

Status and prospects of synthesizing superheavy elements

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Abstract. The nuclear shell model predicts that the next doubly magic shell-closure beyond ^{208}Pb is at a proton number between $Z = 114$ and 126 and at a neutron number $N = 184$. The outstanding aim of experimental investigations is the exploration of this region of spherical “Superheavy Elements”. This article describes the experiments that were performed at the GSI SHIP. They resulted in an unambiguous identification of elements 107 to 112. They were negative so far in searching for elements 113, 116 and 118 at SHIP; however, positive results were reported from experiments in Dubna on elements 114 and 116 and from experiments in Berkeley on element 118. The measured decay data are compared with theoretical predictions. Some aspects concerning the reaction mechanism and the use of radioactive beams are also presented.

PACS. 21.10.Dr Binding energies and masses – 23.60.+e Alpha decay – 25.70.-z Low and intermediate energy heavy-ion reactions – 25.85.Ca Spontaneous fission

1 Introduction

In recent years the exploration of superheavy elements (SHE) received increasing interest both from theoretical and experimental investigation and both from chemical and physical studies. The reasons for the activity awakened again are based mainly on technical developments in the field of computer power, accelerator techniques and detection sensitivity.

Using faster and bigger computers the *properties* of heavy nuclei are studied from multi-dimensional macroscopic-microscopic calculations, self-consistent Skyrme-Hartree-Fock and relativistic mean-field models. The results reveal a rather complex structure of shell effects which determine the stability of nuclei in the superheavy region.

The most difficult problem, however, which is awaiting a theoretical solution, is the understanding of the *synthesis* of superheavy nuclei. The calculation of the involved dynamical processes on a microscopic level is presently the most challenging and work intensive task.

Successful methods for the laboratory synthesis of heavy elements have been fusion-evaporation reactions using heavy-element targets; recoil-separation techniques; and the identification of the nuclei by generic ties to known daughter decays after implantation into position-sensitive detectors. Experiments at low cross-sections necessitate projectile beams of high intensity and stability. Although the intensity limits have not presently been

reached, considerable improvements in accelerator techniques have been made in recent years.

In the following section the results will be discussed from theoretical descriptions of the nuclear ground-state properties. A more detailed description is given of studies of elements 110 to 112 measured at GSI Darmstadt [1]. In addition, a brief overview is given of results obtained at facilities in other laboratories where experiments for the investigation of heavy elements are being performed. The recent results obtained at JINR, Dubna, from studies of hot-fusion reaction producing nuclei up to element 116 are described in more detail elsewhere and in other contributions to this conference [2,3]. Also reported is the present (August, 2001) status of the results obtained from experiments at LBNL, Berkeley, on the synthesis of element 118 using cold fusion [4,5]. Nuclear reactions are discussed for the synthesis of SHEs using stable and radioactive beams. Finally, a summary and an outlook are given.

2 Nuclear structure and decay properties

The basic step which is necessary for the determination of the stability of SHEs is the calculation of the ground-state binding energy. As a signature for shell effects, we can extract from various models the shell-correction energy by subtracting a smooth macroscopic part (derived from the liquid-drop model) from the total binding energy. In macroscopic-microscopic models the shell-correction energy is of course the essential input value which is calculated directly from the shell model. The shell-correction

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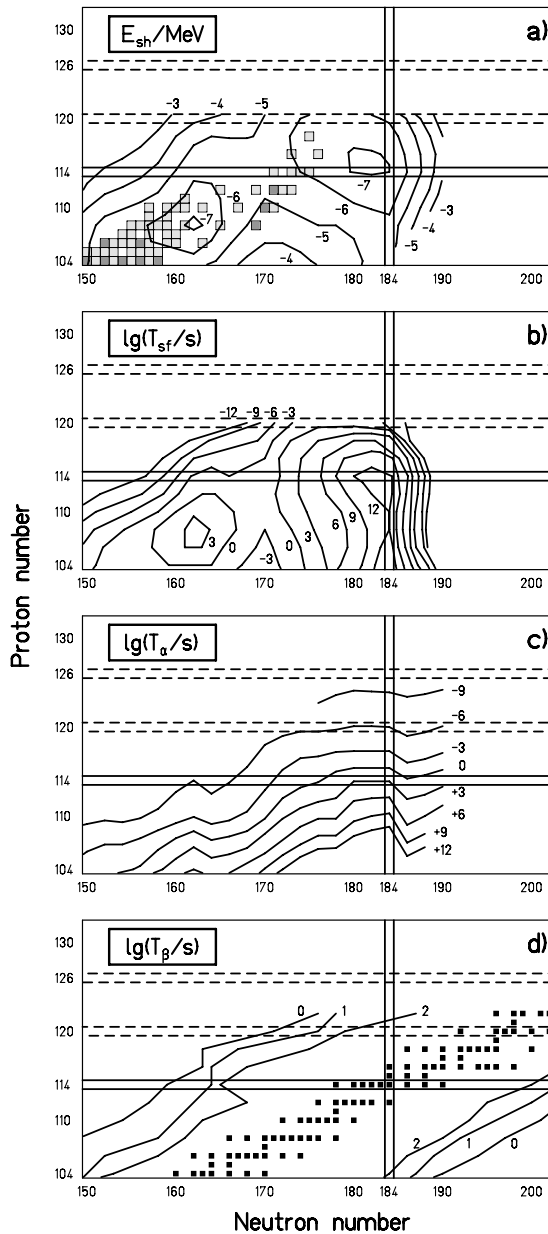


Fig. 1. Shell-correction energy (a) and partial spontaneous fission, α and β half-lives (b-d). The squares in (a) mark the nuclei presently known or under investigation.

energy is plotted in fig. 1a using the data from ref. [6]. Two equally deep minima are obtained, one at $Z = 108$ and $N = 162$ for deformed nuclei with deformation parameters $\beta_2 \approx 0.22$, $\beta_4 \approx -0.07$ and the other at $Z = 114$ and $N = 184$ for spherical SHEs. Different results are obtained from self-consistent Hartree-Fock-Bogoliubov (HFB) calculations and relativistic mean-field models [7–10]. They predict for the spherical nuclei shells at $Z = 114$, 120 or 126 (indicated as dashed lines in fig. 1a) and $N = 184$ or 172, with shell strengths being also a function of the amount of nucleons of the other type.

For the calculation of partial spontaneous fission half-lives the knowledge of ground-state binding energies is

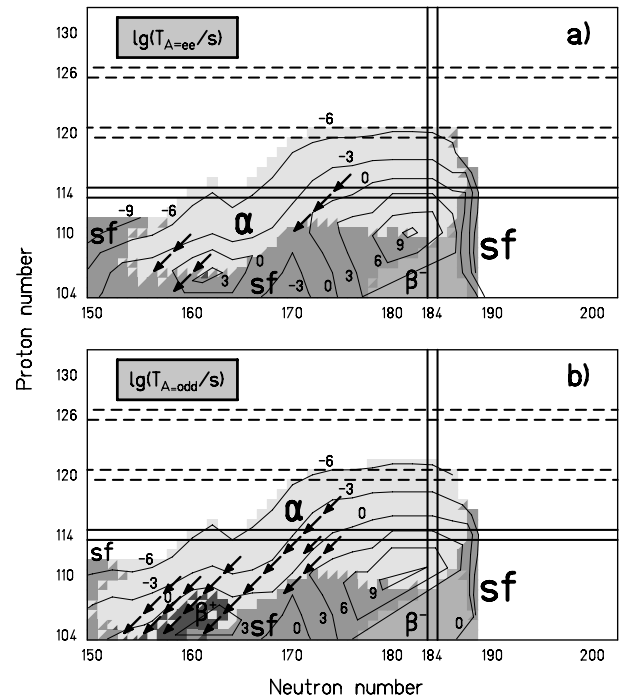


Fig. 2. Dominant half-lives for α , β^+ /EC, β^- -decay and spontaneous fission for even-even mass nuclei in (a) and for odd mass nuclei in (b). The arrows mark decay chains presently known or under investigation.

not sufficient. It is necessary to determine the fission barrier over a wide range of deformation. The most accurate predictions were obtained for even-even nuclei using the macroscopic-microscopic model [6, 11]. Partial spontaneous fission half-lives are plotted in fig. 1b. The landscape of fission half-lives reflects the landscape of shell-correction energies, because in the region of SHEs the height of the fission barrier is mainly determined by the ground-state shell-correction energy. The contribution from the macroscopic liquid-drop part approaches zero. Nevertheless we see a significant increase of fission half-life from 10^3 s for deformed nuclei to 10^{12} s for spherical SHEs. This difference is arising from the width of the fission barrier which becomes wider in the case of the spherical nuclei.

Partial α half-lives decrease almost monotonically from 10^{12} s down to 10^{-9} s near $Z = 126$ (fig. 1c). The valley of β -stable nuclei (marked by black squares in fig. 1d) passes through $Z = 114$, $N = 184$. At a distance from the bottom of the valley, the β half-lives decrease gradually down to values of one second.

The dominating partial half-life is shown in fig. 2a for even-even nuclei. The two regions of deformed heavy nuclei near $N = 162$ and spherical SHEs merge and form a region of α emitters surrounded by fissioning nuclei. The longest half-lives are 1000 s for deformed heavy nuclei and 30 y for spherical SHEs. It is interesting to note that the longest half-lives are not reached for the doubly magic nucleus ${}_{184}^{298}114$, but for $Z = 110$ and $N = 182$. This is a result of the continuously increasing Q_α -values with in-

creasing element number. Therefore, the α -decay becomes the dominant decay mode beyond element 110 with continuously decreasing half-life. The half-lives of nuclei at $N = 184$ and $Z < 110$ are reduced from β^- -decay.

The four-member decay chain of $^{292}116$, the heaviest even-even nucleus, observed in recent experiments in Dubna [3, 12], is also drawn in fig. 2a. The arrows follow approximately the 1 s contour line down to $^{280}110$, which is, in agreement with the experiment, predicted to be a spontaneously fissioning nucleus. The experimental half-lives are 53 ms, -2.6 s, -45 s, -7.6 s, respectively, which are longer than the calculated values by a factor of 10 on average. However, this deviation is well within the accuracy limits of the calculation. *E.g.*, a change of the α energy of $^{288}114$ by 350 keV only changes the half-life by a factor of 10. The decay chains of two other recently synthesized even-even nuclei, $^{270}110$ [13] and ^{270}Hs [14], are also drawn in the figure. In these cases the decay chains end by spontaneous fission at ^{262}Sg and ^{262}Rf , respectively.

In the case of odd nuclei (fig. 2b), the α and fission half-lives calculated by Smolanczuk and Sobiczewski [6, 11] were multiplied by a factor of 10 and 1000, respectively, thus making provisions for the odd-particle hindrance factors. However, we have to keep in mind that the fission hindrance factors have a wide distribution from 10^1 to 10^5 , which is mainly a result of the specific levels occupied by the odd nucleon. For odd-odd nuclei (not shown here), the fission hindrance factors from both the odd proton and the odd neutron are multiplied. The β half-lives given by Möller *et al.* [15] were divided by 10, because first-forbidden transitions were not taken into account in these calculations (see [16] and discussion therein). For the odd and odd-odd nuclei, the island character of the α emitters disappeared and α -decay could propagate down to rutherfordium and beyond. In the allegorical representation where the stability of SHEs is seen as an island in a sea of instability, the even-even nuclei portray the situation during a flood, the odd nuclei during an ebb, when the island is connected to the mainland.

The decay chains of the recently measured even-odd nuclei are also drawn in the figure: $^{277}112$, GSI [1, 17], $^{287}114$ and $^{289}114$, JINR [18], and $^{293}118$, LBNL [4]. Here again, the measured data are predominantly well duplicated in the calculations. The assignment of the three chains observed in Berkeley finds particularly strong support in the excellent agreement with the predicted half-lives or vice versa, if the experimental result will eventually be confirmed (see also subsect. 3.2). The same arguments hold for the first observation of element 114 in Dubna by the decay chain from $^{289}114$ [3, 19].

The interesting question arises, if and how the uncertainty related with the location of the proton and neutron shell closures will change the half-lives of SHEs. Partial α and β half-lives are only insignificantly modified by shell effects, because the decay process occurs between neighboring nuclei. This is different for fission half-lives which are primarily determined by shell effects. However, the uncertainty related with the location of nuclei with the

strongest shell effects and thus longest partial fission half-life at $Z = 114, 120$ or 126 and $N = 172$ or 184 , is inconsequential concerning the longest “total” half-life of SHEs. The regions for SHEs in question are dominated by α -decay. And α -decay will be modified by only a factor of up to approximately 100, if the double shell closure is not located at $Z = 114$ and $N = 184$.

The line of reasoning is, however, different concerning the production cross-section. The survival probability of the compound nucleus (CN) is determined among other factors significantly by the fission barrier. Therefore all present calculations of cross-sections suffer from the uncertainty related with the location and strength of closed shells. However, it may also turn out that shell effects in the region of SHEs are distributed across a number of subshell closures. In that case a wider region of less deep shell-correction energy would exist with corresponding modification of stability and production yield of SHEs.

3 Experimental results

3.1 The new elements 110 to 112

In this section, we present results dealing with the discovery of elements 110 to 112. Detailed presentations of the properties of elements 107 to 109 and of elements 110 to 112 were given in previous reviews [20, 21].

Element 110 was discovered in 1994 using the reaction $^{62}\text{Ni} + ^{208}\text{Pb} \rightarrow ^{270}110^*$ [22]. A total of four decay chains were observed. The main experiment was preceded by a thorough study of the excitation functions for the synthesis of ^{257}Rf and ^{265}Hs in order to determine the optimum beam energy for the production of element 110. The data revealed that the maximum cross-section for the synthesis of element 108 was shifted to a lower excitation energy, different from the predictions of reaction theories.

The isotope $^{271}110$ was synthesized with a beam of the more neutron-rich isotope ^{64}Ni [21]. The important result for the further production of elements beyond meitnerium was that the cross-section was enhanced from 3.5 pb to 15 pb by increasing the neutron number of the projectile by two, which gave hope that the cross-sections could decrease less steeply with more neutron-rich projectiles.

Two more isotopes of element 110 have been reported in the literature. The first is $^{267}110$, produced in the irradiation of ^{209}Bi with ^{59}Co [23, 24]. The second isotope is $^{273}110$, reported to be observed in the irradiation of ^{244}Pu with ^{34}S after the evaporation of five neutrons [25, 26]. Both observations need further experimental clarification.

The even-even nucleus $^{270}110$ was synthesized using the reaction $^{64}\text{Ni} + ^{207}\text{Pb}$ [13]. A total of eight α -decay chains was measured during an irradiation time of seven days. Decay data were obtained for the ground state and a high-spin K isomer, for which calculations predict spin and parity 8^+ , 9^- or 10^- . The new nuclei ^{266}Hs and ^{262}Sg were identified as daughter products after α -decay. Spontaneous fission of ^{262}Sg terminates the decay chain.

Element 111 was synthesized in 1994 using the reaction $^{64}\text{Ni} + ^{209}\text{Bi} \rightarrow ^{273}111^*$. A total of three α chains of the isotope $^{272}111$ were observed [27]. Other three decay chains were measured in a confirmation experiment in October 2000 [28].

Element 112 was investigated at SHIP using the reaction $^{70}\text{Zn} + ^{208}\text{Pb} \rightarrow ^{278}112^*$ [17]. The irradiation was performed in January-February 1996. Over a period of 24 days, a total of 3.4×10^{18} projectiles were collected. Two α -decay chains were observed resulting in a cross-section of 1.0 pb. They were assigned to the one-neutron emission channel.

In May 2000 an experiment was performed aiming at confirming the synthesis of $^{277}112$. During an irradiation time of 19 days a total of 3.5×10^{18} projectiles was collected. One more decay chain was observed [1]. The measured decay pattern is in agreement with the one observed for chain two in the first experiment.

A new result was occurrence of fission which ended the third decay chain at ^{261}Rf . Spontaneous fission of this nucleus was not yet known. In the second chain α energy and lifetime of the last measured decay agreed well with the literature data for ^{257}No ; however, a new α transition of 8.52 MeV with a lifetime of 4.7 s was measured for ^{261}Rf , whereas the literature data give 8.28 MeV and $\tau = 94$ s. In the first chain agreement with the literature was measured for ^{265}Sg ; however, the α -particle of the daughter ^{261}Rf escaped, which prevents from a direct comparison with the decay data of this nucleus from chain two.

However, the new data on the decay of ^{261}Rf , obtained from only three decay chains of $^{277}112$, and the correctness of the assignment was proven in a recent chemistry experiment [14], in which ^{269}Hs was directly produced using the reaction $^{26}\text{Mg} + ^{248}\text{Cm} \rightarrow ^{274}\text{Hs}^*$. After separation of HsO_4 from the other reaction products, a total of five decay chains was measured. A spontaneous fission branching of ^{261}Rf was deduced, and also the α energy of 8.52 MeV was confirmed.

3.2 The reaction $^{86}\text{Kr} + ^{208}\text{Pb} \rightarrow ^{294}118^*$ studied at the Berkeley gas-filled separator and at the GSI SHIP

The BGS became ready for the first experiments by the year 1998. In the autumn of the same year, Smolanczuk [29] was working on a theoretical study of the "Production Mechanism of Superheavy Nuclei in Cold-Fusion Reactions." Using a relatively simple fusion model, he reproduced the measured formation cross-sections of deformed heavy nuclei synthesized in cold-fusion reactions from No, $\sigma = 260$ nb, to $Z = 112$, $\sigma = 1$ pb. The same model, applied for the calculation of cross-sections for the synthesis of spherical superheavy nuclei resulted in the unusually high value of 670 pb for the reaction $^{86}\text{Kr} + ^{208}\text{Pb} \rightarrow ^{293}118 + 1n$.

In order to prove the validity of the theoretical prediction for the synthesis of element 118, an irradiation was performed for eleven days in April-May, 1999, at the new BGS. The fact that the beam of noble gas krypton could

easily be produced from an ECR-ion source available at the cyclotron was particularly helpful.

Three event chains, consisting of an implanted evaporation residue and of subsequently emitted α -particles, were observed [4]. The resulting cross-section was 2.2 pb. This was an unexpected high value when compared with the measured, continuously decreasing yields up to element 112 known at the time; however, when compared with the theoretical prediction, it was much smaller than expected.

In order to confirm the data obtained in Berkeley, the same reaction was investigated at SHIP in the summer of 1999. The experiment is described in detail in [1]. During a measuring time of 24 days a beam dose of 2.9×10^{18} projectiles was collected which is comparable to the Berkeley value of 2.3×10^{18} . No event chain was detected similar to those observed in Berkeley. The cross-section limit resulting from the SHIP experiment was 1.0 pb.

Although the Berkeley data on the synthesis of $^{293}118$ could not be proved, the negative result does not disprove those data. Several reasons could plausibly explain the difference (see [1]), among which statistical fluctuations would be the simplest interpretation. A probability analysis based solely on statistical distributions still gives a probability of 17% that, in two experiments at approximately the same beam dose, three events are observed in one case and zero in the other.

Efforts to confirm the synthesis of element 118 were also made at the accelerator laboratories GANIL in France [30] and RIKEN in Japan [31]. No positive event could be reported.

Further attempts to prove the 1999 data were made by repeating the experiment at the BGS itself. The experiments were performed at the end of 2000 and in April-May 2001. In the new experiments no more decay chain was observed. The re-analysis of the 1999 data files led to unforeseen problems so that the earlier data could not be reproduced. A retraction of the earlier published results was submitted on July 26, 2001 [5].

The new information from Berkeley on the status of their element 118 experiments was not yet available during the conference at the beginning of July, however, it was distributed before the deadline for the written contributions. Therefore I could include the news in my paper, and I would like to comment it briefly.

From the negative results, which were obtained from the recent irradiation in Berkeley, I conclude that they do not disprove the first result. It is extremely difficult, if not presently impossible under acceptable economical conditions, to clearly disprove results measured at the limits of detectability in the case that experimental parameters like beam energy and purity of the separator gas-filling cannot be identically reproduced. Some other reasons are listed in our recent review article [1]. Retracting doubtful results is honorable, because it clears the way for further investigation and the possibility of unambiguous discovery. However, as long as no clear reason can be given that and why the first result was erroneous, it will continue to exist as possibility of being correct. This is especially the

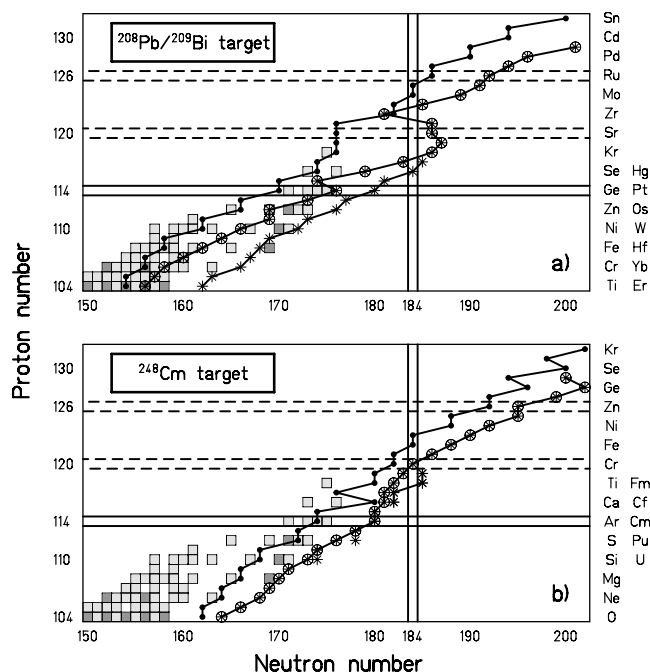


Fig. 3. Compound nuclei produced in reactions using stable and radioactive beams and targets (see text for an explanation of the symbols).

case when the data do not sound unreasonable. For these reasons I kept the Berkeley decay chain in figs. 1 to 3 as a case which must be definitely confirmed or definitely disproved, although the claim for discovery was retracted.

4 Nuclear reactions

Compound nuclei that could be produced concerning the availability of beams and targets are plotted in fig. 3. The graphs also demonstrate the extension in the region of SHEs which will become possible with radioactive beams. The nuclei presently known or under investigation are marked by squares.

In fig. 3a the curve marked with dots (●) shows the most neutron-rich CN that can be produced with ^{208}Pb or ^{209}Bi targets and beams of the most neutron-rich stable isotopes of the elements from Ti to Sn, given in the first column at the right ordinate. The double magic SHEs could be reached only if located at $Z = 126$ and $N = 184$.

The accessible region is extended by 4 to 5 neutrons to the right using radioactive isotopes of the elements from Ti to Sn (circled asterisk in fig. 3a). As possible most neutron-rich radioactive projectiles those isotopes were taken into account that could be produced with intensities of at least $10^9/\text{s}$ according to the data presented in the RIA proposal [32].

Striking is the wide extension of possible CN to the neutron-rich side at $Z = 120$ using beams of Kr, Rb and Sr. The reason is that these nuclei are available as fission fragments and can be accelerated with high yield, too. SHEs both at $Z = 120$ and 126 are well covered using reactions with Pb and Bi targets and radioactive beams.

More neutron-rich nuclei of elements below $Z = 118$ can be produced using the radioactive beams of ^{96}Kr or ^{98}Rb and targets of stable neutron-rich isotopes of elements below Pb from Hg down to Er (second column on the right ordinate, curve marked with symbol *). In these reactions evaporation residues could be produced in a region, where the new chains from element 114, 116 and 118 are ending presently in the unknown. In this region α -decay and spontaneous fission are expected with half-lives from seconds to hours (see fig. 2).

In the lower plot, fig. 3b, the equivalent combinations are given for reactions using a ^{248}Cm target and stable and radioactive beams. Concerning the location of CN, no significant extension into the direction of neutron-rich SHEs results compared with the reactions given in fig. 3a. Apparent from the graph (fig. 3b) is the extraordinary use of ^{48}Ca for the synthesis of neutron-rich SHEs. The surplus of neutrons beyond ^{48}Ca can be only slightly increased using radioactive beams.

Radioactive beams of ^{47}K and ^{46}Ar are likely produced with high yield. Using these beams and in principle feasible actinide targets from U to Fm (the elements are given in the second column at the right ordinate) the surplus of neutrons can be further increased (CN marked by symbol *). However, selecting a reaction using actinide targets, one has to consider the availability of the material and the tremendous increase of safety problems in the handling of targets from U to Cf, Es or Fm.

Finally, we have to notice that SHEs at $Z = 114$ and $N = 184$ cannot be reached even in reactions with radioactive actinide targets and radioactive beams. Concerning the CN on the right from $N = 184$ one has to keep in mind that the fission barrier vanishes rapidly with the neutron number, and therefore these nuclei cannot be synthesized.

Prompted by the recent experiments, a number of theoretical studies are aimed at reproducing the known cross-section data and further extrapolating the calculations into the region of spherical superheavy nuclei [29,33–40]. The measured cross-sections for the formation of ^{257}Rf up to $^{277}112$ are reproduced almost within about a factor of 2 by the various models. However, there are significant differences in the cross-section values for the synthesis of spherical SHEs beyond $Z = 114$.

5 Summary and outlook

The experimental work of the last two decades has shown that the cross-sections for the synthesis of the heaviest elements decrease almost continuously. However, the recent data on the synthesis of elements 114 and 116 in Dubna using hot fusion and 118 in Berkeley using cold fusion seem to break this trend when the region of spherical superheavy elements is reached. Therefore, a confirmation is urgently needed that the region of spherical SHEs has finally been reached and that the exploration of the “island” has started and can be performed even on a relatively high cross-section level.

The progress towards the exploration of the island of spherical SHEs is difficult to predict. However, despite the

exciting new results, many questions of more general character are still awaiting an answer. New developments will not only make it possible to perform experiments aimed at synthesizing new elements in reasonable measuring times, but will also allow for a number of various other investigations covering reaction physics at the limits and spectroscopy of exotic nuclei.

One can hope that, during the coming years, more data will be available in order to promote a better understanding of the stability of the heaviest elements and the processes that lead to fusion. The microscopic description of the fusion process will be needed for an effective explanation of the measured phenomena in the case of low dissipative energies. Then, the relationships between fusion probability and stability of the fusion products may also become apparent.

An opportunity for the continuation of experiments in the region of SHEs at decreasing cross-sections will be afforded by further accelerator developments. High-current beams and radioactive beams are the options for the future. At increased beam currents, values of tens of particle μA 's may become possible, the cross-section level for the performance of experiments can be shifted down into the region of tens of femtobarns, and excitation functions can be measured on the level of tenths of picobarns. The high currents, in turn, require the development of a new target and improvement of the separator. The radioactive beams, not as intense as the stable beams, will allow for approaching the closed neutron shell $N = 184$ already at lighter elements. Interesting will be the study of the fusion process using unstable neutron-rich beams.

The half-lives of SHEs are expected to be relatively long. Based on nuclear models, which are effective predictors of half-lives in the region of the heaviest elements, values from microseconds to years have been calculated for various isotopes. This wide range of half-lives encourages the application of a wide variety of experimental methods in the investigation of SHEs, from the safe identification of short-lived isotopes by recoil-separation techniques to exact mass measurements and atomic physics experiments on trapped ions, and to the investigation of the chemical properties of SHEs using long-lived isotopes.

The recent experiments at SHIP were performed in collaboration with D. Ackermann, F.P. Heßberger, B. Kindler, J. Kojouharova, B. Lommel, R. Mann, G. Münzenberg, S. Reshitko, H.J. Schött (GSI Darmstadt); A. Popeko, A. Yerein (JINR Dubna); S. Antalic, P. Cagarda, S. Šáro (University Bratislava); M. Leino, J. Uusitalo (University Jyväskylä).

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