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# THE SYNTHESIS AND DECAY PROPERTIES OF THE HEAVIEST ELEMENTS

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The experimental investigations performed in the past few years in GSI and FLNR with the aim to synthesize new isotopes of the heaviest elements are addressed in connection with the theoretical predictions on the existence of "islands of stability" in the region of superheavy elements. The experimental data, which demonstrate the enhanced stability of nuclei close to the deformed shells $Z = 108$ and $N = 162$, as well as the reactions of synthesis, are discussed in the light of the possible advance to the heavier long-lived nuclides located close to the spherical shell $N = 184$. First experimental results are presented on the synthesis of superheavy nuclei in $^{48}$Ca-induced reactions. The considerable increase in the half-lives of the isotopes of heavy elements, situated nearby the closed shells, confirm the basic conclusion of theoretical models about the decisive role played by nuclear structure in favour of the stability of superheavy elements.

## 1. NUCLEAR SHELLS AND THE STABILITY OF HEAVY NUCLEI

One of the fundamental consequences of modern nuclear theory is the prediction of the islands of stability in the region of superheavy elements. This hypothesis proposed more than thirty years ago [1,2] has lately become the subject of intensive experimental research.

According to the classical approach, the transition into the region of extremely heavy nuclear masses and high charges is connected with a substantial loss of their stability against spontaneous fission. This is why the existence of regions (islands) of stability of extremely heavy nuclei, which cannot exist within classical liquid drop models by definition, is caused entirely by the quantum effect of nuclear shells. Calculations of nuclear masses and deformations lately made using macro-microscopic approaches of the type of Yukawa+exponential model with Woods-Saxon single particle potentials (YPE+WS) [3,4] and the determination of the nuclear shell amplitude using Strutinsky's method (or the finite-range droplet model with folded Yukawa single particle potentials (FRDM+FY) [5] have confirmed the predictions made earlier on the existence of quite a stable doubly magic spherical nucleus $^{298}_{184}114$ which follows right after $^{208}_{82}\mathrm{Pb}$. The results of calculations presented in Figure 1 demonstrate the influence of the shell effect on the stability of heavy nuclei.

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We shall first consider the spontaneous fission probability. The partial half-lives of spontaneous fission strongly depend on the amplitude of the shell correction. The significant rise in $T_{SF}(N)$ when moving away from the N = 152 shell, which manifests itself noticeably in the radioactive properties of the actinide nuclei, is due to the influence of another neutron shell at N = 162. It is necessary to note that these two shells are related to deformed nuclei. The maximum stability with respect to spontaneous fission is expected for the nucleus $^{270}108$ (N = 162) for which the predicted $T_{SF}$ value can amount to several hours. When the neutron number increases, the ground-state deformation of the nucleus decreases due to the moving away from the deformed shell N = 162. At N > 170 a significant rise of $T_{SF}$ is expected for nuclei up to $^{292}108$ (N = 184), whose partial half-life with respect to spontaneous fission is as long as $T_{SF} \approx 3.10^4$ years.

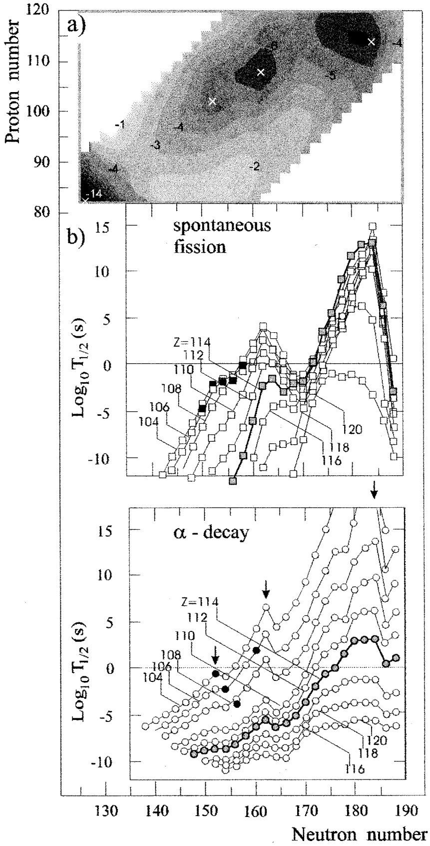

Figure 1. a) Map of the shell corrections (in MeV) to the nuclear liquid drop potential energy. b) The open squares and circles denote calculated half-lives with respect to α-decay and spontaneous fission, the black points – experimental data.

Here we come across a very interesting situation.

If superheavy nuclei possess high stability with respect to spontaneous fission, they will undergo other modes of decay – α-decay and, perhaps, β-decay. The probability for these modes of decay, hence the lifetimes, will be determined by the nuclear masses in the ground state. The latter can be calculated by different models, which are based on different assumptions of the fundamental properties of nuclear matter. In these circumstances any experimental result becomes extremely informative as far as the verification of the theoretical models is concerned. Following the YPE+WS calculations the deformed nucleus $^{268}106$ ($N = 162$) should undergo α-decay with a half-life of $T_\alpha \approx 2$ h. (According to the FRDM+FY calculations – a few days). For the heavier spherical nucleus $^{294}110$ ($N = 184$), $T_\alpha$ increases to several hundred, probably even thousand, years. It is worthwhile noting that in the absence of a nuclear structure (in the liquid drop model) this nucleus should fission spontaneously with $T_{SF} < 10^{-19}$ s. The difference is, as we can see, about 30 orders of magnitude!

Calculations of the energy of the nucleus as a many-body system, carried out in the Hartree-Fock approach, as well as calculations in the relativistic mean field model also indicate a significant increase in the binding energy of the nucleus when approaching the closed neutron shell $N = 184$.

There is yet no agreement among theoreticians regarding the magic proton number for which the maximum binding energy of the spherical nucleus is realized, at the same time the neutron number being $N = 184$. In macro-microscopic models the shell correction amplitude has a maximum value for the nucleus $^{298}114$, irrespective of the variation of the parameters used in the calculations. On the contrary, the calculations performed using the method of Hartree-Fock employing Skyrme effective interaction of the Sly4 (HF+Sly4) type [6], or using a self-consistent relativistic mean-field model with the parameterization of the NL-2Z type (RMF+NL-72) [7] the proton shells are predicted at $Z = 120$ and 126. In the latter case the manifestation of the neutron shell $N = 172$ is also expected. This, however, does not change the main conclusion that in the region of very heavy nuclei there may exist "islands of stability", which in turn substantially can extend the limits of existence of superheavy elements.

## 2. REACTIONS OF SYNTHESIS

It is well known that the first artificial elements heavier than uranium were synthesized in reactions of sequential capture of neutrons during long exposures at high-flux reactors. The long lifetime of the new nuclides made their separation and identification possible using radiochemical methods followed by the measurement of their radioactive decay properties. This pioneering work, which was performed in the Lawrence Berkeley National Laboratory (USA) by Prof. G.Seaborg and colleagues in the period of 1940–1953, led to the discovery of 8 artificial elements with $Z = 93 \div 100$. The heaviest nucleus was $^{257}$Fm ($T_{1/2} \sim 100$ d). The further advance into the region of heavier nuclei was blocked by the extremely short lifetime of $^{258}$Fm ($T_{SF} \sim 0.3$ ms). The attempts to overcome this barrier by means of pulsed neutron fluxes (underground nuclear explosions) also did not go beyond observing $^{257}$Fm.

The transfermium elements with mass $A > 257$ were produced in heavy-ion induced reactions. The interaction of massive nuclei turns out to be a complicated process, characterized by a large number of reaction channels. Only one of them - complete fusion - can lead to the formation of a superheavy nucleus.

The formation cross section of the final nuclei in this channel is defined by the probability of formation and survival of the compound nucleus in the whole available energy region. Having this in mind, fusion reactions, which are used for the synthesis of heavy nuclei, can be conditionally divided into two kinds:

- "cold fusion reactions", based on the use of $^{208}$Pb or $^{209}$Bi targets and characterized by a large Q-reaction-value and, therefore, with a low excitation energy ( $E_x$ ~ 15-20 MeV);
- "hot fusion reactions" in which heavier nuclei, such as isotopes of uranium or transuranium elements, are used as targets ( $E_x$ ~ 30-50 MeV).

We shall start our discussion with the final results. For this purpose in Figure 2 the results are presented on the production cross section of evaporation residues (EVR), obtained in the two types of reactions. We shall try to estimate the potentialities inherent in both methods for the synthesis of nuclei with $Z \geq 112$.

As a whole – this is a difficult task. The theoretical models based on different assumptions on the fusion dynamics of massive nuclei leading to the formation of new elements are tested only by the final results – the rare events of radioactive decay of evaporation residues. The separation of the two stages - the compound nucleus formation and its decay by neutron emission - would be very helpful in providing more information for understanding of the reaction process. The compound nucleus formation cross section can be in principle determined by the characteristics of its main decay mode – the fission into two fragments. Such attempts were made recently in the experiments carried out by M.G.Itkis et al. in FLNR (Dubna) by means of systematic measurements of the angular correlations, mass and energy distributions of the fission fragments as well as in some cases of the fission neutrons [8,9].

As a reference reaction, we shall consider the reaction $^{48}$Ca + $^{206,208}$Pb, leading to the formation of isotopes of No (Z = 102), whose fission barrier height is determined mainly by the amplitude of the shell correction to the nuclear deformation energy.

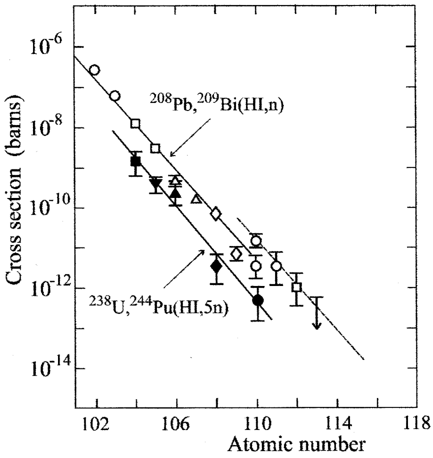

Figure 2. EVR cross sections for cold and hot fusion reactions.

The two-dimensional mass and kinetic energy spectra presented in Figure 3a for the correlated fragments indicate that there are two different channels for their formation: asymmetric mass distribution due to the massive transfer of nucleons from the target nucleus to the projectile and symmetric distribution arising as a result of the fission of the compound nucleus with Z = 102. Comparing the dependence $\sigma_{CN}(E_x)$ with the cross sections $\sigma_{EVR}$ in the xn-channels (x = 1– 4), it is possible to determine the survival probability of the compound nucleus $^{254,256}$No [10] in a wide excitation energy range (Figure 4). On the other hand, the survival probability could also be calculated within the statistical model. Further, the dependence $B_f(E_x)$ can be determined from the best fit between calculation and experiment. The use of cold fusion reactions for the advent into the region of heavier elements brings forth a considerable drop in $\sigma_{CN}$. When the projectile mass increases by 10 a.m.u. (i.e., $^{58}$Fe instead of $^{48}$Ca), only a small part of the fragments in the region of symmetric masses can be attributed to the fission of the compound nucleus $^{266}$Hs (Z = 108) (Figure 3b). At the same time, the experimentally observed drop by 3 orders of magnitude in the cross section for $^{265}$Hs in comparison to $^{255}$No (both nuclei are formed in the 1n-evaporation channel at $E_x$ ~15 MeV) is due to the small survival probability of the heavy nucleus (Figure 5). The survival probability, as it is well known, is defined by the neutron binding energy in the nucleus $^{266}$Hs ($B_n$ = 8.03 MeV and the height of its fission barrier $B_f$ ~ 5 MeV).

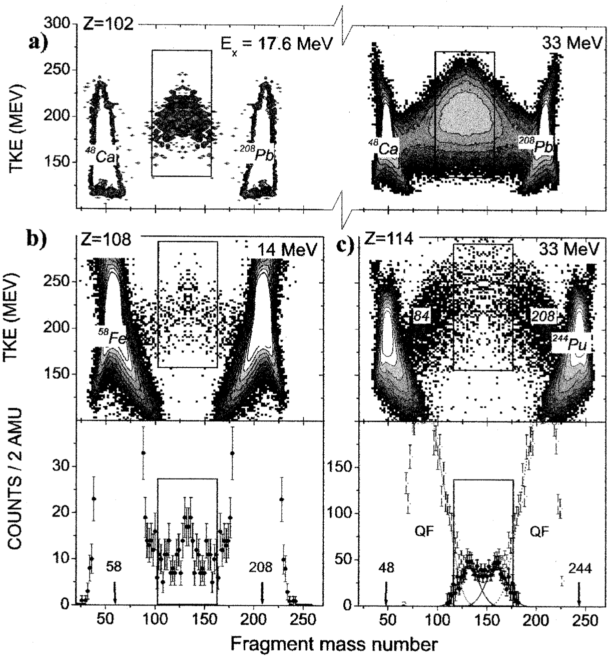

Figure 3. TKE vs $A_f$ for correlated fragments, obtained in the reactions:
a) $^{48}$Ca + $^{208}$Pb, b) $^{58}$Fe +$^{208}$Pb, c) $^{48}$Ca + $^{244}$Pu.
The events marked with a square are attributed to the fission of a compound nucleus.

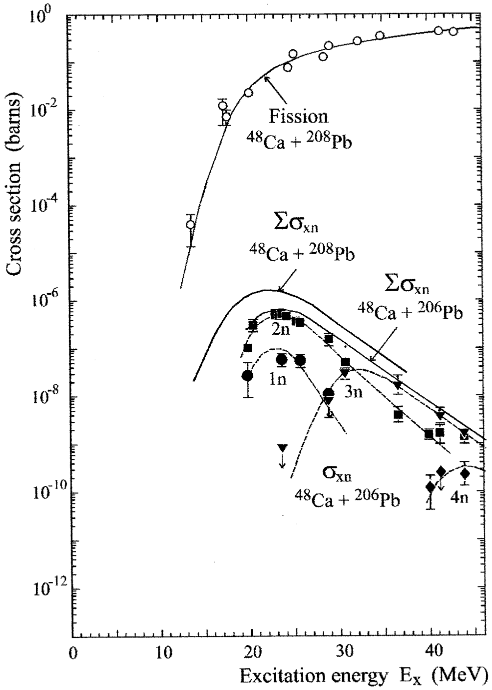

Figure 4. Cross sections for compound nucleus (fission) and evaporation residues, obtained in the $^{48}Ca + ^{206,208}Pb$ reaction. The lines are drawn through the experimental points to guide the eye.

In the synthesis of heavier nuclei this trend is kept. In the reaction $^{86}Kr + ^{208}Pb$ only the upper limit of $\sigma_{CN}$ was obtained for $^{294}118$ at the level of $\sigma_{CN} \leq 0.5$ μb [11]. We should note that this limit is approximately 200 times less than the cross section for the formation of the compound nucleus $^{266}Hs$ [12] at the same excitation energy $E_x \sim$ 14 MeV.

The other means - the transition to more asymmetric reactions is connected to the increase of the target mass. The transition from $^{208}Pb$ to the isotopes of actinide elements such as $^{238}U$ and $^{244}Pu$ (the target mass is increased by 30-36 a.m.u.) qualitatively changes the picture of the fusion process.

The main part of the fission fragments, as it is shown in Figure 3c, is connected to the asymmetric quasifission of the nucleus $^{292}114$ into two fragments with masses close to $A_1$ = 208 and $A_2$ = 84. Only a small part of the fragments in the region of the medium mass nuclei can be attributed to the fission of the compound nucleus $^{292}114$. In this region the fission fragment mass distribution also is asymmetric with maximal close to $A_{f1}$ = 132 and $A_{f2}$ = 160. The asymmetry in the fission of the superheavy nucleus is most probably due to the shells Z = 50 and N = 82 in the $^{132}Sn$ nucleus, formed as a light fragment. A similar situation, as it is well known, is observed also in the fission of actinide elements (Z = 90 - 98) with the only difference in that the shell effect manifests itself in the formation of the heavy fragment.

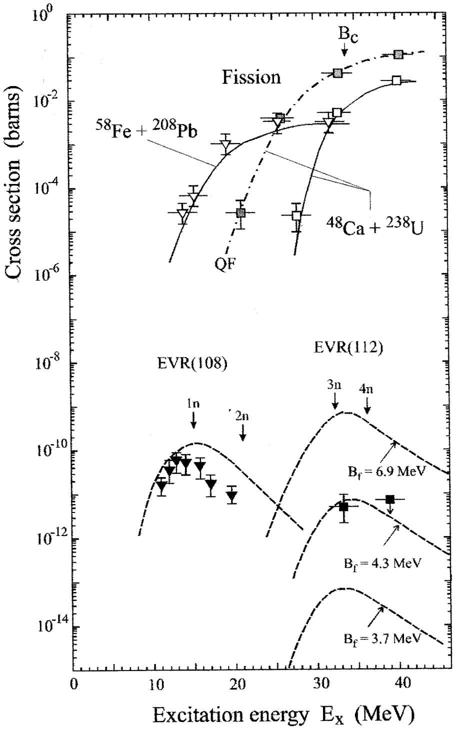

Figure 5. Experimental cross sections, obtained in fusion-fission reactions with $^{208}$Pb-targets and $^{48}$Ca, $^{58}$Fe-beams at different excitation energies. Open symbols denote symmetric fission; black symbols - EVR; dot-dashed line – quasifission; dashed lines - calculated cross sections for EVR.

In asymmetric hot fusion reactions, where the compound nucleus excitation energy even at the Coulomb barrier amounts to 45-50 MeV, the survival probability strongly depends on $E_x$. For this reason the choice of the nuclei participating in a hot fusion reactions leading to compound nuclei with a minimal excitation energy is of great importance. Most promising from this point of view seem to be the fusion reactions induced by $^{48}$Ca-ions. Due to the considerable mass excess of the doubly magic nucleus $^{48}$Ca, the excitation energy of the compound nuclei with Z = 112, 114 at the Coulomb barrier amounts to about 30 MeV. However, even in these favourable conditions $\sigma_{EVR}$ is very sensitive to the fission barrier of the compound nucleus. Following the calculations of Meyers and Swiatecki [13] predicting for the $^{286}$112 nucleus a fission barrier of about 6.9 MeV, we can expect that the maximum cross sections $\sigma_{EVR}$ in the reaction $^{238}$U($^{48}$Ca,3-4n)$^{283,282}$112 may reach hundreds of *pb*. However, $\sigma_{EVR}$ decreases by almost 3 orders of magnitude for $B_f \sim 3.7$ MeV, obtained in the macro-microscopic calculations of R.Smolanczuk [4]. This may be the reason for which previous attempts to produce superheavy elements in $^{48}$Ca-induced reactions for which the production cross sections amounted to hundreds of picobarn could not be successful [14-16]. Hence, for the synthesis of superheavy elements the sensitivity of the new experiments in comparison to earlier ones has to be improved by hundreds of times.

## 3. EXPERIMENTAL APPROACH AND SET-UP

The planning of experiments on the synthesis of heaviest elements is determined to a great extent by their radioactive properties and above all by the lifetime of the atoms to be synthesized. The lifetime, as mentioned earlier, can vary in a wide range depending on how justified are the predictions concerning the influence of nuclear shells on the stability of heavy nuclides with different Z and A. Hence the experimental set-up should be sufficiently fast. On the other hand, the evaporation residues, whose yield is extremely small, should be quickly separated from the enormous background of incidental reaction products, which are formed with an 8-10 orders of magnitude higher probability.

These conditions can be achieved if the separation of the products is performed in-flight (during $10^{-6} \div 10^{-5}$ s), taking into account the kinematical characteristics of the reaction channels. Such an operation can be performed by Wien velocity selectors (the separator SHIP in GSI) [17] or the energy selector (the separator VASSILISSA in JINR) [18], where the reaction products are separated according to the electric rigidity in transverse electric fields. Essentially, such operations can also be performed by gas-filled separators, where the separation of the recoil atoms is achieved by the magnetic rigidity in a gaseous hydrogen or helium atmosphere at a pressure of about 1 torr [19].

The efficiency of the kinematic separators depends on the ratio of the masses of the interacting nuclei. For fusion reactions induced by relatively light projectiles ($A_p \leq 20$) it amounts to only a few percent, but increases to $30 \div 50\%$ when going to ions with mass $A_p \geq 40$. The experimental set-ups have also high selectivity. In the focal plane practically all the background from the primary beam is eliminated and the products of incomplete fusion reactions are suppressed by a factor of $10^4 \div 10^7$, depending on the kinematical characteristics of the different channels leading to their formation. This is, however, not enough for the identification of the extremely rare events corresponding to the production of atoms of a new element. Therefore, the selection of the sought nuclei is further accomplished with a sophisticated registration device (Figure 6).

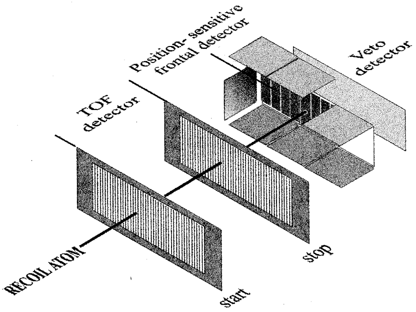

Figure 6. The detector array used as a focal plane detector.

The recoil atoms, which have reached the focal plane, are implanted into a multistrip silicon semiconductor detector with an active area of ~ 40 ÷ 50 $cm^2$. The length and width of the strips, as well as their number, is determined by the size of the object image on the focal plane of the separator. Each strip had longitudinal position sensitivity. The position resolution depends on the particle type (recoil nucleus, α-particle or spontaneous fission fragments). However, more than 95% of all particles, accompanying the decay of the implanted atom, are confined in an interval $\Delta_x$ ~ 1.0 - 1.5 mm. The front detector is surrounded by side detectors so that the entire array has the shape of a box with an open front wall. In this way the detection efficiency for particles resulting from the decay of the implanted nucleus (α-particles or two fission fragments) is increased to 85 ÷ 87%. For distinguishing between the signals of the recoil nucleus and those belonging to the particles from its decay, a time-of-flight (TOF) detector is situated before the front detector. The signals from the TOF detector are used also for determining the velocity of the implants.

The selection of the events according to their generic decay significantly improves the background conditions. The parent nucleus, implanted into the detector, can be reliably identified if the decay chain of its sequential α- and β-decays leads to nuclei with known properties. This method has been successfully used in the experiments on the synthesis of new elements with $Z = 107 \div 112$ whose isotopes have insignificant neutron excess ($N\text{-}Z \leq 53$). Advancing into the region of spherical, more neutron-rich nuclei this advantage is lost. Here the decay of the parent nucleus results in the formation of hitherto unknown nuclei, whose properties can be only predicted with precision allowed by the theoretical calculations.

At the same time, if the basic theoretical prediction on the existence of the "island of stability" is justified, in any decay chain of sequential α- and β-decays the daughter nuclei move more and more away from the closed spherical shells. Finally the decay chains will be terminated by spontaneously-fissioning nuclei. In principle, such a decay scheme appears to be a reliable sign of the formation of a heaviest nucleus. From a technical point of view the appearance of such an event should strongly differ from other possible correlated decays. Following the signal from the recoil atom, which has been implanted into a definite position in the front detector, and the measurement of its time of flight by the TOF detector, in the same position window in definite time intervals, signals will occur due to the emission of an α-particle with an amplitude corresponding to its energy ~8.5 ÷ 10 MeV (without a TOF signal). Next, a signal with high amplitude will be registered, which is the result of the spontaneous fission fragments with total kinetic energy TKE ~ 200 MeV.

## 4. ENHANCED STABILITY OF NUCLEI CLOSE TO THE DEFORMED SHELLS Z = 108, N = 162

The synthesis and the decay properties of nuclides in this region as a matter of fact provide the first test of the theoretical concepts on the role of shell effects in nuclei, where the liquid drop fission barrier is practically absent.

Most interesting for this purpose seem to be the even-even isotopes of Sg ($Z = 106$), which can be synthesized with relatively high cross section (≥ 100 *pb*) in cold and hot fusion reactions. For nuclei with $Z = 106$ situated between the two shells $N = 152$ and $N = 162$, the increase of the neutron number should, according to theory [3], considerably enhance their stability with respect to both α-decay and spontaneous fission (Figure 7).

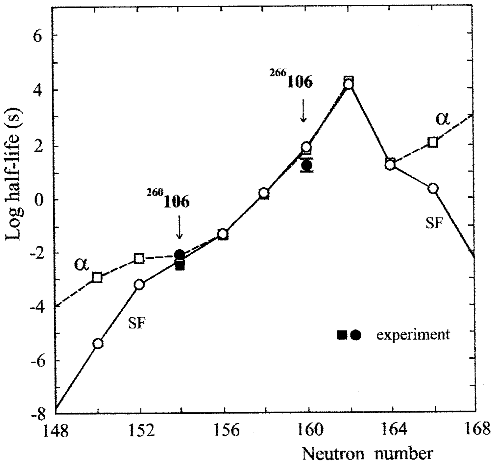

Figure 7. Half-lives for α-decay and spontaneous fission for the Sg-isotopes (Z = 106). Black symbols – experiment; open symbols – calculated values [3].

For the synthesis of the heavy isotopes of element 106 we chose the reaction $^{22}$Ne + $^{248}$Cm at a beam energy close to the Coulomb barrier, where the maximum cross section expected is for the 4n- and 5n-evaporation channels. In a 360-hour irradiation of the $^{248}$Cm-target with a total beam dose of $1.6 \cdot 10^{19}$, at the gas-filled separator two isotopes of element 106 with masses 265 and 266 were synthesized [20].

The two nuclei $^{265}$Sg (N = 159) and $^{266}$Sg (N = 160) undergo predominantly α-decay. The α-decay energy of the even-even nuclide $^{266}$Sg ($Q_\alpha$ = 8.76 MeV) determines its half-life $T_{1/2} = 20 \pm 10$ s. On the basis of 6 registered correlated events (α – SF), observed in the decay of the nucleus $^{266}$Sg, the half-life of the daughter nucleus $^{262}$Rf (Z = 104, N = 158) was also determined. It undergoes spontaneous fission with $T_{SF} \sim 1.2$ s. The radioactive properties of the new nuclides indicate a considerable rise in the stability of heavy nuclides with respect to spontaneous fission while approaching the closed shells Z = 108 and N = 162 (Figure 7) and quantitatively agree with the macro-microscopic calculations of Patik et al. [3].

The data obtained for the Z = 106 nuclei force the conclusion that for even heavier nuclei with Z > 106 the fission modes will not bring forth a revision of the optimistic theoretical predictions on the existence of a wide region of stability of heavy nuclei. They will predominantly undergo α-decay as long as the shell corrections to the deformation energy will restrain spontaneous fission. These conclusions are confirmed by the experimental investigations, carried out at GSI, where many new α-radioactive isotopes with atomic numbers up to Z = 112 were synthesized and their properties determined [21-23]. To an equal extent these conclusions are true also for the heaviest isotopes with Z = 106 - 110, obtained in FLNR in hot fusion reactions [20,24,25].

In Figure 8 three decay chains of the isotope $^{277}$112 are presented. They were observed in the experiments carried out by S.Hofmann et al. in the $^{70}$Zn + $^{208}$Pb reaction [26]. The energies and times of the sequential α-decays of this nucleus, as well as those for nuclei with Z < 112, as it will be shown below are also in good agreement with the calculated values of

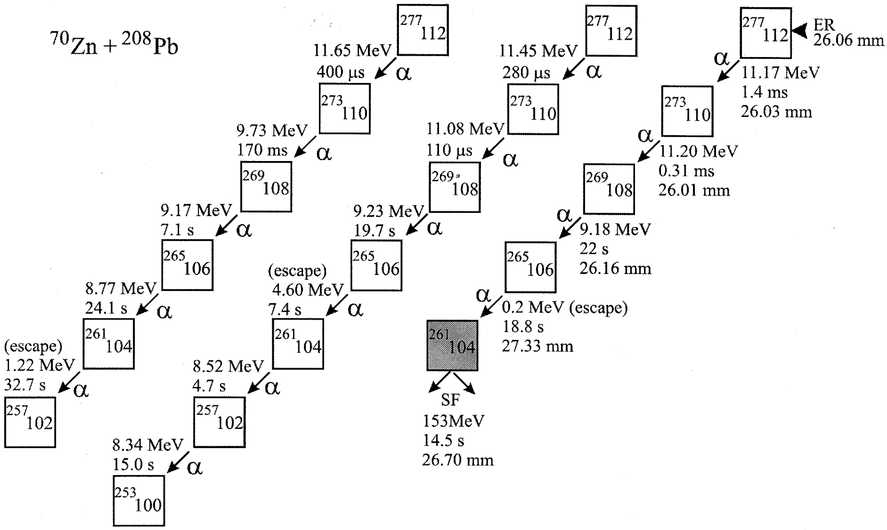

Figure 8. Decay chains of the isotope $^{277}112$.

nuclear masses close to the shells Z = 108 and N = 162. We should note that in one of the three decay events of the even-odd nucleus $^{277}112$ the decay chain terminates by spontaneous fission. With the increase of the neutron number (going away from N = 162), the spontaneous fission probability will be rising up to the moment when the stabilizing effect of the next shells will start to play a role.

## 5. TRANSITION TO THE REGION OF SPHERICAL NUCLEI. THE $^{48}$Ca + $^{244}$Pu REACTION

According to the macro-microscopic theory, the maximum contribution of nuclear shells appears at Z = 114 and N = 184 (Figure 1). No combination of stable or even long-lived isotopes can bring us to the very neutron-rich isotopes of element 114. One may hope to approach at least the boundaries of this unknown region so as to come under the influence of the N = 184 spherical shell. For this purpose the fusion reaction $^{48}$Ca + $^{244}$Pu was chosen, as it leads to the formation of the compound nucleus $^{292}114$ (N = 178). The experiments were performed at the Dubna Gas-Filled Recoil Separator.

The target consisted of the enriched isotope $^{244}$Pu (98.6 %), supplied by our collaborators from LLNL (Livermore). The energy of the $^{48}$Ca-beam in the middle of the target was chosen equal to 236 MeV. Taking into account the energy losses in the target and the weak variations of the beam energy during the long-term irradiations, the excitation energy of the compound nuclei should be in the range 31.5 to 39 MeV. However, for each event of a recoil atom registered in the detector array we could determine the exact (prompt) beam-energy value and the target-segment in which the event originated. The average intensity of the $^{48}$Ca-beam on the target was about $4 \cdot 10^{12}$ pps at a rate of material consumption equal to about 0.3 mg $h^{-1}$.

In these conditions two practically identical experiments were carried out.

In one of them at a beam dose of $1\cdot10^{19}$ ions two identical α-decay chains terminated by spontaneous fission were registered [27]. The genetic relation in the chain was determined according to their position. All 4 signals (EVR, $\alpha_1$, $\alpha_2$, SF) appeared within a position interval of 0.5 mm, which indicates that there is a strict correlation between the observed decays (Figure 9a). In the limits of the detector energy resolution and the statistical uncertainty in the decay times, the two events coincide in all measured parameters. The probability of random coincidences imitating recoil nuclei and their correlated decays is estimated to be less than $5\cdot10^{-13}$.

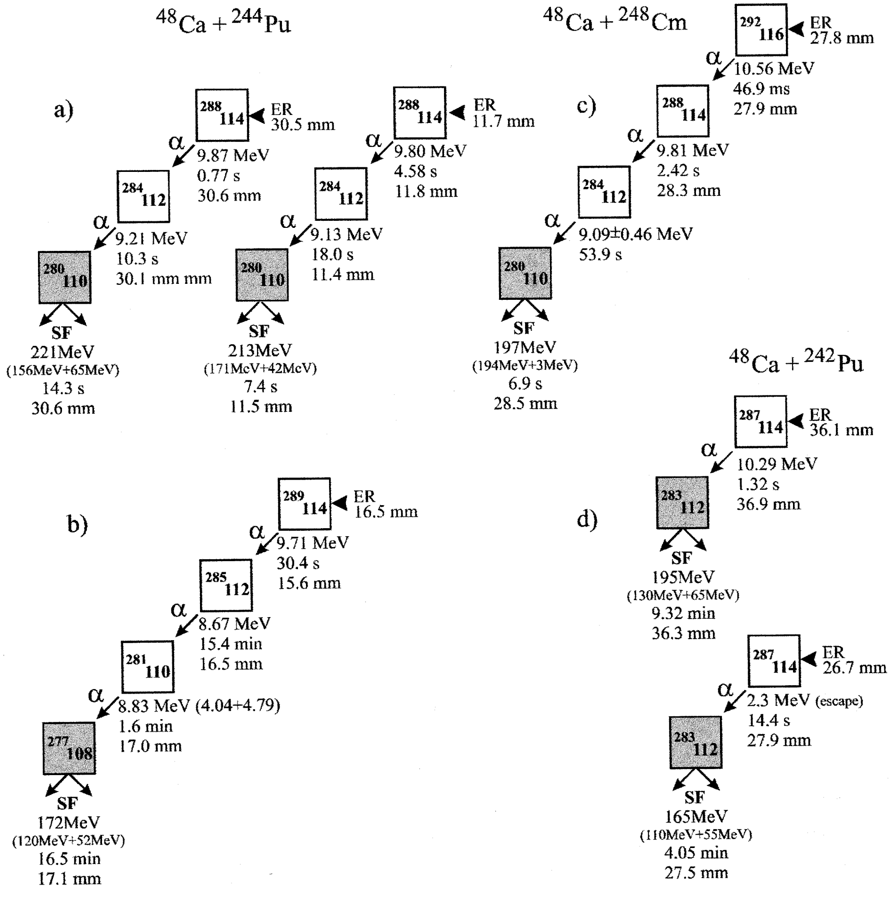

Figure 9. Decay sequences, observed in the $^{48}$Ca + $^{242,244}$Pu, $^{248}$Cm reactions. For the SF fragments the energy deposit in the front and side detectors is shown. For all registered signals positions in the strip detector are presented.

We should note that in this long-term experiment only two spontaneous fission events were observed: they are characterized by a large value of the fission fragment kinetic energy (TKE ~ 235 MeV) and both are preceded by the same α-decay sequences. The projectile energy, measured at the moment of registration of the given events, corresponds to an excitation energy of the compound $^{292}114$ nucleus equal to $E_x = 37 \pm 2$ MeV. At this energy the most probable 4n-evaporation channel leads to the formation of the isotope $^{288}114$ (N=174). Evaporation channels accompanied by the emission of charged particles (protons, α-particles) are strongly suppressed due to the high Coulomb barrier of the heavy nucleus. Channels leading to the formation of heavy spontaneously fissioning isotopes with $Z \geq 98$ ($Q > 80$ MeV) are also hindered.

The parent nucleus $^{288}114$ undergoes α-decay with a decay energy $Q_\alpha = 9.98 \pm 0.05$ MeV and a half-life $T_\alpha = 1.8^{+2.1}_{-0.6}$ s. For the even-even nuclei the value of $Q_\alpha$ is directly connected with the mass difference between the parent and daughter nuclei. According to the Geiger-Nuttall rule, the energy and probability of α-decay, $Q_\alpha$ and $T_\alpha$, determine the atomic number of the parent nucleus. Following the relation $T_\alpha(Q_\alpha)$ as a version of the Viola and Seaborg formula with constant coefficients, describing the α-decay of all known 58 even-even isotopes with $Z > 82$ and $N > 126$ [4], it follows that the first-step α-decay in the observed decay sequences and the third event – from the decay of the $^{292}116$ nucleus (see below) is due to the nucleus with $Z = 114.4^{+1.2}_{-0.8}$. The daughter nucleus, the even-even isotope $^{284}112$ also undergoes α-decay with $Q_\alpha = 9.30 \pm 0.05$ MeV and a half-life $T_\alpha = 19.0^{+22.7}_{-6.7}$ s. Its atomic number can be estimated as $Z = 111.2^{+1.2}_{-0.7}$. Finally, the "grandchild", the nucleus $^{280}110$ undergoes spontaneous fission with a half-life $T_{SF} = 7.5^{+13.7}_{-2.9}$ s. For the two observed binary events the fission fragment energy release in the detectors is about 40 MeV higher than the value obtained in the reaction $^{208}$Pb($^{48}$Ca,2n) for the known spantaneously fissioning nucleus $^{252}$No. Regardsless of the relatively wide fragment kinetic energy distributions (TKE) manifested in spontaneous fission, this value also gives evidence that the "grandchildren" in the decay chain are the result of the fission of a sufficiently heavy nucleus ($Z > 106$).

In a second experiment (chronologically, it was the first one), with a beam dose of $5.2 \cdot 10^{18}$ ions, another longer α-decay sequence was observed terminating by spontaneous fission [28]. Applying the same criteria as in the previous case, we came to the conclusion that this, still single, event was due to the decay of the neighbouring isotope of element 114. In this decay chain, all 5 signals (recoil nucleus, $\alpha_1$, $\alpha_2$, $\alpha_3$, SF) appeared within a position interval of 1.5 mm, which also is a strong indication that there is a correlation between the observed decays (Figure 9b). The total time between the implantation of the recoil nucleus and the spontaneous fission amounts to 34 min. The probability of random coincidences imitating such a decay in any point of the working area of the detector was less than 0.6%. For that place, where the decay occurred (the given position window in the considered strip), this probability was even less ($\sim 10^{-4}$).

For all the steps of the decay sequence the basic rule for α-decay, giving the relation between the decay energy $Q_\alpha$ and the half-life $T_\alpha$, was also satisfied. Considering the experimental conditions and the observed decay characteristics, the origin of the decay chain is most probably found in the isotope $^{289}114$, which has been produced in the 3n-evaporation channel. Despite the high efficiency of the detector array, in the long decay sequence the probability to lose one of the α-particles is ~34%. Then the observed spontaneous fission can be attributed to the decay of the nucleus $^{273}106$. The probability to lose the first α-particle of the chain is about 8.5%.For this reason the first transition with a 91.5% probability can be considered as due to the decay of EVR.

The decay properties of the neighboring odd isotope $^{289}114$ agree well with the above-mentioned properties of the even-even nucleus $^{288}114$. For this nucleus, as it was expected, an increase is observed in the halflives $T_\alpha$ and $T_{SF}$ that is due to the additional odd neutron.

The production cross section of the new nuclides amounts to about 1 pb. In spite of the small value of the cross section, obtained in both experiments, background from spontaneous fission is practically negligible. At a total beam dose of $1.5 \cdot 10^{19}$ ions, in addition to the observed decays only two decay events were registered, which could be attributed to the short-lived spontaneously fissioning nucleus $^{244m}$Am ($T_{SF)} = 0.9$ ms) with energy release in the detectors amounting to 149 and 153 MeV. From this point of view the sensitivity of the experiment can in the future be raised by increasing the intensity of the $^{48}$Ca-beam.

## 6. EXPERIMENTS WITH THE $^{238}$U AND $^{242}$Pu TARGETS

If the identification performed in the preceding two experiments with the $^{244}$Pu target is correct, it is not difficult to predict the properties of another isotope, viz. $^{287}114$ (N = 173). It should predominantly undergo α-decay to the daughter nucleus $^{283}112$, which was formed in a previous experiment in the $^{48}$Ca + $^{238}$U reaction [29]. In the given case we can expect a short decay sequence (α-SF), implying a few-seconds α-decay half-life, followed by spontaneous fission with considerably longer (of the order of minutes) half-life. This isotope of element 114 can be synthesized via the 3n-evaporation channel in the $^{48}$Ca + $^{242}$Pu reaction.

The experiments were performed at the VASSILISSA separator [30]. The rotating 0.2 mg/cm$^2$ thick $^{242}$Pu target was bombarded by a 235 MeV $^{48}$Ca-beam with a total beam dose of $7.5 \cdot 10^{18}$ ions. The most probable deexcitation channel of the compound nucleus $^{290}114$ ($E_X \approx 33.5$ MeV), corresponding to the emission of 3 neutrons, leads to the formation of the even-odd isotope $^{287}114$ (N = 173).

In this experiment 4 spontaneous fission events were observed.

Two spontaneous fission events were registered 59 and 20 μs after the implantation of corresponding position-correlated recoil atoms, respectively. We could assign these events to the SF-isomer $^{241mf}$Pu(24-μs), produced in the one-neutron transfer reaction on the $^{242}$Pu-target nucleus. In the case of the two other events, the search for α-decays preceding the spontaneous fission revealed the two decay chains presented in Figure 9d.

In one of the sequences only one α-particle ($E_\alpha = 10.29$ MeV) was detected by the front detector 1.32 s after the implantation of a heavy recoil. Spontaneous fission was observed 9.32 min later. All three signals (recoil nucleus, α, SF) appeared within a position interval of 0.82 mm, which indicated that there is a correlation between the observed decays. The second decay chain is similar to the previous one with the exception that the detector registered only a part of the α-particle energy emitted in the back hemisphere (the open window). The probability that both events are due to random coincidences imitating the decay chains (recoil nucleus, α, SF) in the given position intervals is less than $10^{-4}$.

In both events the parent nucleus undergoes α-decay. The half-life of the parent nucleus, derived on the basis of the two events, amounts to $T_\alpha = 5.5^{+10}_{-2}$ s. The detected daughter nuclei undergo spontaneous fission. Their decay properties are comparable with the ones of the spontaneously-fissioning nuclide produced earlier in the $^{48}$Ca + $^{238}$U reaction [29]. All four spontaneous fission events, observed in these two reactions, within the experimental error, can be described by the same half-life $T_{SF} = 3.0^{+2.8}_{-1.0}$ min and can be attributed to the decay of one and the same nucleus. In the $^{48}$Ca + $^{238}$U reaction this nuclide was produced directly as an EVR in the 3n-evaporation channel, while in the $^{48}$Ca + $^{242}$Pu reaction it is the daughter of the α-decay of the parent $^{287}$114 nucleus ($E_\alpha = 10.29$ MeV).

The production cross section of the new isotope of element 114 amounts to about 2 pb.

Its half-life and the decay sequence are shorter than the ones of the previously observed heavier isotope $^{289}$114, formed in the reaction $^{48}$Ca + $^{244}$Pu. Such a trend is expected, according to theory, with the decrease of the neutron number of the superheavy nucleus, or in other words with moving away from the closed N = 184 shell.

## 7. OBSERVATION OF THE DECAY OF $^{292}$116

On June 14, 2000, an experiment aimed at the synthesis of superheavy nuclei with Z=116 in the complete fusion reaction $^{248}$Cm + $^{48}$Ca was started at FLNR.

Already after the conference, on 19 July, on the 35-th day of irradiation, with the accumulated beam dose of 6.6×10$^{18}$ ions, the first event sequence was observed, that can be assigned to the implantation and decay of the isotope of element 116 with mass number 292, see Figure 9c.

Implantation of a heavy recoil in the focal-plane detector was followed, in 46.9 ms, by α-particle with $E_\alpha$=10.56 MeV. This sequence switched the ion beam off, for one hour and further decays were detected under lower-background conditions. Second α-particle with $E_\alpha$=9.81 MeV was observed 2.42 s later. Then, in 53.87 s the third α-decay was registered by the side detector with the energy of 8.63 MeV. The energy deposited by this α-particle in the focal-plane detector was lower than the detection threshold of 0.92 MeV. Thus it's total energy is determined with larger uncertainty, $E_\alpha$=9.09±0.46 MeV; the probability that the third α-particle appeared in the chain (Δt~1 min) due to random count can be estimated as ~1%.

Finally, 6.93 s after the last α-decay, two coincident fission fragments with sum energy of 197 MeV were registered by both the focal-plane and the side detectors. The low energy of one fission fragment measured by side detector for this event means large energy lost by this fragment in the dead layers.

Positions of the four events (EVR, $\alpha_1$, $\alpha_2$, and SF) were measured to be within a window of about 0.5 mm, all events appeared within time interval of 63.26 s, which points to a strong correlation between them. The probability of the random origin of the observed event chain is negligible ($<10^{-10}$).

All the decays following the first 10.56-MeV α-particle agree well with the decay chains of $^{288}$114, previously observed in the $^{244}$Pu+$^{48}$Ca reaction, see Figure 9a.

Thus, it is reasonable to assign the observed decay the nuclide $^{292}$116, produced via evaporation of 4 neutrons in the complete-fusion reaction $^{248}$Cm+$^{48}$Ca. Decay energy of the new observed nuclide is Q = 10.71 MeV, its half-life estimated from one event is $T_\alpha = 33^{+155}_{-15}$ ms.

Experiments are in progress.

## 8. COMPARISON WITH THEORY

The data on the radioactive decay of nuclides, produced in the given series of investigations, can be compared with the predictions of different theoretical models.

Unfortunately, in many cases the calculations go no further than look for the region of greatest stability of superheavy nuclides without giving the definite properties of nuclei comprising this region. Therefore we shall consider only those few cases, which can be directly applied for comparison with the experiment.

First we would like to point out that the heaviest isotopes with Z = 110, 112 and 114, produced in the $^{48}$Ca-induced reactions, undergo α-decay. Spontaneous fission in this region of nuclei is observed only for elements when $(N-Z) \leq 61$.

For the even-odd nuclei $^{277}$108 (N = 169) and $^{283}$112 (N = 171) the spontaneous fission half-lives turn out to be respectively 5 and 3 orders of magnitude higher than the predicted values for the neighbouring even-even nuclei. Such a difference can be explained by the presence of the extra neutron, which significantly diminishes the probability for spontaneous fission of the heavy nucleus. For the even-even nucleus $^{280}$110 (N = 170) the experimental value ($T_{SF} \sim 10$ s) is also about 3 orders of magnitude longer than the calculated one, obtained in ref. [3]. Keeping in mind the uncertainty in calculating the probability for spontaneous fission, connected with tunnelling through the potential barrier, this difference could be taken as an evidence of the larger contribution of the shells to the deformation energy of the nucleus.

Some conclusions can be drawn on the basis of the ground state properties of the observed nuclei (Table 1).

The experimental values of the α-decay energies of all known nuclides with $Z \geq 100$ and $N \geq 148$ are shown in Figure 10, together with the calculated values $Q_\alpha$, obtained in the macro-microscopic theory, for all even-even isotopes of the same elements [3,4].

Certainly, the experimental results well reflect the expected by theory changes $Q_\alpha(N)$ for different N and Z, including the region of superheavy elements, where a transition from deformed to spherical shells is predicted. Quantitatively, for nuclei located in the transition region between the deformed neutron shells N = 152 and N = 162 a negligible difference $\Delta Q_\alpha \leq 0.2$ MeV is observed between the calculated and experimental values. When going to the spherical shell in the region N = 170 ÷175 the difference grows up to $\Delta Q_\alpha \leq 0.5$ MeV.

Other calculations, carried out by S.Cwiok, W.Nazarewicz and P.H.Heenen using the Hartree-Fock-Bogoliubov method with selected Skyrme forces, concerned the heaviest nuclide $^{289}$114, produced in the $^{48}$Ca + $^{244}$Pu reaction [6]. Within this approach, which can definitely be applied to other nuclei also, the quite good agreement obtained between the calculation and experiment ($\Delta Q_\alpha \leq 0.25$ MeV) for the chain $^{289}$114 – $^{285}$112 – $^{281}$110. The calculations of the α-decay energies of heavy nuclei, performed recently by M.Bender [7] in the relativistic mean field theory, where, according to the author, the spin-orbit interactions of the particles are calculated more precisely, very well agree with the experimental values of $Q_\alpha$ for the sequence of the even-even nuclei $^{288}$114 – $^{284}$112, but greatly differs from $Q_\alpha$(exp) for $^{292}$116. For the decay sequences of the even-odd nucleus $^{289}$114 the calculated $Q_\alpha$ values differ from the experimental ones by $\Delta Q_\alpha \leq 0.3$ MeV. According to this model, the small values of $Q_\alpha$ and the corresponding long half-lives of the isotopes of element 114, which were obtained in the $^{48}$Ca + $^{244}$Pu reaction, are due to the appearance of local deformed sub-shells with Z = 114 and N = 174. This, however, does not exclude the existence of spherical shells in the more neutron-rich nuclei.

Table 1
Experimental and calculated $Q_\alpha$ values for the $\alpha$-decay chains of the isotopes Z=110-116

| Z | A | $Q_{exp.}$ (MeV) | $Q_{theor.}$ | | | |
|---|---|---|---|---|---|---|
| | | | YPE+WS | FRDM+FY | SHFB | RMF |
| 110 | 273 | 9.87<br>11.24±0.02<br>11.36 | - | 10.09 | - | 10.82 |
| 110 | 280 | ≤9.4[a] (SF) | 9.8 | 9.05 | 9.8 | 8.98 |
| 110 | 281 | 8.96±0.18 | - | 8.55 | 9.44 | 8.68 |
| 112 | 277 | 11.82<br>11.62±0.02<br>11.33 | - | 12.08 | - | 11.32 |
| 112 | 283 | 10.39±0.02 | - | 9.01 | - | - |
| 112 | 284 | 9.3±0.05 | 9.8 | 8.69 | 9.42 | 9.3 |
| 112 | 285 | 8.80±0.05 | - | 8.59 | 8.76 | 9.02 |
| 114 | 288 | 9.98±0.05 | 10.3 | 9.16 | 9.35 | 9.83 |
| 114 | 289 | 9.85±0.05 | - | 8.87 | 10.16 | 9.38 |
| 116 | 292 | 10.71±0.05 | 11.07 | 10.82 | 10.61 | 11.65 |

YPE+WS – macroscopic-microscopic Yukawa-plus-exponential model with Woods-Saxon single-particle potentials.
FRDM+FY – macroscopic-microscopic finite-range droplet model with folded Yukawa single-particle potentials.
SHFB – self-consistent Skyrme-Hartree-Fock-Bogoliubov model with pairing.
RMF – self-consistent relativistic mean-field model.

[a] The $Q_\alpha$ limit was calculated from experimental $T_{1/2}$ value using the formula by Viola and Seaborg with parameters [4].

More definitely a comparison between theoretical calculations within different approaches and experiment, from our point of view, can be made for the even-even isotopes, where $\Delta Q_\alpha$, as it is well known, corresponds to the mass difference in the ground state. As it can be seen in Figure 11, the variation in $\Delta Q_\alpha$ for 5 known even-even nuclei with Z = 106 – 114 amounts to about ±0.7 MeV (this is approximately 7% of the calculated energy $Q_\alpha$). It is clear that theoreticians have the possibility to improve the agreement with experiment, while the experimentalists can add new even-even nuclides to the already known ones. For this purpose, cold fusion reactions can be used as before using $^{207}$Pb targets and hot fusion reactions of Th-Cm targets with $^{36}$S and $^{48}$Ca ions.

Finally, it is necessary to point out that the decay properties of the heaviest nuclei that have been synthesized up till now confirm the basic conclusions of the shell model on the decisive role of the nuclear structure on the stability of superheavy elements.

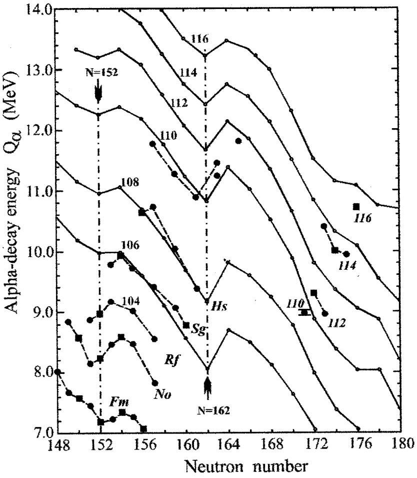

Figure 10. Solid lines and open symbols denote calculated values $Q_\alpha$ for even-even isotopes, obtained in the framework of the macro-microscopic theory [3,4]. The black symbols denote the experimental $Q_\alpha$ values; the points – even-odd nuclei, squares – even-even nuclei. The dashed line is drawn through the experimental points to guide the eye.

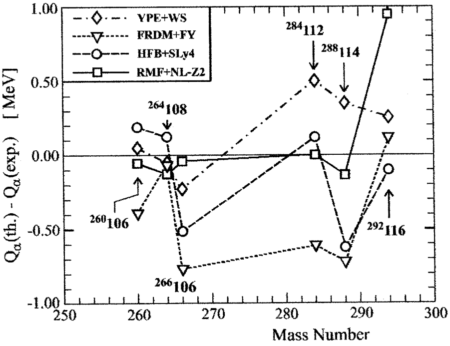

Figure 11. The difference $\Delta Q_\alpha$ = $Q_\alpha$(th) - $Q_\alpha$(exp) for the heaviest even-even nuclei with Z = 106 – 114. The line through the points correspond to calculations performed in different models (indicated in the Figure).

This paper presents the results obtained by a large group of physicists, many of them are co-authors of the original publications. The experiments were performed at the heavy ion accelerator complexes of GSI (Darmstadt) and FLNR (Dubna) in collaboration with LLNL (Livermore), RIKEN (Saitama), the Institute of Physics and Department of Physics of the Comenius University (Bratislava) and the Department of Physics of the University of Messina.

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Синтез тяжелейших элементов и свойства их распада

Синтез и исследование радиоактивных свойств новых элементов рассматриваются в плане существования «островов стабильности» гипотетических сверхтяжелых ядер, предсказанных теорией более 35 лет тому назад. Экспериментальные данные, демонстрирующие повышенную стабильность ядер вблизи деформированных оболочек $Z = 108$, $N = 162$, обсуждаются в плане продвижения к более тяжелым и значительно более стабильным нуклидам вблизи предсказанных сферических оболочек $Z = 114 - 122$ и $N = 184$, следующих за дважды магическим ядром $^{208}$Pb. Приводятся результаты опытов по синтезу изотопов 114 и 116 элементов в реакциях слияния с ионами $^{48}$Ca. Показано, что в этих реакциях наблюдаются цепочки распада отдельных атомов, состоящие из последовательных $\alpha$-распадов и оканчивающиеся спонтанным делением. Энергии и вероятности распадов сравниваются с предсказанием различных теоретических моделей, описывающих структуру тяжелых ядер. Полученные результаты рассматриваются как первое экспериментальное указание существования областей стабильности сверхтяжелых ядер, значительно расширяющих предел существования химических элементов.

Работа выполнена в Лаборатории ядерных реакций им. Г.Н.Флерова ОИЯИ.

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Oganessian Yu.Ts. E7-2000-259

The Synthesis and Decay Properties of the Heaviest Elements

The synthesis and the study of radioactive properties of new elements is considered with respect to the existence of the «islands of stability» of hypothetical superheavy elements predicted by the theory more than 35 years ago. Experimental data demonstrating an enhanced stability of nuclei in the vicinity of deformed shells with $Z = 108$ and $N = 162$ is discussed from the point of view of advent into more heavy and much more stable nuclides near the predicted spherical shells $Z = 114 - 122$ and $N = 184$ following after the doubly magic nucleus $^{208}$Pb.

The author presents the results of experiments on the synthesis of isotopes of elements 114 and 116 in the fusion reactions with $^{48}$Ca. In these reactions the decay chains of heavy atoms consisting of sequential $\alpha$-decays interrupted by spontaneous fission have been observed.

The decay energies and probabilities are compared with predictions of different theoretical models describing the structure of heavy nuclei. The obtained results are considered as the first experimental evidence of the existence of domains of stability of superheavy nuclei which substantially extends the boundaries of existence of chemical elements.

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

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