focal-plane detector) and $E_{F2} = 38$ MeV (part of the energy of the second fragment registered by the side detector) ($E_{tot} = 174$ MeV). This event was the only SF event detected by both the focal-plane and side detectors in this experiment. During the beam-off period following the last α particle, no α particles with $E_{\alpha} > 7$ MeV were registered in the same position of strip 6 of the focal-plane detectors.

For this decay chain, the position deviations of the detected signals from the recoiling nucleus and its subsequent sequential decays (α and SF) are consistent with the position resolution of that particular strip detector (see Fig. 2). The position deviation of the α particle with $E_{\alpha} = 8.93$ MeV is within two ER- α position resolutions for strip 6. The correlated positions coupled with the short decay times indicate true, nonrandom correlations between the registered decays. Indeed, using rather wide windows around the decays observed in the experiment, the probabilities of detecting random beam-off α particles in the total energy range of $E_{\alpha} = 8$ –11 MeV and within 1 s are about 7×10^{-5} applying a position window of ± 3 mm for events registered by the focal-plane detector and 3×10^{-3} for α particles detected by the side detectors only. The same probability for SF events

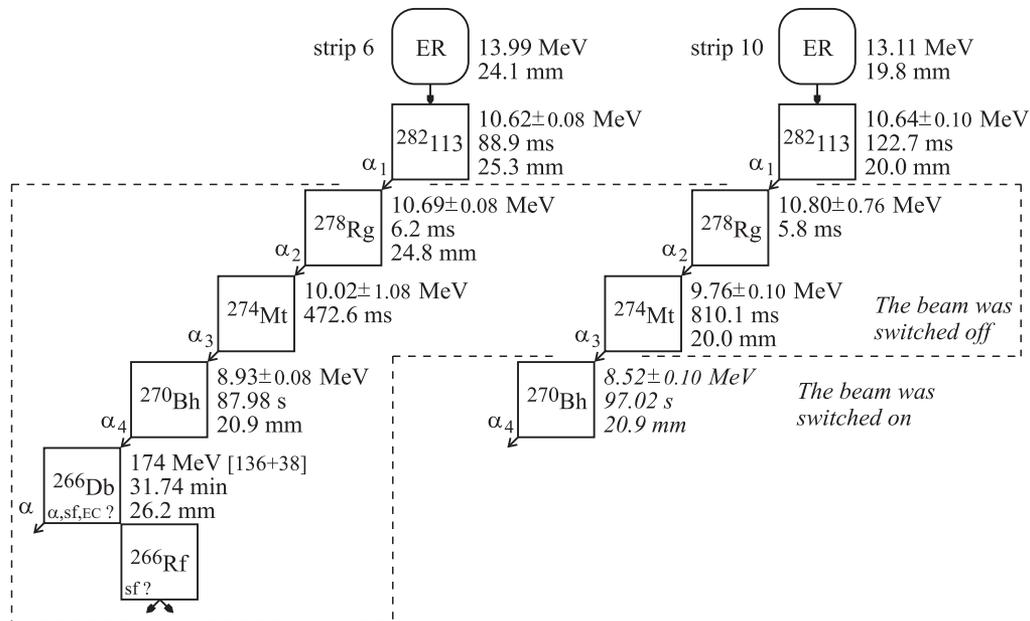


FIG. 2. Time sequences in the decay chains of $^{282}_{113}$ observed in the $^{237}\text{Np}+^{48}\text{Ca}$ reaction. The measured α -particle energies, decay times, and positions of the observed events with respect to the top of the focal-plane detectors are shown.

is about 5×10^{-9} per second. Thus, the number of random decay chains that would start from any of the ER- α pairs that switched the beam off followed by three more α decays and terminated by an SF event is less than 10^{-12} [18].

One more decay chain was registered in strip 10 (Fig. 2). Similar to the first decay chain, a recoil signal with $E_{\text{ER}} = 13.11$ MeV was followed by an α particle with an energy and decay time ($E_{\alpha_1} = 10.64$ MeV, $\Delta t = 122.7$ ms) that switched the beam off, and then two additional α particles were detected by the side detector only ($E_{\alpha_2} = 10.80 \pm 0.76$ MeV, $\Delta t = 5.8$ ms) and focal-plane detector ($E_{\alpha_3} = 9.76$ MeV, $\Delta t = 810$ ms), respectively. The initial conditions for prolonging the beam-off interval were not fulfilled (the second α did not generate a signal in the strip and the third α had an energy $E_{\alpha} < 9.9$ MeV, see above), the beam was switched on and the irradiation was continued. The number of random decay chains (ER- α_1 - α_2 - α_3) expected in this case is about 3×10^{-6} . The next α particle with an energy $E_{\alpha} = 8.52$ MeV (similar to the last one in the first decay chain) was found in the same strip 97 s later. However, one should note that the assignment of an α particle with $E_{\alpha} = 8.52$ MeV to the observed decay chain is somewhat tentative because the probability of detecting a similar signal of random origin in a 100-s time interval was about 0.4. No SF events were observed in strip 10 during the remainder of the experiment (about 10 d).

Based on the similar α -particle energies and decay times of the first three α transitions, we assign both decay chains to the same parent nucleus, namely $^{282}_{113}$ produced in the $^{237}\text{Np}(^{48}\text{Ca},3n)$ reaction (see Fig. 2). The production cross section calculated based on the two events is $\sigma_{3n} = 0.9^{+1.6}_{-0.6}$ pb, which is in agreement with expected value [15] for the $^{237}\text{Np}(^{48}\text{Ca},3n)^{282}_{113}$ reaction (see above) and with the

experimentally observed behavior of cross sections $\sigma_{xn}(Z_{\text{CN}}, N_{\text{CN}})$ measured in fusion-evaporation reactions of ^{238}U , $^{242,244}\text{Pu}$, ^{243}Am , $^{245,248}\text{Cm}$, and ^{249}Cf targets with ^{48}Ca ions (see, e.g., Fig. 5 in Ref. [2]).

The experimental α -decay energies, Q_{α}^{exp} , of the newly synthesized isotopes and previously known odd- Z nuclei with $Z \geq 101$ are plotted in Fig. 3. The α -decay energies attributed to the isotopes $^{282}_{113}$ and ^{278}Rg agree well with expected values resulting from the trend of the $Q_{\alpha}(N)$ systematics measured for the neighboring isotopes $^{278,283,284}_{113}$ and $^{274,279,280}\text{Rg}$. For the measured α -particle energies of the new isotopes $^{282}_{113}$ ($E_{\alpha} = 10.63 \pm 0.08$ MeV, $T_{1/2} = 73^{+134}_{-29}$ ms) and ^{278}Rg ($E_{\alpha} = 10.69 \pm 0.08$ MeV, $T_{1/2} = 4.2^{+7.5}_{-1.7}$ ms), one can estimate half-lives for *allowed transitions* using the formula of Viola and Seaborg [22]. The ratios between experimental (T_{exp}) and calculated (T_{calc}) half-lives (6 ms for $^{282}_{113}$ and 1 ms for ^{278}Rg) result in hindrance factors of 4–10 for the isotopes $^{282}_{113}$ and ^{278}Rg , similar to those estimated for the isotopes ^{280}Rg ($N = 169$) and ^{276}Mt ($N = 167$) (see Table V in Ref. [3]).

The measured T_{exp} values closely reproduce the calculated values for the heavier isotopes $^{283}_{113}$ and $^{284}_{113}$, as well as for their parent nuclei $^{287,288}_{115}$. A rise in the hindrance factor for the α decay of lighter nuclei ($^{282}_{113}$, $^{278-280}\text{Rg}$, etc.) can be caused by an increase of their deformation in the ground state (see also [1–4]). One can expect that the deformation of the heaviest nuclei increases with the reduction of the neutron number of the descendant nuclei, in agreement with macroscopic-microscopic calculations [5]. The quadrupole deformation parameter β_2 was calculated to be 0.066, 0.072 for $^{287,288}_{115}$ and 0.149, 0.138 for $^{283,284}_{113}$, and increases to 0.196, 0.207, 0.202, and 0.200 for $^{282}_{113}$ and $^{278-280}\text{Rg}$, respectively. Note also that changes of the α -decay energies in

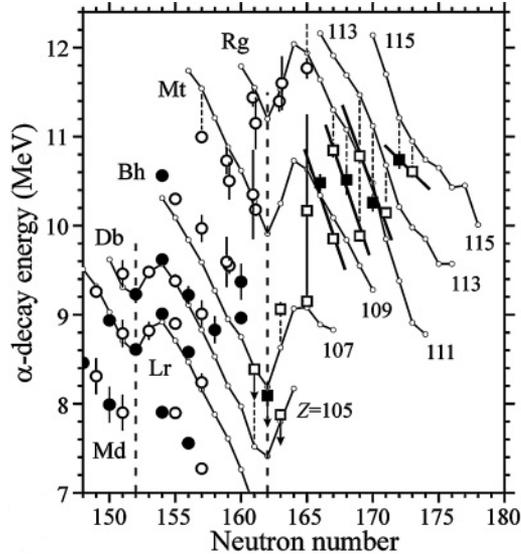


FIG. 3. α -decay energy vs. neutron number for odd- Z elements Md ($Z=101$) through element 115 (solid and open symbols—*even-N* and *odd-N* isotopes, respectively [9,13,16,19–21], squares—data produced in ^{48}Ca -induced reactions [3]). The values shown for $^{266,267,268}\text{Db}$ isotopes are upper limits, since these nuclides have not been observed to decay by α emission. Small open symbols connected by lines show the theoretical Q_α values calculated for the same isotopes in the MM model [4].

five consecutive α transitions $^{282}113 \rightarrow ^{278}\text{Rg} \rightarrow ^{274}\text{Mt} \rightarrow ^{270}\text{Bh} \rightarrow ^{266}\text{Db}$ (together with data for neighboring isotopes) demonstrate the influence of the neutron shell at $N=162$.

From the $Q_\alpha(N)$ data (Fig. 3) and theoretical predictions [5,16], one would expect the α -particle energy of ^{274}Mt to be about 10.3–10.6 MeV. The detected α -particle energy in the second chain (unfortunately, in the first chain this value was measured with poor precision) was less than expected by 0.5–0.8 MeV; this could have been caused by α decay of ^{274}Mt to excited states in the daughter nucleus ^{270}Bh (see, e.g., data for $^{283}112$, $^{289}114$ [2] or odd- Z isotopes ^{272}Rg , ^{268}Mt , and ^{264}Bh [19–21]). Thus, we propose the following decay properties for ^{274}Mt : $E_\alpha = 10.0 \pm 1.1$ and 9.76 ± 0.10 MeV, $T_{1/2} = 440^{+810}_{-170}$ ms.

For the last α decay observed in the first decay chain of $^{282}113$, the α -particle energy as well as half-life are in agreement with those expected for ^{270}Bh ($E_\alpha = 8.93 \pm 0.08$ MeV, $T_{1/2} = 61^{+292}_{-28}$ s, $T_{\text{calc}} = 5$ s), see Fig. 3.

The first decay chain was terminated by SF decay with an apparent life time of 31.7 min. The origin of this decay can be spontaneous fission of ^{266}Db , or its ε decay with a life time of 31.7 min followed by the relatively short-lived spontaneous fission of the even-even isotope ^{266}Rf ($T_{\text{SF}} = 23$ s as predicted in Ref. [23]).

In the second decay chain presented in Fig. 2, the SF event was not observed. One cannot entirely exclude the possibility that the SF event was lost during registration of any preceding signal within a time interval of 85 μs (dead time of the detection system). From the average counting rate of beam-on events, the probability of missing a SF is about

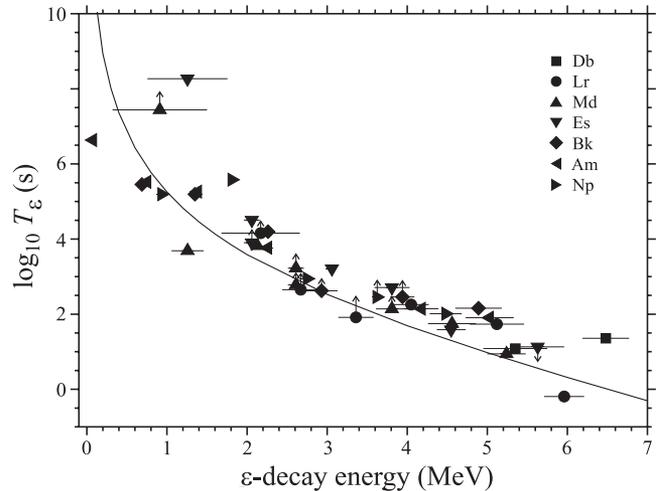


FIG. 4. Partial ε/e^- -decay half-life vs. decay energy for odd-odd isotopes of elements Np–Db [13]. The solid line shows the fit for odd-odd nuclei [24].

2% in this experiment. However, the nonobservation of a SF event in this case could arise in the decay properties of ^{266}Db .

One cannot exclude *a priori* three different decay modes of ^{266}Db , namely, spontaneous fission, α or ε decays. Using experimental data for the lighter isotopes $^{262,263}\text{Db}$ [13] along with the dependence of partial SF half-lives predicted for neighboring even-even nuclei [23], one would expect an SF half-life for ^{266}Db exceeding a few hours. For the ε -decay energy of this isotope one can take a value of 2.86 ± 0.65 MeV [16], which agrees with calculations [5–7], and results in a partial half-life of 20 min (with a factor of 10 uncertainty, see Fig. 4), which is comparable with a decay time of 31.7 min. Thus, the last event in the first chain can originate from SF decay of ^{266}Db or its daughter nucleus ^{266}Rf .

Along with SF and ε decay, the isotope ^{266}Db can undergo α decay. The α -decay energy of $Q_\alpha = 8.19 \pm 0.30$ MeV suggested for ^{266}Db in Ref. [16] is in reasonable agreement with the $Q_\alpha^{\text{exp}}(N)$ data shown in Fig. 3; therefore, one would expect a partial α -decay half-life of ~ 1 –100 min for the allowed transition of this isotope. Nonobservation of the α particle due to the decay of ^{266}Db in the first decay chain, which was observed completely during a beam-off period, could be explained by its nonregistration by the detectors. In the two decay chains, seven α particles were registered in all with a detector that has an 87% efficiency for the detection of α particles, so the loss of one α particle seems rather probable. In that case, the spontaneous fission in the first chain belongs to the known isotope ^{262}No ($T_{\text{SF}} = 5$ ms) that follows ε decay of ^{262}Lr ($T_\varepsilon = 3.6$ h) [10,11].

However, an SF event was not registered in the second chain within ten days following the ER. The nonobservation of an SF event could be caused by a possible α -decay branch for ^{262}Lr that leads to the long-lived isotope ^{258}Md ($T_{1/2} = 51.5$ d, $b_{\text{SF}} < 3 \times 10^{-3}$ [12]) with a half-life exceeding the total time of the experiment.

The isotope ^{262}Lr undergoes mainly ε decay (a 20% α -decay branch is reported as an upper limit by the authors

of the original paper [10,11]). However, given the estimated value $Q_\alpha = 8.01 \pm 0.20$ MeV [16], one would expect a partial half-life of about 10 min for an allowed α decay of this isotope. Comparison of this half-life with the experimentally measured value $T_{1/2} = 3.6$ h [10,11] can indicate a perceptible probability for the α decay of ^{262}Lr , which would explain the missing of a SF event in the second decay chain.

Because of the low number of observed decays of the parent isotope $^{282}\text{113}$, the decay modes of nuclei at the end of the α -decay chain of $^{282}\text{113}$, discussed above, should be considered somewhat speculative, but do explain the results of the experiment. Unfortunately, the production of more parent nuclei in the $^{237}\text{Np} + ^{48}\text{Ca}$ reaction is limited by the low production cross section for evaporation residues. The $^{241}\text{Am}(^{48}\text{Ca}, 3n)^{286}\text{115} \rightarrow ^{282}\text{113}$ reaction can be preferable for the investigation of light neighboring isotopes of elements 115 and 113.

In the $^{237}\text{Np} + ^{48}\text{Ca}$ reaction, two decay chains originating from the new isotope $^{282}\text{113}$ were observed for the first time. The production cross section of the $3n$ -evaporation channel of the reaction was measured to be about 1 pb.

Atomic and mass numbers of the isotope of element 113 produced in this reaction were determined through a comparison of the decay characteristics of the observed nuclei

with radioactive properties of neighboring isotopes $^{283}\text{113}$ and $^{284}\text{113}$, produced earlier in the $^{243}\text{Am} + ^{48}\text{Ca}$ reaction [3], guided by comparison with theoretical predictions.

The production cross section of element 113 in the reaction $^{237}\text{Np} + ^{48}\text{Ca}$ exceeds that of the lighter isotope of element 113 produced in the cold fusion reaction $^{209}\text{Bi} + ^{70}\text{Zn}$ by more than one order of magnitude [9]. The decay properties of five new isotopes with $Z = 105\text{--}113$ are in agreement with the modern macroscopic-microscopic models of nuclei.

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