DISCOVERY OF THE ISLAND OF STABILITY FOR SUPERHEAVY ELEMENTS

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Abstract

The existence of a domain of hypothetical Super Heavy Elements (SHE) forming a region (island) with high stability in the vicinity of the doubly magic nucleus $^{298}114$ was postulated in the mid-1960s. For more than 30 years, scientists searched hard for naturally occurring SHEs and unsuccessfully attempted to synthesize them using heavy ion accelerators. Over the past 15 years a breakthrough in heavy-element synthesis has been achieved, using rare †actinide targets irradiated with ^{48}Ca beams. More than 52 neutron-rich nuclei including the isotopes of the new element 113-118 and their α-decay products where synthesized for the first time.

INTRODUCTION

The first ideas of the form of atomic nucleus and properties of nuclear matter in the framework of the planetary model of atom (E. Rutherford, 1911) were formulated in a macroscopic concept, the so-called liquid-drop nuclear model (G. Gamow, 1928 [1]). In this approach, the nucleus is considered a drop of a liquid with great density ($2.5\cdot10^{14}$ g/cm^3), that keeps practically all the mass and the whole positive charge of the atom. Stability of extremely heavy nuclei is determined by spontaneous fission. Half-life with respect to spontaneous fission of the isotope ^{238}U (fission barrier height $B_f\approx6$ MeV), is 10^{16} years (G. Flerov and K. Petrzhak, 1940 [2]). Note, in absence of fission barrier ($B_f\approx0$) such an extremely heavy nucleus would undergo fission in a time $T_{SF}\approx10^{-19}$ S. The difference is more than 42 orders of magnitude! According to liquid-drop model the fission barrier practically wash out for elements with Z≥100 [3].

In general, despite the success of the charge liquid drop model in describing the nuclear properties it has been noticed for long that the energies of the nuclei in the ground and strongly deformed states obtained in the experiments somewhat deviate from the calculations. These deviations are regular and thus point to the presence of structure of nuclear matter. Taking into account the structure of nuclear matter, first by introducing a correction to the potential energy of the nuclear drop macro-microscopic model (MMM), and later in purely microscopic model calculations, such as Hartree-Fock-Bogoliubov (HFB) and Relativistic mean field (RMF) m†ade in the 60-ies of the last century allowed to eliminated these disagreements. At the same time, the limits of nuclear masses predicted by new theory were quite unexpected.

One of the fundamental outcomes of the nuclear shell model is a prediction of new closed shells in very heavy (superheavy) nuclei that cannot exist in liquid-drop model (Fig. 1). It turned out that upon approach to closed sphere

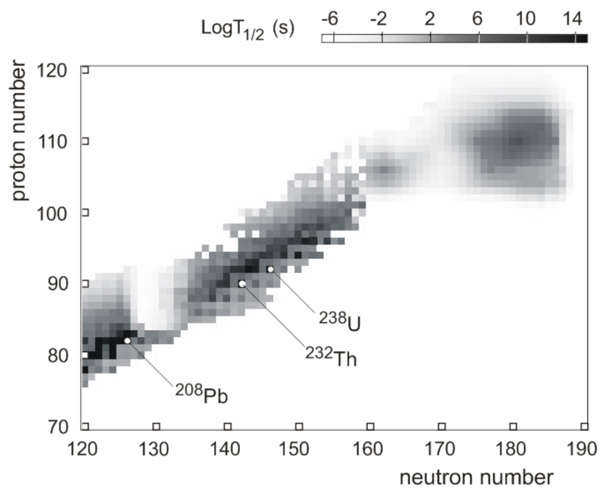

Figure 1: Part of the nuclear map. Black squares up to the ^{208}Pb are stable nuclei. Over ^{209}Bi, the lifetime of the nuclei sharply decreases, but increases again in the isotopes Th and U to values comparable with the age of the earth. In the meanwhile a sharp drop in the stability is observed again for trans-uranium elements with the increase in the atomic number. However, according to the predictions of modern theory, in the region of very heavy (superheavy) elements, there may appear a region (an island) of sufficiently long-lived nuclei.

cal shells Z=114 (not excluded also 120, 122 and 126) and N=184 next to the doubly magic nucleus ^{208}Pb, a strong increase in nuclear stability should be observed [4]. The new nuclei in the vicinity of the closed shells have large neutron excess. In the chart of the nuclides they form a vast area – the "island of stability" of the isotopes of superheavy elements that are characterized by very high stability to various types of decay.

REACTIONS OF SYNTHESIS

Verification of theory about possible existence of such an "island" is very difficult first of all because of the necessity of synthesizing the nuclei with such a large mass and substantial excess of neutrons. Known methods of production of artificial elements heavier than uranium do not allow reach the "island". As it is seen in Fig. 2, reactions

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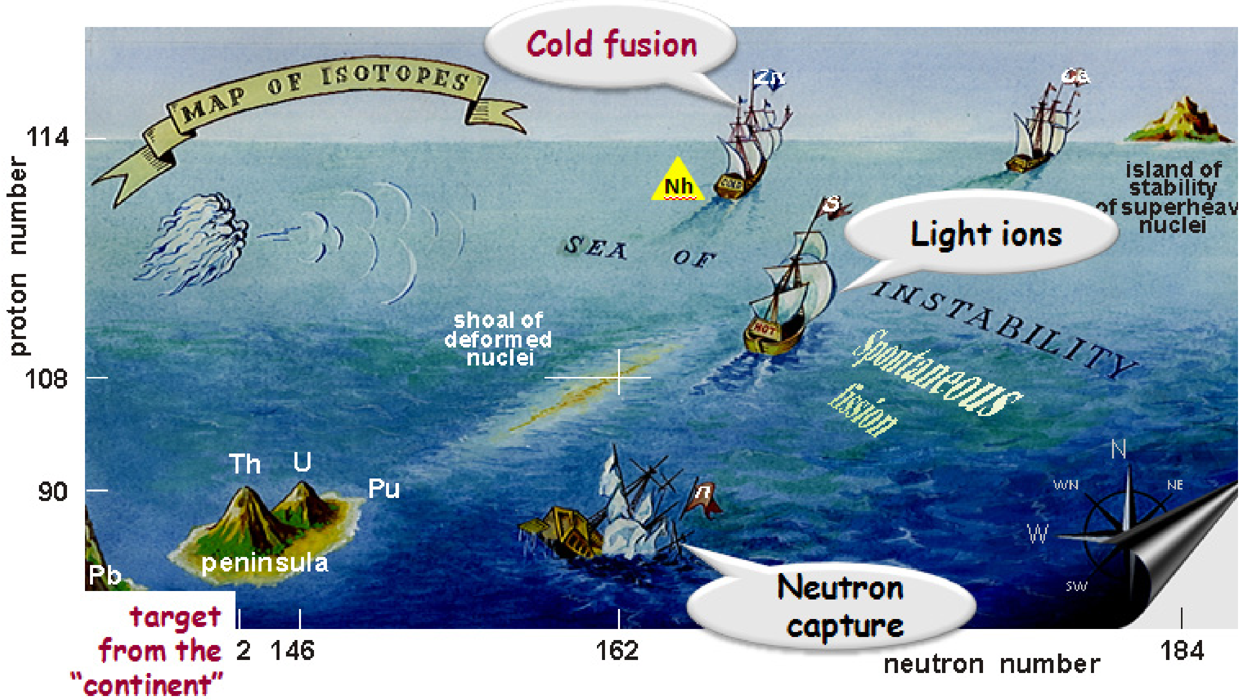

Figure 2: Reactions of synthesis.

of multiple neutron capture, even in extremely high neutron flux from nuclear explosion, do not allow shift from ^{238}U to the neutron excess in uranium isotopes by more than 20 mass units. Reactions of target nuclei of uranium and its close neighbours with lighter ions – isotopes of C-Ne – allow cover only half a way to the "island". Along with this, reactions of fusion of the target nuclei ^{208}Pb/^{209}Bi with massive projectiles up to ^{70}Zn result in formation of the isotopes with Z≤113 but they have large deficit of neutrons.

Nuclei produced in such reactions appear to be out of effect of the shell N=184; they have very short half-lives: $T_{1/2}=10^{-4}–10^{-3}$ s and produced with extremely small cross sections. A larger excess of neutrons could be expected in the reactions of incomplete fusion. But the transfer reactions of large clusters (A> 50) in the interactions of extremely heavy nuclei: ^{238}U + ^{238}U or ^{238}U + ^{248}Cm have very small cross sections. We returned again to the complete fusion but chose reactions with maximum available neutron excess both in target and projectile nuclei.

In this approach, the heaviest nuclei and their decay products were synthesized using as target material the accessible neutron-rich isotopes of artificial elements such as ^{244}Pu, ^{248}Cm, ^{249}Bk or isotopes $^{249-251}$Cf produced in high-flux nuclear reactors. Rare and very expensive isotope ^{48}Ca (with abundance of 0.19% in natural composition) was used as projectile with energy near the height of the Coulomb barrier ($E_{LAB}\approx$ 5.0 - 5.5 MeV·A).

Superheavy nuclides, the isotopes of elements 112-118 and their decay products, were detected using kinematic gas-filled recoil separator placed at an angle $\theta=0^0$ to the beam direction (Fig. 3). Recoiling nuclei produced in

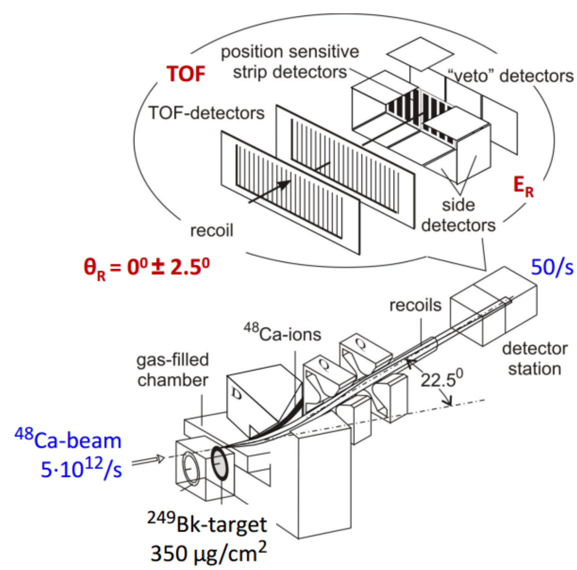

Figure 3: Kinematic recoil separator used for synthesis of SHE. The recoil products emitted from the target continue to move in hydrogen medium at a pressure of about 1torr. Separation of reaction products occurs in a magnetic field. The distance from the target to the focal detectors is 4 m; the time of flight this distance by the heavy nucleus is about 1μs.

complete fusion reactions are separated from the ^{48}Ca beam-particles and other unwanted reaction products by their kinematic characteristics (emission angle and kinematic energy) over time of flight from target to detectors that is about 1μs [5]. Detectors mounted in the focal plane

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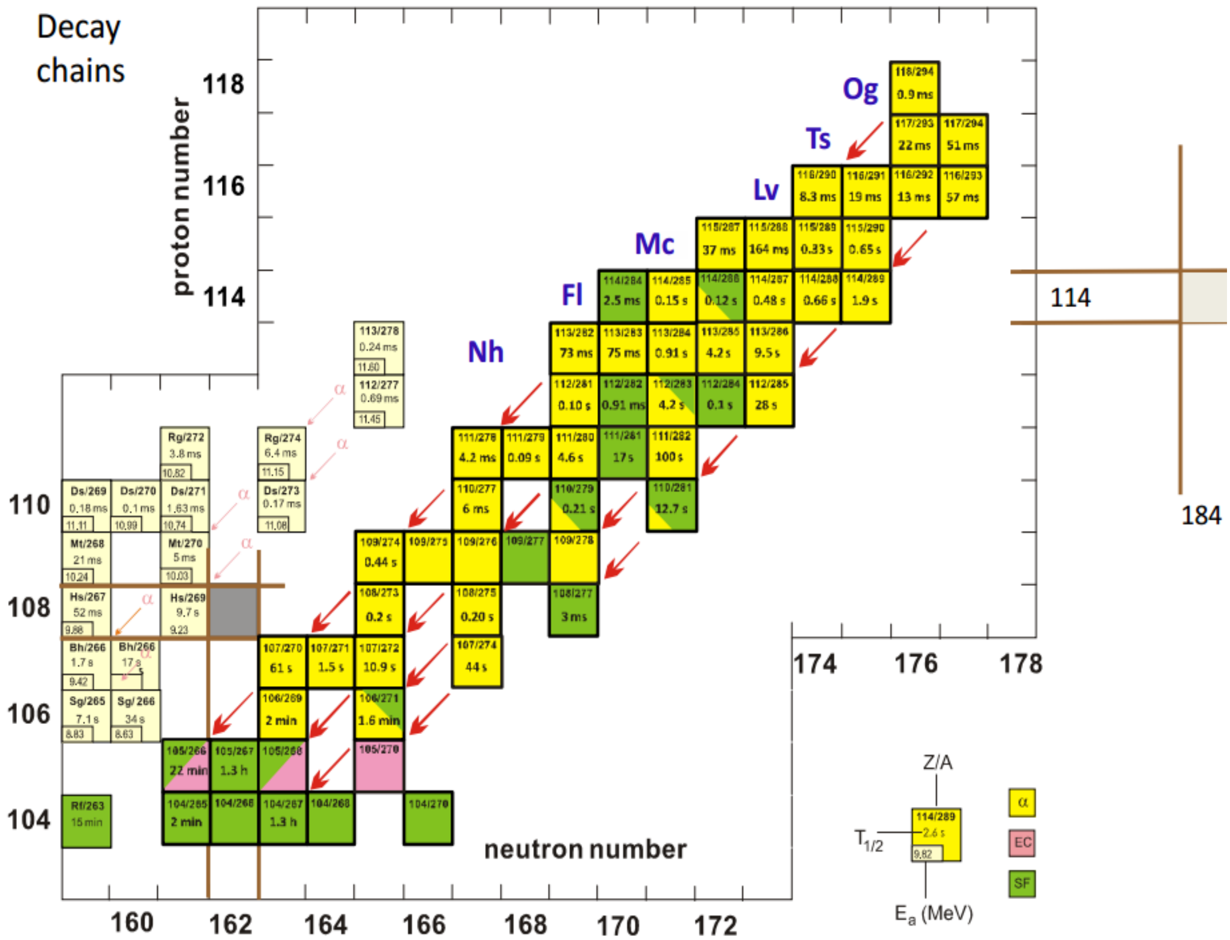

Figure 4: Decay chains of SHE obtained in the fusion reactions of ^{238}U, $^{239-244}$Pu, ^{243}Am, $^{245.248}$Cm, ^{249}Bk and ^{249}Cf nuclei with ^{48}Ca nuclei. Each chain consists of successive alpha transitions and ends with spontaneous fission. For each nucleus in the chain, the type and probability of decay (half-life) are indicated. The measured experimental values of the alpha decay energies and total kinetic energies of the fission fragments are not given so as not to load the graph.

of the separator determine position of the implanted recoils and register its alpha-decay or spontaneous fission (SF). From calculations in MM-model is expected, successive alpha-decays (one or several) should end in spontaneous fission. Using coordinate signals from the focal plane detector, it is possible to identify with high reliability such radioactive family originating from the decay of the implanted super heavy nucleus.

Amid all the reaction channels, the fusion of the complex nuclei involving considerable restructuring of a 300-nucleon system of two contacting nuclei into a single compound nucleus is a complex and rare process. To this one should add that on the second stage that is cooling of the compound nucleus excited up to energy of 40 MeV via emission of 3-4 neutrons fission strongly competes with neutron emission. As a result, new nucleus (element) is formed with a cross section of about 1pb (10^{-36} cm^2) that is ~10^{-12} of the total reaction cross section. This determines extremely rare registration of the events of formation of new element: from one per day to one per month.

DECAY PROPERTIES

As can be seen in Fig. 4, new nuclides indeed undergo successive α-decays that end in spontaneous fission [6-8]. Decay of even-even nuclei results in rather short chains while nuclei with odd number of protons and/or neutrons show longer chains, as spontaneous fission is hindered due to the absence of pairing effect in the fissioning nucleus. In experiment, for every alpha-transition the energy and time of alpha-decay were measured together with coordinates; as mentioned above, the latter were used to establish genetic links in the radioactive family of a superheavy nucleus. Despite low formation cross section of the superheavy nuclides, the probability of random correlation of signals that could imitate a decay chain, even for a single event, is 10^{-11} to 10^{-3} depending on chain length and measuring conditions.

Finally, in reactions of fusion of target nuclei – a variety of isotopes of elements ranging from uranium to californium with nuclei of ^{48}Ca, seven elements with Z=112-118 located on the edge but already in the area of a hypothetical “stability island” were synthesized. Decay properties of the heaviest nuclei (together with their decay products, 52 new nuclides with Z=104-118 and N=169–177 were observed, see Fig. 4) can be compared to predictions of the macroscopic-microscopic theory.

From the experimental data we see that having moved 86 mass units away from the last doubly magic nucleus ^{208}Pb towards the heaviest nuclei we observe their amazing survivability. In very heavy nuclei at the limit of the existence of the strongly charged nuclear matter approaching magic number of protons and neutrons due to the effect of new shells the potential energy in the ground-state of the heaviest nuclei decreases and there appears the fission barrier that makes possible the existence of superheavy elements. This pushes the limit of nuclear masses to the region A>300 and considerably expands the limits of the existence of chemical elements.

Fundamental predictions of the microscopic theory concerning the existence of the superheavy nuclides have got first experimental confirmation.

CHEMISTRY OF NEW ELEMENTS

Another problem is associated with the electron structure of the superheavy atoms.

To what extent the chemical behaviour of the super heavy elements whose atomic number has reached today 118 and will most likely increase further, follows their light homologues? The answer to this question is one of actual problems of atomic physics. One has to pay attention to the fact that the expected change of the electron energy with increased atomic number (Moseley's law) can be disrupted by the simultaneously increased contribution of the so-called "relativistic effect" associated with approaching of the speed of orbital electrons to the speed of light. In the limiting case, the change of the electronic structure of superheavy atoms (relativistic contraction) in elements with $Z \geq Z_{crit}$ may lead to violation of the law of periodicity of chemical properties established by D.I. Mendeleev in 1869.

Relatively long lifetime of several isotopes of superheavy elements allowed us to start investigation of their chemical properties. By now, first studies of formation of intermetallic compounds of elements 112 and 114 with atoms of gold: [Cn·Au] and [Fl·Au] were carried out and their properties could be compared with the known data for [Hg·Au] and [Pb·Au], respectively [9,10].

The experimental values of adsorption enthalpy from which the enthalpy of sublimation and then boiling point was calculated, show a sharp increase in volatility when going from lighter to heavier homologies in pairs Hg/Cn and Pb/Fl respectively. Such a strong change in the physical and chemical properties that relate to groups 12 and 14 of the table of elements leave open the question of the chemical behaviour of element 118 as member of group 18 of the noble gases [11].

Further progress of work on synthesis of superheavy elements is linked with the development of the new experimental complex – "Factory of Superheavy Elements". Putting in operation of the new heavy-ion accelerator, increase of production of target materials at the high-flux reactor, implementation of new technologies and development of new experimental equipment will allow for the SHE-Factory to increase the experimental sensitivity by about a factor of 100 times.

Experiments aimed at the synthesis of the superheavy elements were carried out employing the heavy-ion cyclotron of the Flerov Laboratory of Nuclear Reactions (JINR) in collaboration with Livermore National Laboratory (LLNL, USA), Oak Ridge National Laboratory (ORNL, USA), Institute of the Atomic Reactors (IAR, Russia) Vanderbilt University (USA) and Paul Scherrer Institute (PSI, Switzerland).

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