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THE SYNTHESIS OF SUPERHEAVY NUCLEI IN THE ^{48}Ca + ^{244}Pu REACTION: ^{288}114

In the bombardment of a ^{244}Pu target with ^{48}Ca ions, we observed two identical decay sequences of genetically linked events, each consisting of an implanted heavy atom, two subsequent α-decays, and terminated by a spontaneous fission. The measured α-energies and corresponding half-lives of the sequential chain members were: E_α=9.84 MeV ($T_{1/2}$=1.9 s) and 9.17 MeV ($T_{1/2}$=9.8 s); for the spontaneous fission ($T_{1/2}$=7.5 s), the total energies deposited in the detector array were 213 and 221 MeV. The decay properties of the synthesized nuclei are consistent with the consecutive α-decays originating from the parent even-even nucleus ^{288}114, produced in the $4n$-evaporation channel with a cross section of about 1 pb. ^{288}114 and ^{284}112 are the heaviest known α-decaying even-even nuclides, following the production of ^{260}Sg and ^{266}Sg (Z=106) and the observation of one α-decay of ^{264}Hs (Z=108). The observed radioactive properties of ^{288}114 and the daughter nuclides match the decay scenario predicted by the macroscopic-microscopic theory.

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During the last year the first convincing evidence for the synthesis of superheavy nuclei around the theoretically predicted spherical shell closures at Z=114 and N=184 was presented [1-4].

These reports included the results of our experiment, in which a ^{244}Pu target was bombarded by ^{48}Ca ions with beam dose of 5.2×10^{18} during September-December, 1998, employing the Dubna Gas-filled Recoil Separator. We observed a decay sequence consisting of an implanted heavy atom, three subsequent α-decays (E_α = 9.71 MeV, 8.67 MeV and 8.83 MeV), and a spontaneous fission (SF), all correlated in time and position [2]. The most reasonable explanation for this decay chain is consecutive α-decays starting from the parent nucleus ^{289}114, produced in the 3n-evaporation channel.

In March-April, 1999, a ^{242}Pu target was bombarded with ^{48}Ca ions (beam dose 7.5×10^{18}) at the separator VASSILISSA. Two decay chains were assigned to the α-decay of the parent nucleus ^{287}114, with $T_{1/2}$ = 5.5 s and E_α = 10.29 MeV [3]. Both decay chains were terminated after the first α-decays by spontaneous fission of the daughter nucleus ^{283}112, a nuclide previously observed in the ^{238}U + ^{48}Ca reaction [1].

The synthesis of ^{293}118 and its sequential α-particle emission to the daughter isotopes with Z=116-106 in the bombardment of ^{208}Pb with 2.3×10^{18} 449-MeV ^{86}Kr ions using the Berkeley separator BGS was announced in April-May, 1999. Three decay chains were observed, each consisting of an implanted atom and six subsequent α-decays. Another experiment with this reaction was carried out at the same bombarding energy at GSI, in Darmstadt, using the separator SHIP. No correlated α-decay chains were observed yet, with a similar beam dose of 2.9×10^{18} Kr ions [5].

In June-October, 1999, we continued the bombardment of a ^{244}Pu target with ^{48}Ca projectiles. Most of the details of the experiment are given in our previous publication [2]. As before, the rotating target consisted of 9 separate sectors made of the enriched isotope ^{244}Pu (98.6%) in the form of PuO$_2$, deposited onto 1.5-μm Ti foils to a thickness of ~0.37 mg cm^{-2}. We used a ^{48}Ca bombarding energy of ~236 MeV in the middle of the target. Taking into account the energy loss in the target (~3.4 MeV), the small variation of thickness of individual target sectors, the beam energy resolution and the variation of the beam energy during the long-term irradiation, we estimated the excitation energy of the compound nucleus ^{292}114 to be in the range of 31.5-39 MeV [6], which corresponds to the evaporation of 3 or 4 neutrons.

In the present series of experiments we used an upgraded data acquisition system. This allowed us to determine the individual target sector which gave rise to each detected recoil atom, as well as the ^{48}Ca bombarding energy at this time. Thus a narrower range of excitation energies could be assigned to each particular recoiling compound nucleus.

A typical beam intensity on target was 4×10^{12} pps. Over a period of 60 days a total of 1.0×10^{19} projectiles was delivered to the target.

Evaporation residues (EVR) recoiling from the target were separated in flight from beam particles, scattered ions and various transfer-reaction products by the Dubna Gas-filled Recoil Separator [7], passed through a time-of-flight (TOF) detector, and were implanted in the focal-plane detectors, consisting of twelve 4-cm-high×1-cm-wide

position-sensitive strips. To detect α's escaping the focal-plane detector, eight side detectors of the same type without position sensitivity were arranged in a box surrounding the focal-plane detector. The total α-particle detection efficiency was ~87% of 4π. A set of 3 similar "veto" detectors was situated behind the focal-plane detectors in order to eliminate signals from low-ionizing light particles, which could pass through the focal-plane detector (300μm) without being detected in the TOF system. At a beam intensity of 4×10^{12} pps, the overall counting rate of the detector system was ~15 s^{-1}. We estimate that 40% of the recoiling Z=114 nuclei formed in the ^{244}Pu target would be implanted in the focal-plane detector.

Alpha-energy calibrations were periodically performed using the α peaks from nuclides produced in the test reaction ^{206}Pb + ^{48}Ca. The fission-energy calibration was obtained by detecting fission fragments from the SF of ^{252}No. The energy resolution for detection of α-particles in the focal-plane detector was ~50 keV; for detection by the side detectors of α's escaping from the focal-plane detector, the energy resolution was ~190 keV. We determined the position resolution of the signals of correlated decays of nuclei implanted in the detectors: For sequential α-α decays the FWHM position resolution was 1.1 mm; for correlated EVR-α signals, 0.8 mm; and for correlated EVR-SF signals, 0.5 mm.

According to experimental data obtained in the previous experiments [1-3] and the predicted decay properties of the nuclei, the 2-to-4 neutron evaporation products from the compound nucleus ^{292}114 [8,9] should have α-decay chains terminated by spontaneous fission.

We observed only two fission events in the present ^{244}Pu + ^{48}Ca bombardment. Both SF events were observed as two coincident signals (two fission fragments) with energies E_{F1}=156+65 MeV and E_{F2}=171+42 MeV. The items in each sum are the energies deposited in the focal-plane and side detectors, respectively. We searched the data backwards in time from these events for preceding α particles with E_α>8 MeV [8,9] and/or EVRs, in the same positions. The latter were defined as events characterized by the measured energies, TOF signals and estimated resulting mass values, that were consistent with those expected for a complete-fusion EVR, as determined in the calibration reaction. The full decay chains including these two fission events are shown in Fig. 1, with the suggested assignment of decays to specific nuclei.
Both decay chains are consistent with one another, in all the measured parameters. The first α-particles have similar energies E_α=9.87 MeV and E_α=9.80 MeV, and were detected in the focal-plane detector 0.77 s and 4.58 s after the implantation of the recoil nuclei in strips 2 and 8, respectively. The second α-particles in corresponding chains, having the energies E_α=9.21 MeV and E_α=9.13 MeV, were observed at the same locations after 10.34 s and 18.01 s.
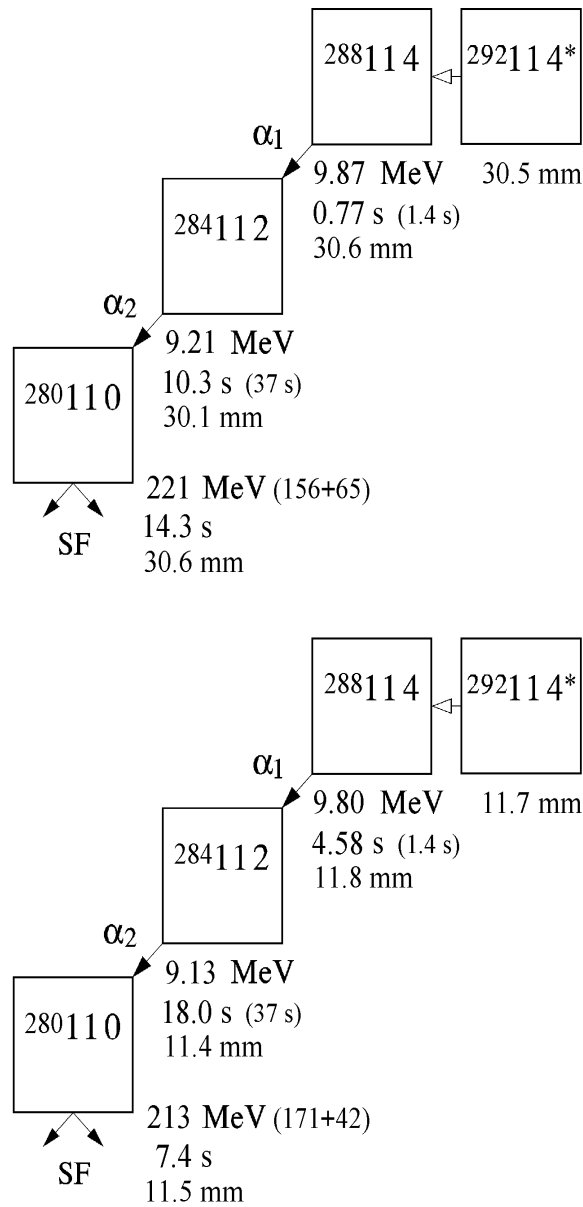
288114   292114*

α_1

9.87 MeV   30.5 mm
0.77 s (1.4 s)
30.6 mm

284112

α_2

9.21 MeV
10.3 s (37 s)
30.1 mm

280110

221 MeV (156+65)
14.3 s
30.6 mm

SF

288114   292114*

α_1

9.80 MeV   11.7 mm
4.58 s (1.4 s)
11.8 mm

284112

α_2

9.13 MeV
18.0 s (37 s)
11.4 mm

280110

213 MeV (171+42)
7.4 s
11.5 mm

SF

Fig. 1. *Time sequences in the observed decay chains. The expected half-lives corresponding to the measured E_α values for the given isotopes are shown in parentheses following the measured lifetimes. Positions of the observed decay events are given with respect to the top of the strip.*

Finally, 14.26 s and 7.44 s later, the SF events were observed. All events in the two decay chains appeared within time intervals of 25.4 s and 30.0 s and position intervals of 0.5 mm and 0.4 mm (Fig. 1), respectively.

To estimate the probability of a random origin of the candidate decay chains we used two approaches. By applying a Monte Carlo technique [2,10] we determined the probability per fission of finding such correlated events at any position of the detector array. Also, following the procedure described in Ref. [11], we calculated probabilities of these decay sequences being caused by the chance correlations of unrelated events at the positions in which the candidate events occurred. The results from the two methods were similar; the probability that both decay chains consist of random events is less than 5×10^{-13}, calculated in the most conservative approach.

We observed two three-member decay sequences. If we assume that they actually consisted of four decays (i.e., the spontaneous fission was due to element 108), the probability of missing one α-event in both decay chains would be less than 3%.

The ^{48}Ca beam energy in the middle of the target corresponded to an excitation energy of 36-37 MeV of the compound nuclei in both cases, expected for evaporation of 4 neutrons from the ^{292}114 compound nucleus. We should also note that the observed chains, including two alpha-decays and terminated by SF, match the decay scenario predicted for the even-even nuclide ^{288}114 [8,9]. For all the chain members, the basic rule of α-decay, which defines the relation between Q_α and T_α of the even-even nuclides with Z=114 and 112, is fulfilled. To illustrate this, Fig. 1 presents the expected half-lives corresponding to the measured α-particle energies for the genetically related nuclides with the specified atomic numbers. For the calculation of half-lives, the formula by Viola and Seaborg has been used, with parameters fitted to the T_α values of 58 even-even nuclei with Z>82 and N>126, for which both T_α and Q_α were measured [8,9]. Conversely, substituting experimental $T_{1/2}$ and E_α values in this formula results in atomic numbers of $114.4^{+1.6}_{-0.8}$ and $110.2^{+1.5}_{-0.8}$ for the mother and daughter nuclides, respectively. The measured total energies deposited in the detector array for both fission events exceed the average value measured for ^{252}No by about 40 MeV. Despite the relatively wide distributions of the total kinetic energies in spontaneous fission, this also indicates the fission of a rather heavy granddaughter nucleus, with Z>106 [12].

From the above considerations, we can conclude that the detected decay chains originate from the parent even-even nuclide ^{288}114, produced in the ^{244}Pu + ^{48}Ca reaction via the 4n-evaporation channel. On the basis of two detected events, the cross section of this reaction is about 1 pb.

Experiments with the ^{244}Pu + ^{48}Ca reaction were performed under essentially the same experimental conditions as reported in [2], with a resulting integral beam dose of 1.5×10^{19} ions. From the experimental dependencies of the cross sections of symmetric fission induced by ^{48}Ca ions on the ^{238}U and ^{244}Pu targets [12] and recent calculations [13], evaporation of 3 or 4 neutrons from the compound nucleus ^{292}114 could be expected with similar probabilities, in the investigated excitation energy range of 31.5-39 MeV. In this respect, the observation of the two neighboring isotopes of element 114 in the present series of experiments is to be expected.

The new decay sequences evidently originate from a different parent nucleus than the single chain that was observed previously in the reaction ^{244}Pu + ^{48}Ca and attributed to the decay of ^{289}114 [2]. The new alpha-decaying nuclides are characterized

by higher decay energies than the corresponding members of the chain observed in [2], while SF terminates the decay sequence at an earlier stage. Thus, comparison of the decay properties gives partial support for the assignment of the previously observed longer sequence to the decay of a heavier odd-mass nuclide [289]114 [2]. We plan to continue our experiment using a ~5 MeV lower projectile energy to search for additional decays of [289]114.

Note that [288]114 and [284]112 are the heaviest known α-decaying even-even nuclides, following the production of [260,266]Sg (Z=106) [14,15] and the observation of one α-decay of [264]Hs (Z=108) [16].

The radioactive properties of the new observed nuclides are in qualitative agreement with predictions of the macroscopic-microscopic nuclear theory [8,9]. The properties of the new nuclides also agree with those that could be expected by comparison with those of previously synthesized neighboring odd isotopes [287,289]114 [2,3] and [285]112 [2]. The latter point is demonstrated in Fig. 2, in which the alpha-decay energies of known isotopes of even-Z elements with $Z \geq 100$ together with theoretical Q_α values [8,9] for even-even nuclides with $Z=106$-114 are shown.
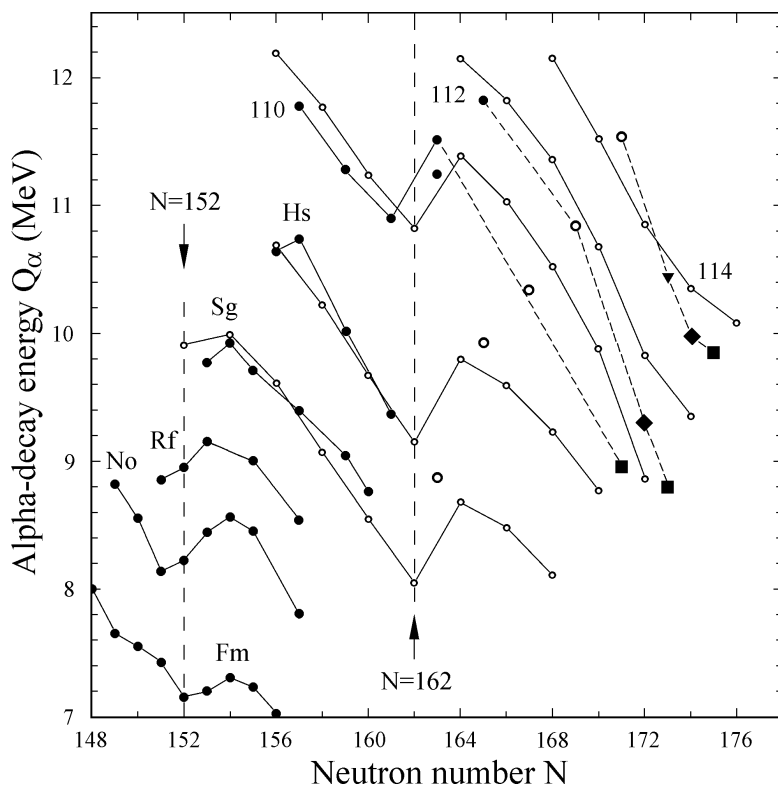

Fig. 2. *Alpha-decay energy vs. neutron number for isotopes of even-Z elements with $Z \geq 100$ (solid circles) [14-19]. Open circles show data from Ref. [4]; triangles, from Ref. [3]; solid squares, from Ref. [2]; and diamonds, data from the present work. Open circles connected with solid lines show theoretical Q_α values [8,9] for even-even $Z=106$-114 isotopes.*

Comparison of the measured decay properties of the new even-even superheavy nuclei $^{288}114$ (E_α=9.84±0.05 MeV, $T_{1/2}$=1.9$^{+3.3}_{-0.8}$ s), $^{284}112$ (E_α=9.17±0.05 MeV, $T_{1/2}$=9.8$^{+17.9}_{-3.8}$ s), and $^{280}110$ ($T_{1/2}$=7.5$^{+13.7}_{-2.9}$ s) with theoretical calculations [8,9] indicates that nuclei in the vicinity of spherical shell closures with Z=114 and N=184 could be even more stable than is predicted by recent theory. It can be seen in Fig. 2 that alpha-decay energies of the heaviest new even-even nuclides with Z=112 and 114 are 0.4-0.5 MeV less than the corresponding predicted values. The heaviest even-odd nuclides follow this trend as well. Such a decrease in Q_α values leads to an increase of partial α-decay lifetimes by an order of magnitude. Calculations are far less definite regarding spontaneous fission; however, we note that the observed spontaneous fission half-life of $^{280}110$ exceeds the predicted value [8] by more than two orders of magnitude.

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