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PROBING SHN STRUCTURE AND STABILITY — SYNTHESIS AND DECAY STUDIES AT S^3*

D. Ackermann [a], F. Agert [a,b,†], A. Bahini [a]
D. Boilley [a,b], H. Donglo [a,b], S. Kumar [a,b], J. Piot [a]
H. Savajols [a], Ch. Stodel [a]

and the S^3 Collaboration

[a]Grand Accélérateur National d'Ions Lourds (GANIL)
CEA/DRF-CNRS/IN2P3, Caen, France
[b]Université de Caen Normandie, Caen, France

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The superheavy nuclei (SHN), residing at the edge of nuclear stability at the highest atomic charge Z and mass A, are in the focus of scientific efforts worldwide due to the high discovery potential offered by their investigation. In this paper, we report on some of the major challenges and how we intend to approach them at GANIL, with advanced instrumentation and state-of-the-art experimental methods, as well as reaction theory approaches to the SHN production mechanism. Decay Spectroscopy After Separation (DSAS) is an efficient tool to study nuclear structure features of the heaviest nuclear species such as single particle trends towards the predicted next spherical shell closures beyond ^{208}Pb, and deformation and exotic shapes, leading also to the formation of meta-stable states, like, *e.g.*, K-isomers. The understanding of the reaction mechanism governing heavy collisions employed for the synthesis of the heaviest nuclei, despite decades of experimental and theoretical efforts, is still a challenging task. Being a fundamental topic by itself, mastering reaction theory and producing reliable cross-section predictions are essential for a successful experimental program. Detailed nuclear structure studies of the heaviest nuclei, as well as the synthesis of superheavy elements (SHE) are presently still hampered by the limited efficiencies of the existing experimental facilities. To overcome this restriction, substantial efforts are being made to upgrade and develop existing and new facilities worldwide, online since recently or planned for the future, including the linear accelerator facility SPIRAL2 at GANIL, equipped with the versatile separator–spectrometer set-up S^3.

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† Present address: CEA, DES, IRESNE, DER, SPRC, LEPh, 13108 Saint-Paul-lez-Durance, France.

1. Introduction

The history of the investigation of the heaviest nuclear species since the middle of the last century has led to a substantial body of data for the hitherto known 206 isotopes from einsteinium to the heaviest element for which observation has been reported, oganesson, as shown in Fig. 1 where the decay modes are shown and Fig. 2 which illustrates the nuclides' half-lives (upper panel) and year of discovery (lower panel).

In this paper, we discuss the approach to the study of SHN at GANIL, illustrating the envisaged methods of nuclear structure studies, based on state-of-the-art results, as well as the development of reaction theory and its tools needed for efficient experiment planning at the future facility. We also present the investigation of single-particle-level trends, K isomerism, and decay-mode competition with an emphasis on β decay and its non-observation for isotopes with $Z > 105$.

The major technique providing the bulk of information is Decay Spectroscopy After Separation (DSAS), an efficient tool for studying nuclear structure features of superheavy nuclei (SHN), discussed in detail in a recent review [1]. The unpaired nucleon in odd–even and even–odd nuclei can serve as an efficient probe to study these metastable states and other nuclear structure effects. High K values are often generated by the coupling of higher-order quasi-particle excitations of high-J orbitals in the vicinity of the deformed shell gaps for neutrons and protons at $N = 152$ and $Z = 100$, making K-isomers a systematic feature in that region of the Segrè chart, and a sensitive probe for deformation developing towards the spherical SHN. Complex configurations of low-lying states in heavy nuclei can also have consequences for decay mode competition and nuclear stability, providing indications that might also be relevant for the location of the long-searched-for *island of SHN with enhanced stability*.

The non-observation of β-decay beyond dubnium ($Z = 105$) is evident in Fig. 1, which is at least partly due to the insensitivity of the methods, hitherto applied for their investigation. Recent theoretical work by Ravlič and Nazarewicz [2] predicts competitive β-decay probabilities for some of those very heavy species up to flerovium ($Z = 114$). Examples of nuclides for which β decay (EC) is more likely than the assumed decay by spontaneous fission (SF) are the dubnium isotopes ^{263,266,268}Db, which are marked by pink triangles in Fig. 1 in the online version of this paper or Fig. 2.1 in Ref. [1].

To produce those heavy nuclei, a detailed understanding of the reaction mechanism needed for their synthesis is essential. The theoretical treatment of the 3-step process of the fusion–evaporation reaction leading to the synthesis of heavy nuclei depends critically on ingredients like the model-

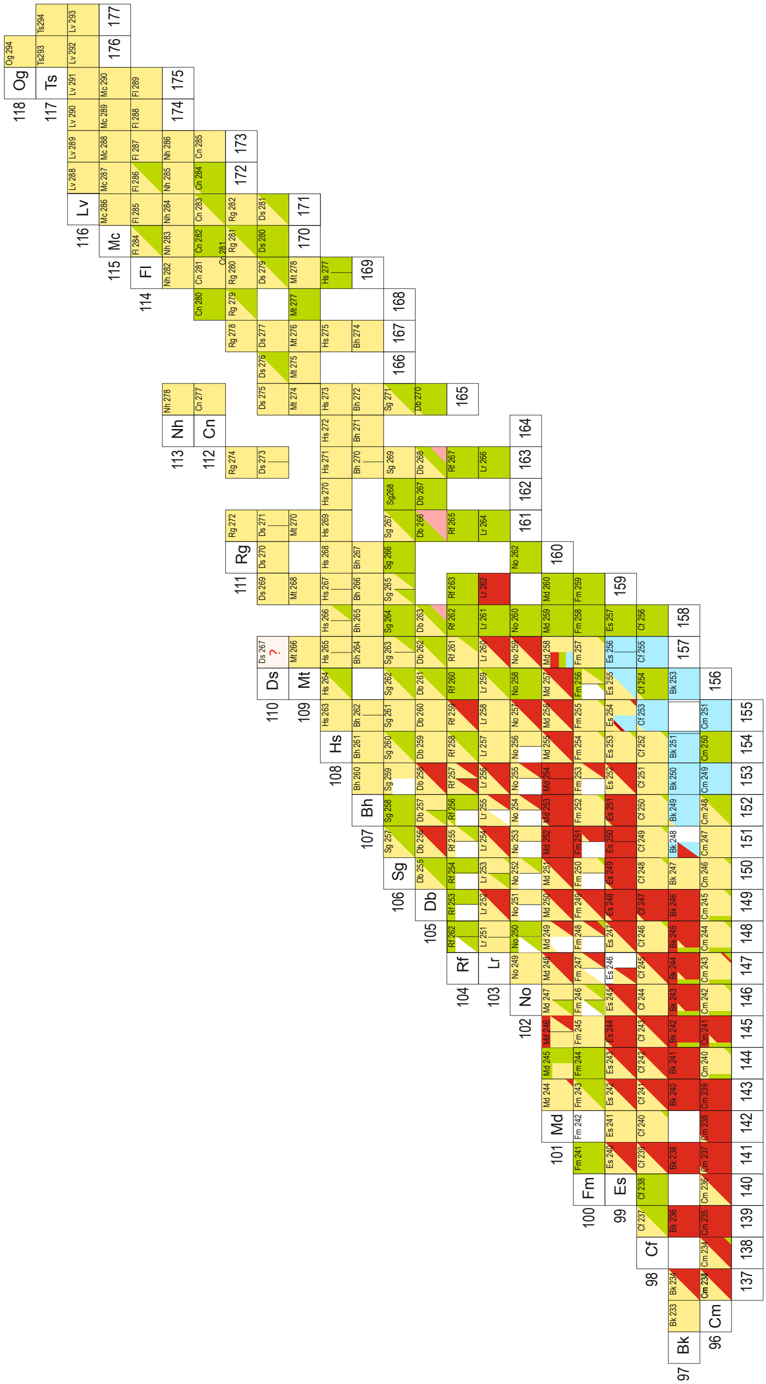

Fig. 1. (Colour on-line) Excerpt of the chart of nuclides showing the heaviest nuclei observed ($Z = 96$–118 and $N = 137$–177), updated from Ref. [1].

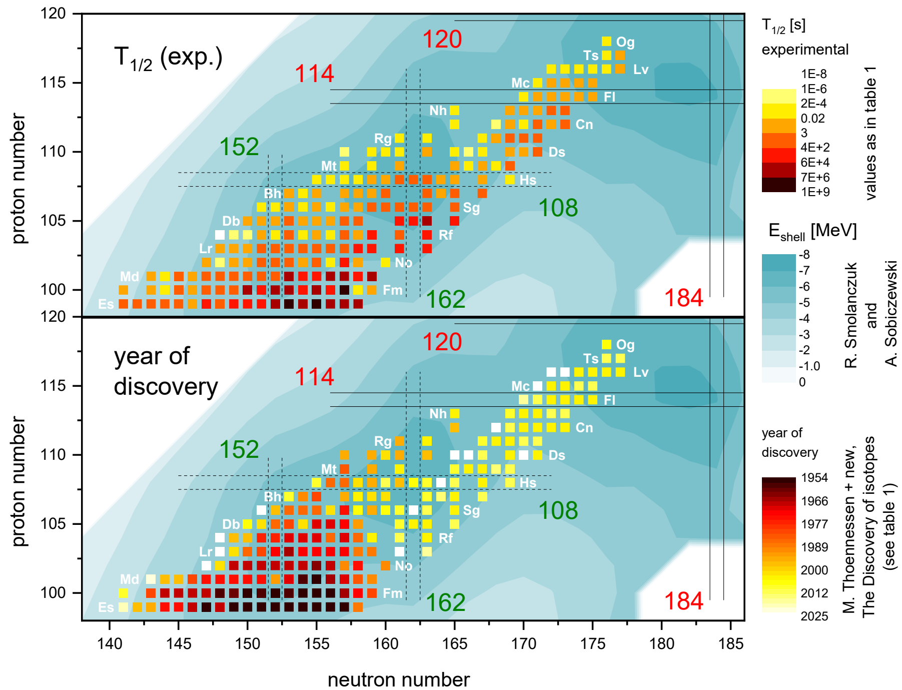

Fig. 2. (Colour on-line) Shell correction energies from Refs. [6, 7] as a function of proton and neutron numbers with (upper panel) experimental half-lives $T_{1/2}$ as listed in Ref. [1], and (lower panel) years of discovery from Ref. [8], including updates from the «Discovery of Nuclides Project» at `https://people.frib.msu.edu/~thoennes/isotopes/`, for the heaviest nuclei from einsteinium to oganesson. The predicted closed shells on the *island of stability* for spherical superheavy nuclei (SHN) for protons at $Z = 114$ and 120, and neutrons at $N = 184$ are indicated by solid black lines. The subshell closures or shell gaps for deformed shell-stabilized nuclei for protons at Z=108, and for neutrons at $N = 152$ and 162, are indicated by dashed black lines. (Reproduced from Ref. [1].)

dependent fission barriers, which lead to inconsistencies in the complementary description of compound-nucleus formation as pointed out, *e.g.*, by Naik *et al.* in 2007 [3], and further elaborated by Lü and Boilley in 2016 [4]. Improving the description of the process leading to the final product of the fusion–evaporation process, the evaporation residue (ER), is an ongoing effort that is pursued at GANIL continuously, *e.g.*, by improving the statistical model code KEWPIE2 [5].

After this introduction, the specific application of DSAS for K-isomers and decay mode competition, and the role of weak decays in the region of SHN are discussed in Section 2. The theoretical efforts pursued at GANIL to improve the description of the reaction mechanisms involved in the synthesis of the SHN are presented in Section 3. In Section 4, the future perspectives of the field in view of the advent of the separator–spectrometer installation S^3 at the linac facility SPIRAL2 of GANIL are outlined.

2. DSAS for SHN — some examples

While the scope of the search for new superheavy elements (SHE) is very prestigious, but limited to the synthesis of new elements and isotopes, revealing only basic decay properties of the heaviest species, DSAS aims at and has the potential to study detailed nuclear structure properties of SHN. From the understanding of single-particle levels and their isotopic/isotonic trends, conclusions can be drawn on the location of the next spherical shell closures at the *island of stability*. The investigation of isomeric states, in particular K-isomers of deformed nuclei and their development towards the highest Z and N, as well as decay mode competition (SF, α- and β-decay), yields valuable information on nuclear stability and the onset of sphericity.

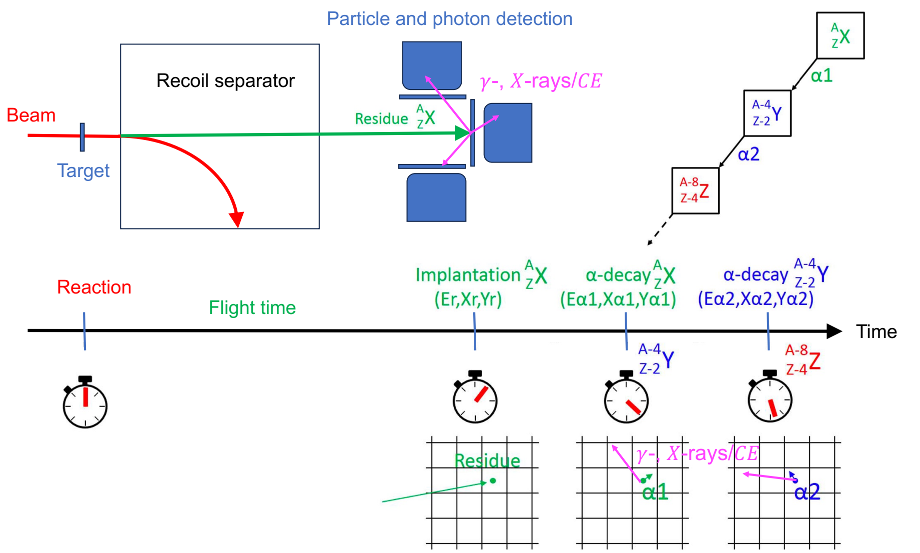

Fig. 3. (Colour on-line) DSAS and genetic correlations: the recoiling nucleus ^A_ZX is implanted in a position-sensitive detector at position (X_r, Y_r). It subsequently decays via α emission into its neighborhood at position ($X\alpha1$, $Y\alpha1$) to the daughter nucleus ($^{A-4}_{Z-2}Y$) which itself decays to the granddaughter ($^{A-8}_{Z-4}Y$) at position ($X\alpha2$, $Y\alpha2$). In addition to the recoils and α particles, γ-, x-rays, and CEs are detected in coincidence. The technique allows for correlations in position and/or time of the recoil implantation, and its subsequent decay due to the inclusive detection of the particles and photons involved. (Figure and caption are taken from Ref. [9].)

To study SHN by investigating their various decay modes, after they were produced as ER, with partly extremely low cross sections, highly efficient separation schemes are needed to suppress the primary beam and products from competing reaction channels. The various separation schemes have been discussed in Ref. [9], listing the major existing separator facilities. The separated heavy and superheavy species are studied, employing comprehensive particle and photon detector systems that provide efficient and highly resolving detection of α particles, electrons, fission fragments, γ- and x-rays. Particle and photon correlations, employing energy, time and position information, allow for the construction of so-called genetic decay chains, consisting of the various decay generations, as shown in Fig. 3 from Ref. [10]. More details on this powerful and well-established tool to investigate rare activities, including single-event detection, are given in Ref. [11].

The advanced DSAS technologies, including fast-timing sensitivity and low-energy threshold conversion electron (CE) detection as described, *e.g.*, in Ref. [12], are essential to investigate rare and short-lived states, and have the potential to reveal the hitherto almost completely missing information on β-decay properties of nuclides for all elements beyond dubnium (see Fig. 1).

2.1. K-isomers

In the region of deformed heavy nuclei around and beyond the shell gaps at $Z = 100$ and $N = 152$, K-isomers play a major role, as illustrated in Fig. 4, where the known cases in isotopes from curium to darmstadtium are shown. For a more comprehensive discussion of metastable states in SHN, see our recent review [10]. The presence of high-J orbitals in that region of the Segrè chart favours the generation of high values of K. In particular, the unpaired nucleon in odd–even and even–odd nuclei, such as ^{249,251}Md [13] or ^{255}No [14, 15], can serve as an efficient probe to study these metastable states and other nuclear structure effects. While for even–even isotopes often 2-quasi-particle excitations across a shell gap lead to high-K numbers, the meta-stable states in odd-mass nuclei are formed as 3-quasi-particle or even 5-quasi-particle states [15], where high-K values are produced by 2- and 4-quasi-particle excitations coupled to the odd unpaired nucleon.

^{270}Ds and its α-decay daughter ^{266}Hs are the heaviest nuclei for which a K-isomer has been observed, with the specific features that the delay due to isomerism is observed in α-decay times and that the isomeric decay proceeds slower than the decay of the ground state [16, 17]. While for even–even nuclei, due to their distinct level structure, the α-decay spectrum exhibits a well-defined and separated decay-energy peak structure, dominated by the transition of the ground state of the mother to the ground state in the daughter nucleus, the 31 known α-decay energies of ^{270}Ds are rather fragmented. Two major structures are observed around 11 MeV and 12 MeV, each spread over several hundred keV, a behaviour that is observed rather for odd-A and odd-Z nuclei, due to the higher level densities at low excitation energy E^* caused by the unpaired nucleon(s). This feature is presently being analysed, employing advanced nuclear structure theory.

In Fig. 4, the six even–odd nobelium and rutherfordium isotopes, and the three mendelevium and lawrencium isotopes, for which high-K isomers have been reported, are presented with their major properties. Up to date, no high-K isomer has been reported for an odd–odd nucleus with $Z \geq 96$.

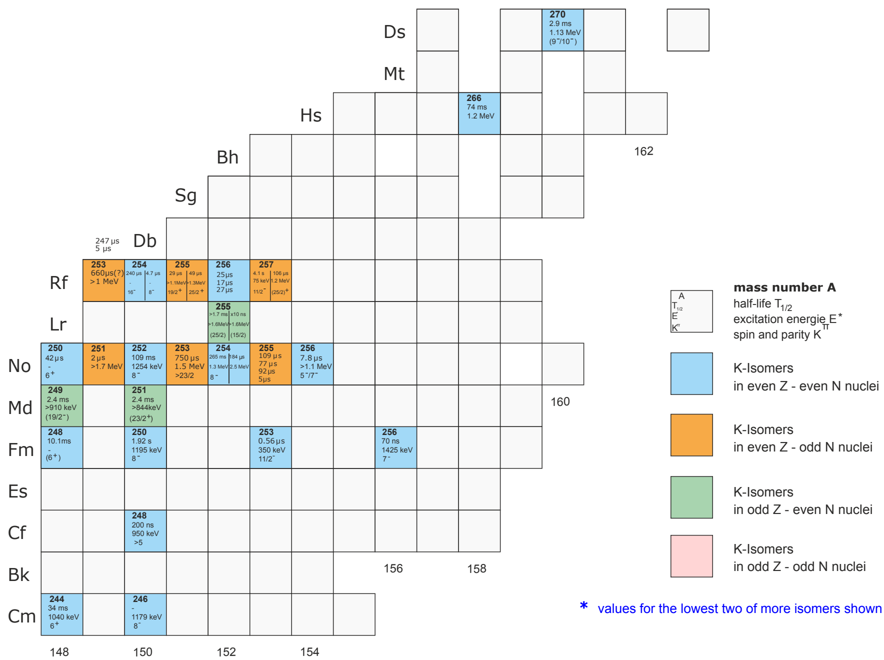

Fig. 4. (Colour on-line) Summary of K-isomers for the heaviest nuclei at and above $Z = 96$. (Taken from Ref. [1].)

The high intensities provided by the new LINAC of GANIL/SPIRAL2, together with the high transmission of S^3 and its comprehensive focal plane detection array SIRIUS (see Section 4 for references) will allow the extension of these studies towards the next higher neutron shell gap $N = 162$ and towards the onset of the deformation development towards sphericity, generally expected for the next proton and neutron shell closures beyond ^{208}Pb. One of the first tasks in this context will be to clarify not-yet-understood features observed for the α-decaying K-isomers ^{266}Hs and ^{270}Ds.

2.2. Decay mode competition

Complex configurations of low-lying states in heavy nuclei can also have consequences for decay mode competition and nuclear stability, providing indications that might also be relevant for the location of the long-searched-for *island of SHN with enhanced stability*. One example is the observation of low excitation energies for single-particle states originating from orbitals that are supposed to define the shell gaps for spherical SHN, like in the case of the ^{247}Md $\rightarrow$ ^{243}Es decay [18]. Here the decay mode competition between *SF* and α decay resulted in largely different SF–α branching ratios with $b_{\mathrm{SF}} = 8.6 \times 10^{-3}(10)$ for the $7/2^{-}[524]$ ^{247gs}Md and $b_{\mathrm{SF}} = 0.20(2)$ for the $1/2^{-}[521]$ ^{247m}Md. The higher spin value of the ground state leads apparently to a substantial fission hindrance, which was also predicted by theory, *e.g.*, in Ref. [19] for the superheavy isotope ^{271}Rg. For this nucleus, a fission barrier 2.5 MeV higher was predicted for the $11/2^{+}$ excited state with respect to the $1/2^{-}$ ground state.

In the same α-decay study, the two-proton single-particle states $1/2^{-}[521]$ and $3/2^{-}[521]$, originating from the orbitals $f_{5/2}$ and $f_{7/2}$, which are predicted to define the $Z = 114$ proton shell closure, could be populated and identified. While theory predictions, *e.g.*, in Ref. [20], calculate an energy gap of ≈ 1 MeV, the ^{243}Es level scheme places the $3/2^{-}[521]$ as the ground state and the $1/2^{-}[521]$ at ≈ 70 keV above [18, 21]. That contradicts the prediction of $Z = 114$ as the next proton shell closure beyond ^{208}Pb.

A similar situation is observed for ^{259}Sg and its α decay to ^{255}Rf, for which fission from the $11/2^{-}$ ground state is hindered with respect to $1/2^{+}$ first excited state.

As far as the competition of SF with β decay is concerned, we note that for nuclides beyond dubnium with $Z > 105$, no β decay has been observed, which is partly due to the insensitivity of this decay mode to the hitherto performed experiments. A possible solution for this problem is provided by the detection of CEs, employing fast digital electronics as described, *e.g.*, in Ref. [12]. In fact, the endpoints of decay chains starting from moscovium and tennessine isotopes could be expected to decay by EC, which is indicated in Fig. 1 in the online version of this paper or Fig. 2.1 in Ref. [1] by pink triangles for ^{263,266,268}Db. A recent theory study supporting this assumption will be discussed in Section 2.3.

2.3. Weak decay of SHN

As mentioned above (Section 2.2), no experimental evidence has been registered on weak decays for nuclei beyond dubnium ($Z = 105$). However, in a recent approach from theory based on relativistic nuclear density functional theory and the quasiparticle random-phase approximation, Ravlič and Nazarewicz [2] report on a novel approach to understand the weak decay of SHN.

The authors underline the importance of first forbidden (FF) transitions, in particular, 1^- transitions which were not considered in earlier approaches to the problem, and they find competitive transition probabilities for β decay in comparison to fission. They investigated isotopes of elements from mendelevium to oganesson with their calculated β^+/EC decay rates comparing to the known half-lives shown in Fig. 4 of Ref. [2].

The EDF calculations, using DD-PC1 and DD-PCX functionals, yield 45 nuclides with an EC decay branching $> 5\%$. Only six of these are even–even isotopes. The majority of these isotopes are found in the lighter elements, but they also extend beyond dubnium up to one case in flerovium (^{290}Fl). For the dubnium isotopes mentioned above, the calculated values show that EC is competitive for ^{263}Db and even dominating (shorter EC half-life) for ^{266}Db, supporting the suggestions in Fig. 1 in the online version of this paper or Fig. 2.1 in Ref. [1], where possible EC decay is marked by pink triangles.

These predictions strongly support experimental verification and the need to search for weak decay modes in SHN. A possible experimental approach, employing CE and x-rays spectroscopy, has been demonstrated for the case of the ^{258}Db EC-decay into the ground and an excited state of ^{258}Rf [22]. At GANIL S^3 with its DSAS detector array SIRIUS (for references, see Section 4) will be ideal for this type of investigation.

3. Reaction mechanism studies and cross-section predictions

To successfully plan and perform experiments to study SHN, a reliable prediction of their production cross section is essential. To provide a software tool for this task at GANIL, the code KEWPIE and its more advanced version KEWPIE2 [5] have been introduced, being under continuous development.

The fusion–evaporation process to synthesize heavy and superheavy nuclei is often described as a 3-step process consisting of the capture, the compound nucleus (CN) formation, and the survival against fission

$$\sigma_{\rm ER} = \frac{\pi\hbar^2}{2\mu E_{\rm cm}} \sum_{\ell} (2\ell+1)\, T(\ell) \times P_{\rm form}(\ell) \times P^{xn}_{\rm survival}(\ell)\,, \tag{1}$$

with $T(\ell)$, $P_{\rm form}(\ell)$, and $P^{xn}_{\rm survival}(\ell)$ being, respectively, the Coulomb barrier transmission, CN-formation, and survival probabilities as functions of the angular momentum ℓ. Here, μ is the reduced mass of projectile and target nuclei, $E_{\rm cm}$ kinetic energy in the centre-of-mass frame, see, *e.g.*, Ref. [33].

Apart from the uncertainties in the modelling of the capture, which can be directly compared to experimental data, major problems are caused by the difficulties in predicting the formation probabilities, which are impossible to measure, as well as the uncertainties in the predicted fission barriers, leading to large uncertainties in the survival probabilities. This has been illustrated by Lü *et al.* [4] by systematically comparing CN-formation probabilities ($P_{\rm form}$) for different fission barrier calculations constrained by the reproduction of experimental cross sections for superheavy ERs. In Fig. 5, taken from Ref. [4] as one example, mean CN formation probability values $\bar{P}_{\rm form}$ for two sets of theoretical fission barriers are compared to various model predictions, discussed earlier by Naik *et al.* [3]. Due to the compensation of the higher fission barriers resulting from the macroscopic–microscopic finite-range liquid-drop model (FRLDM) by Möller *et al.* [25] (red circles in Fig. 5) as compared to the Lublin–Strasbourg drop (LSD) model [23] (red squares in Fig. 5), the resulting CN-formation probabilities differ by more than an order of magnitude.

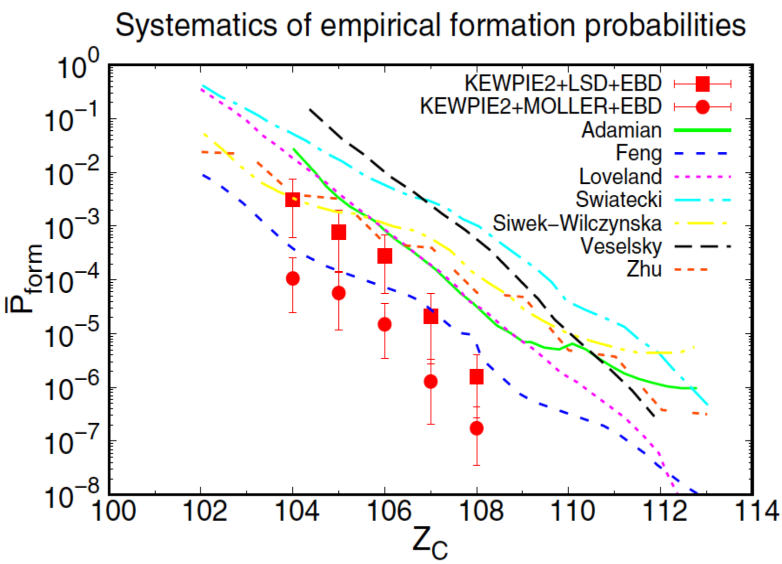

Fig. 5. (Colour on-line) Systematic comparison of the calculated mean values of the empirical formation probability for the cold-fusion reactions leading to the synthesis of the elements from $Z = 104$ to $Z = 108$ (*cf.* Table I in Ref. [4]). The deduced formation probabilities are based upon two different fission-barrier models, Lublin–Strasbourg drop (LSD) [23] with shell corrections from Ref. [24], and the macroscopic–microscopic finite-range liquid-drop model from Möller *et al.* [25]. Lines correspond to theoretical predictions from Refs. [26–32]. (Figure and caption taken from Ref. [4].)

By comparing, in addition, the empirical barrier-distribution (EBD) method [26, 34] as in Fig. 5, to the Wentzel–Kramers–Brillouin (WKB) approximation with a proximity potential [35, 36], Lü *et al.* point out how critical model assumptions are for the prediction of absolute SHN synthesis cross sections, clearly demanding substantial further theoretical efforts to enable reliable model predictions. In a recent and continuing effort, Donglo and co-workers revisited the fusion-by-diffusion approach [33] to get a new version of the KEWPIE2 code [37].

4. Opportunities at SPIRAL2/S^3 — an outlook

The understanding of the reaction mechanism governing heavy collisions employed for the synthesis of the heaviest nuclei, despite decades of experimental and theoretical efforts, is still a challenging task [4]. Being a fundamental topic by itself, mastering reaction theory and producing reliable cross-section predictions are essential for a successful experimental program.

Detailed nuclear structure studies of the heaviest nuclei as well as the synthesis of SHE are presently still hampered by the limited efficiencies of the existing experimental facilities. To overcome this restriction, among other facilities online since recently or planned for the future worldwide, the linear accelerator facility SPIRAL2 at GANIL in Caen, France [38], equipped with the versatile separator spectroscopy set-up S^3 [39], will be operational shortly. The SPIRAL2 LINAC [38] has started operation of its first phase, being limited to the highest intensities to projectile masses $A \leq 40$. The hitherto performed experiments confirm that the design goals resulting in intensities of, *e.g.*, at least 10 particle μA for ^{48}Ca, will be reached, which is about one order of magnitude more than provided by the previous generation accelerator facilities, comparable to the performance of the SHE factory of the Flerov Laboratory for Nuclear Reaction FLRN/JINR in Dubna [39]. The complete capabilities of this installation will be available with the second phase of the project, the new injector NEWGAIN [40]. It will provide the highest intensities for all ions, and together with the separator–spectrometer set-up S^3 [41] will be one of the world's most competitive facilities for the investigation of SHN.

With its separation and detection capabilities S^3 [42], equipped with the detection array for Spectroscopy and Identification of Rare Isotopes Using S^3 (SIRIUS) [43] as well as the S^3 Low Energy Branch (S^3 LEB) [44], has the potential to advance SHE/SHN research to the next level. The latter will be offering tools for laser spectroscopy, mass measurement, and the setup for laser/trap-assisted DSAS SEASON. With SIRIUS detailed DSAS studies as described in Section 2, will be possible for isotopes at and beyond the next proton and neutron deformed shell gaps at $Z = 108$ and $N = 162$, respectively, with promising implications for the search for the next spherical shell closures beyond ^{208}Pb.

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