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THE SYNTHESIS OF SUPERHEAVY NUCLEI IN THE $^{48}Ca + ^{244}Pu$ REACTION

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Синтез сверхтяжелых ядер в реакции $^{48}Ca + ^{244}Pu$

При облучении мишени из ^{244}Pu ионами ^{48}Ca был наблюден распад атома, имплантированного в детектор, состоящий из трех последовательных α-распадов и спонтанного деления. Все звенья цепочки распадов коррелированы во времени и позиции. Измеренные энергии α-частиц и соответствующие временные интервалы составляют: $E_\alpha = 9{,}71$ МэВ ($\Delta t = 30{,}4$ с), 8,67 МэВ ($\Delta t = 15{,}4$ мин) и 8,83 МэВ ($\Delta t = 1{,}6$ мин); для спонтанного деления ($\Delta t = 16{,}5$ мин) полное энерговыделение составляет около 190 МэВ. Большие энергии α-частиц и длинные времена распадов, а также прерывание цепочки спонтанным делением свидетельствуют о распаде ядер с большими атомными номерами. Данная цепочка является хорошим кандидатом на α-распад материнского ядра $^{289}114$, образующегося в канале с испарением трех нейтронов с сечением около 1 пб. Значительное повышение времен жизни новых ядер с $Z = 112$ и 110 — дочерних продуктов распада ядра с $Z = 114$ (фактор $\sim 10^6$) относительно известных наиболее тяжелых изотопов элементов 112 и 110, можно рассматривать как прямое доказательство существования «острова стабильности» сверхтяжелых элементов.

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The Synthesis of Superheavy Nuclei in the $^{48}Ca + ^{244}Pu$ Reaction

In the bombardment of a ^{244}Pu target with ^{48}Ca ions, we observed a decay sequence consisting of an implanted heavy atom, three subsequent α-decays, and a spontaneous fission, all correlated in time and position. The measured α-energies and corresponding time intervals were: $E_\alpha = 9.71$ MeV ($\Delta t = 30.4$ s), 8.67 MeV ($\Delta t = 15.4$ min) and 8.83 MeV ($\Delta t = 1.6$ min); for the spontaneous fission ($\Delta t = 16.5$ min) the total deposited energy was approximately 190 MeV. The large alpha-particle energies together with the long decay times and spontaneous fission terminating the chain offer evidence of the decay of nuclei with high atomic numbers. This decay chain is a good candidate for originating from the α-decay of the parent nucleus $^{289}114$, produced in the 3n-evaporation channel with a cross section of about 1 pb. The significant increase in the lifetimes of the new $Z = 112$ and 110 daughters of the $Z = 114$ nuclide (by a factor of $\sim 10^6$) with respect to the known heaviest isotopes of elements 112 and 110 can be considered direct proof of the existence of the «island of stability» of superheavy elements.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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Introduction

A fundamental outcome of macro-microscopic nuclear theory is the prediction of an "island of stability" of superheavy elements. Calculations performed over more than 35 years with different versions of the nuclear shell model predict substantial enhancement of the stability of heavy nuclei when approaching the closed spherical shells Z=114 and N=184. The isotopes of element 114 with neutron numbers from 174 to 176, close to the predicted spherical nuclei and, consequently, being relatively stable, can be produced in the fusion reaction of ^{244}Pu with ^{48}Ca ions [1].

With the doubly-magic ^{48}Ca projectile, the resulting compound nucleus ^{292}114 is weakly excited; its excitation energy at the Coulomb barrier is about 30 MeV. Correspondingly, nuclear shell effects are still expected to persist in the excited nucleus, increasing the survival probability of the evaporation residues (EVRs), as compared to "hot fusion" reactions ($E^* \approx 45$ MeV), which were used for the synthesis of heavy isotopes of elements with atomic numbers Z=106, 108 and 110 [2]. Additionally, the high mass asymmetry in the entrance channel should decrease the dynamical limitations on nuclear fusion arising in more symmetrical reactions.

In spite of these advantages, previous attempts to synthesize new elements in ^{48}Ca-induced reactions with actinide targets gave only upper cross section limits for their production [3]. The first positive result was obtained in 1998 when two spontaneous fission (SF) events were observed in the ^{48}Ca+^{238}U reaction with an integrated beam dose of 3.5×10^{18} ions. These were assigned to the decay of a new isotope of element 112 produced in the reaction ^{238}U(^{48}Ca,3n)^{283}112 (N=171) with a cross section of 5±2 pb [4].

In the ^{48}Ca+^{244}Pu reaction the ^{292}114 compound nuclei are expected to deexcite by emission of 2 to 4 neutrons. From calculations by Pustylnik, based on the experimental cross sections of "hot fusion" reactions, and diffusion model calculations by Wada [5] the maximum cross section for producing evaporation residues in the ^{48}Ca+^{244}Pu reaction is expected to be the 3n-evaporation channel at an excitation energy of 35 MeV. The absolute cross sections of the xn-channels are estimated with larger uncertainty; the calculated cross section at the maximum of the 3n-channel varies from 1 to 10 pb.

Smolanczuk *et al.* [6,7], who successfully reproduce the decay properties of the most neutron-rich known heavy nuclei, have calculated that the even-even isotopes ^{288}114 and ^{290}114 will have partial α-decay half-lives T_α=0.14 s and 0.7 s, respectively. Their predicted SF half-lives are considerably longer: $T_{SF}=2\times10^3$ s and 4×10^5 s, respectively. For their daughter nuclei - isotopes of element 112 - the values of T_α and T_{SF} are more comparable, but α-decay should still prevail. The α-decay granddaughters - the isotopes of element 110 - are expected to decay primarily by spontaneous fission. For the odd isotopes, in particular for the $^{289}114$ nucleus, the predictions are less definite; the odd neutron can lead to hindrance of α-decay and, especially, spontaneous fission. Here one expects competition between the two decay modes in the daughter products with $Z \leq 112$ and somewhat longer chains of sequential α-decays than in the case of the neighboring even-even isotopes.

We note that T_α calculations by Möller *et al.* [8] for $^{288-290}114$ give values exceeding those of [6,7] by orders of magnitude (e.g., T_α of 7×10^4 s for $^{289}114$). This, however, does not change the expected decay pattern for these isotopes of element 114 and their daughters. We can expect a sequence of two or more α-decays terminated by a spontaneous fission as it recedes from the stability region around N=184.

We present here initial results of an experiment to synthesize nuclei with Z=114 in the vicinity of predicted spherical nuclear shells in the complete fusion reaction $^{48}Ca+^{244}Pu$.

Experiment

A $^{48}Ca^{+5}$ beam was extracted from the ECR ion source and injected into the Dubna U400 heavy ion cyclotron. The average intensity of the ion beam on the target was 4×10^{12} pps at the material consumption rate of about 0.3 mg h^{-1}. The beam energy was verified with a precision of ~1 MeV, by measuring the energies of scattered ions, and by a time-of-flight technique.

The targets consisted 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}. Each target had an area of 3.5 cm^2 in the shape of an arc segment with an angular extension of 40° and an average radius of 60 mm. Nine targets were mounted on a disk that was rotated at 2000 rpm across the beam direction.

We used a ^{48}Ca bombarding energy of 236 MeV, corresponding to the calculated maximum of the 3n-evaporation channel to form the isotope $^{289}114$. EVRs recoiling from the target were separated in flight from beam particles and various transfer-reaction products by the Dubna Gas-filled Recoil Separator [9], passed through a time-of-flight (TOF) system, and were implanted in the focal-plane detectors. At a beam intensity of 4×10^{12} pps, the overall counting rate of the detector system was 15 s^{-1}. The collection efficiency of the separator was estimated from the results of test experiments in the bombardment of ^{nat}Yb and enriched $^{204,206-208}Pb$ targets with ^{48}Ca ions. We deduce that 40% of the recoiling Z=114 nuclei formed in the ^{244}Pu target would be implanted in the focal plane.

The TOF detector was used to measure the time of flight of recoiling nuclei (detection efficiency of ~99.7%) and to distinguish the focal-plane detector signals of particles passing through the separator from those of the radioactive decay of previously implanted nuclei. The focal-plane detector consisted of three 40×40 mm^2 silicon *Canberra Semiconductor* detectors, each with four 40-mm-high×10-mm-wide strips having position sensitivity in the vertical direction. To increase the detection efficiency for α's escaping the focal-plane detector, we arranged 8 detectors of the same type without position sensitivity in a box surrounding the focal-plane detector. Employing these side detectors increased the α-particle detection efficiency to ~87% of 4π. A set of 3 similar "veto" detectors was situated behind the front detectors in order to eliminate signals from low-ionizing light particles, which sometimes pass through the focal-plane detector without being detected in the TOF system. Alpha-energy calibrations were periodically performed using the α peaks from nuclides produced in the test reactions mentioned above. The fission-energy calibration was obtained by detecting fission fragments from the SF of ^{252}No [10]. The energy resolution for detection of α-particles in the focal-plane detector was ≈45 keV; for detection by the side detectors of α's escaping from the focal plane, the energy resolution was ≈180 keV. We determined the position resolution of the signals of correlated decays of nuclei implanted in the detectors: For sequential α-α decays the position resolution (at 95% confidence level) was ±0.8 mm; for correlated EVR-α signals, ±1.2 mm; and for correlated EVR-SF signals, ±1.1 mm.

Results

The experiment was performed during November and December, 1998. Over a period of 40 days a total of 5.2×10^{18} projectiles was delivered to the target.

In the analysis of the experimental data, we assumed that the "island of stability" of superheavy nuclides has a border at which nuclei are unstable against spontaneous fission. As long as any α-decay chain leads to the edge of the "island of stability", it should be terminated by spontaneous fission. In test experiments, we observed that 95% of SF events from the ^{252}No implants produced in the $^{206}Pb+^{48}Ca$ reaction are characterized by total deposited energy exceeding 130 MeV (without corrections for the pulse-height defect). Four such events were observed in the $^{244}Pu+^{48}Ca$ bombardment.

Two events, with measured energies E=149 MeV and E=153 MeV, were detected 1.13 ms and 1.07 ms, respectively, after the implantation of corresponding position-correlated recoil nuclei. For one of these SF events both fission fragments were registered by the focal-plane and side detectors. We assign these events to the spontaneous fission of the 0.9-ms $^{244mf}Am$ isomer, a product of transfer reactions with the ^{244}Pu target. Another signal was registered at the edge of the sensitive layer of the outermost detector strip and since its analysis is not straightforward, we have deferred it to a later time.

The last SF event was observed as two coincident signals (two fission fragments) with energy deposited in the focal-plane detector E_{F1}=120 MeV and in the side detector E_{F2}=52 MeV; E_{tot}=172 MeV. Correcting for pulse-height defect using calibration data mentioned above, this would mean a total deposited energy of

190 MeV. We searched the data backwards in time from this event for preceding α particles in the same position with E_α>8 MeV [6-8]. The entire position-correlated decay chain is shown in Fig.1a. An α-particle was detected in the focal-plane de-

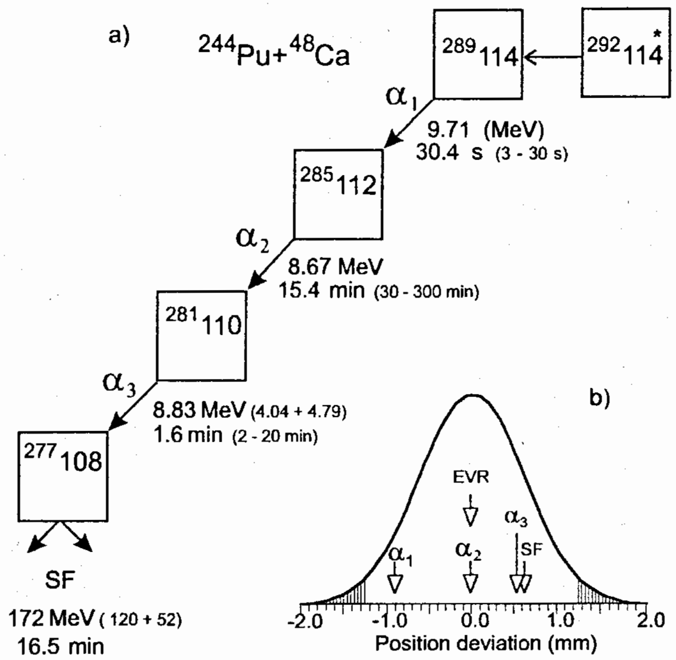

Fig.1 a) Time sequence in the observed decay chain. The expected half-lives corresponding to the measured E_α values for the given isotopes are shown in parentheses following the measured lifetimes. Hindrance factors of 1 and 10 were assumed for α-decay of nuclei with an odd neutron number.
b) Position deviations, in mm, of the observed decay events from the recoil nucleus. The curve shows the position distribution for correlated EVR-α signals; open area corresponds to 95% confidence level

tector 30.4 s after the implantation in the middle of the 8th strip of a recoil nucleus with a measured energy E_{EVR}=6.1 MeV. This value fits the energy expected for element-114 recoils, and the TOF signal is consistent with that expected for a complete-fusion EVR, as determined in the calibration reactions. The energy of the first α-particle was E_α=9.71 MeV. A second α-particle, having an energy E_α=8.67 MeV, was observed at the same location after 15.4 min. A third α-particle, escaping the front detector leaving an energy $E_{\alpha 1}$=4.04 MeV and absorbed in the side detector with $E_{\alpha 2}$=4.79 MeV (E_{tot}= 8.83 MeV), was measured 1.6 min later. Finally, 16.5 min later, the SF event was observed.

All 5 signals (EVR, α_1, α_2, α_3, SF) appeared within a position interval of 1.6 mm (Fig.1b), which strongly indicates that there is a correlation among the observed decays. Assuming that the decay sequence for a valid event will terminate with SF, we developed a Monte Carlo technique to estimate the probability of the candidate event being due to random correlations. Artificially, 10^5 fissions were inserted into the data distributed at random positions and times over the entire detector array and entire experiment duration. We used a search algorithm similar to the one described in the preceding paragraph to find correlations of an EVR-like event followed by three α-particle signals within the energy interval of 8.5-10 MeV in the 34 minutes preceding each artificial fission, using position criteria corresponding to 95% confidence level. The probability per fission of finding such a correlated event was determined to be N_b=0.006. Even without further restricting the energies of sequential events or event-to-event times within the given interval, this value is quite low.

Another N_b calculation was performed for strip 8 at the position in which the candidate event occurred. For a position-correlation window of 1.6 mm the signals from recoil nuclei were observed with a frequency of 2 h^{-1}. The signals of α-particles with E_α=8.1-10.5 MeV occurred with a frequency of 1 h^{-1}. Thus, calculated from event rates alone, the probability of this decay sequence being caused by the chance correlation of unrelated events in strip 8 is 5×10^{-5}.

All events of the decay chain are correlated in time and position. These events arise as a result of the α-decay of a parent nucleus (E_α=9.71 MeV) and continue until SF takes place. This matches the decay of a superheavy nucleus that is predicted by theory. For the whole decay chain the basic rule for α-decay, defining the relation between Q_α and T_α, is fulfilled. This can be seen in Fig.1a where the expected half-lives, corresponding to the measured α-particle energies for the isotopes with the specified atomic numbers of the radioactive family, are shown. For the calculation of half-lives, the formula of Viola and Seaborg with parameters from Refs. [6,7] has been used, with hindrance factors of 1 and 10. The detected sequential decays have larger $T_{1/2}$ vs. E_α values than the known radioactive nuclides. The best candidate for the parent nucleus is the even-odd isotope $^{289}114$, produced in the

3n-evaporation channel. This one event corresponds to a cross section of about 1 pb. The decay properties of the observed nuclei are also in agreement with calculations [6,7], assuming reasonable hindrances for the decay of nuclei with odd neutron numbers.

In our experiment we observed a four-member decay sequence. If we assume that it actually consisted of five decays (the spontaneous fission was due to $^{273}106$), the probability of missing any one of the four α-events is about 34%, but the probability of missing any *particular* α event in the chain and observing the other three is only about 8.5%.

The lifetimes of the new isotopes, in particular $^{285}112$ and $^{281}110$, appear to be approximately 10^6 times longer than those of the known nuclei $^{277}112$ and $^{273}110$ [2,11], which have eight fewer neutrons. Therefore, the observed decay properties of the synthesized nuclides, together with the data obtained earlier for $^{283}112$ [4], can be considered the first experimental proof of the existence of enhanced stability in the region of superheavy elements.

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