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How accurately can we predict synthesis cross-sections of superheavy elements?

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Synthesis of superheavy elements beyond oganesson is facing new challenges as new target-projectile combinations are necessary. Guidance from models is thus expected for future experiments. However, hindered fusion models are not well established and predictions in the fission barriers span few MeVs. Consequently, predictions are not reliable. Strategies to constrain both fusion hindrance and fission barriers are necessary to improve the predictive power of the models. But, there is no hope to get an accuracy better than one order of magnitude in fusion-evaporation reactions leading to superheavy elements synthesis.

Keywords: superheavy elements; nuclear reactions; uncertainty analysis.

I. INTRODUCTION

After the recent successes which lead to fill-up the last line 3 of Mendeleev's periodic table [1], the synthesis of superheavy 4 elements is facing new challenges. The heaviest synthetic el-5 ements have all been created in collisions of two heavy nu-6 clei. For a historical review, see Ref. [2]. However, one has 7 to find new target-projectile combinations to extend further 8 the periodic table. For cold fusion reactions, the expected 9 cross-sections are too low with present facilities to expect to 10 synthesise a new element in a reasonable timeframe. And, for 11 hot fusion reactions, there is no available target in sufficient 12 quantity anymore to be associated with the ⁴⁸Ca beam. Heav-13 ier projectiles must be used. Thus, one has to find new opti-14 mum target-projectile combinations. Guidance from models 15 is expected to optimise future experiments, and various pre-16 dictions have been continuously published in the scientific lit-17 erature. Accurate predictions are necessary as a small change 18 in the cross-section could mean months of beam time for an 19 experiment.

However, a direct comparison shows that predictions dis-21 agree with each other [3, 4] even if the models can reproduce 22 existing data. One of the reasons is the so-called fusion hin-23 drance, i.e. the strong reduction in the fusion cross-section with respect to what is calculated by a simple extrapolation 25 of fusion models with light nuclei. Its origin is well under-26 stood and it is now widely acknowledged that the dynamical 27 trajectory for the fusing system must pass over a conditional saddle point in a multidimensional space in order to form a compound nucleus, in contrast to light systems for which the conditional saddle point lies outside the point of hard contact in heavy-ion reactions. Dissipation also plays a crucial role 32 to understand the fusion hindrance. But, there is no consen-33 sus on the dynamical models, leading to large discrepancies in predictions.

The total fusion-evaporation cross-section is a combination 36 of three steps described by three different models, namely the

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37 capture cross-section that brings the two nuclei in contact, 38 the formation probability to reach the compound shape and 39 the survival probability accounting for neutron evaporation 40 from this excited compound nucleus which competes with the 41 predominant fission decay mode:

$$\sigma_{ER} = \sigma_{cap} \times P_{CN} \times P_{sur}. \tag{1}$$

43 Capture and survival phases can be described by extrapolated 44 models used for the fusion of light nuclei. They are supposed 45 to be the best known parts of the reaction. Thus, the physics 46 used to estimate σ_{cap} and P_{sur} is well established; however, 47 parameters entering the models are not well constrained lead-48 ing to uncertainties in the predictions.

The formation probability that is very specific to heavy 50 ions collisions, is responsible for the hindrance phenomenon. 51 There is no consensus on the dynamical model nor on the pa-52 rameters. The physics used to describe the formation phase 53 faces several open questions, and, as pointed out in Refs. $_{54}$ [3, 4], P_{CN} calculated by various models spans two or three 55 orders of magnitude. See Fig. 1. Thus, there is no hope to 56 produce reliable predictions without assessing the formation

References [3, 4] also show that once multiplied by σ_{cap} 59 and P_{sur} , all models converge to experimental data. This $_{\rm 60}\,$ means that the uncertainties in σ_{cap} and P_{sur} are large enough to compensate the discrepancies of the various formation models and can be adjusted to get experimental data [5, 6].

Thus, to improve the predictive power of the models, we 64 also have to find ways to reduce uncertainties in σ_{cap} and 65 P_{sur} . Capture cross-section can be directly measured and the 66 models are well constrained. Discrepancies between two reasonable models are lower than an order of magnitude [5]. Regarding the survival probability, the decay of the compound nucleus formed in the collision is dominated by fission. This means that a small change in the fission width will not affect much the fission probability but will induce great variations of the survival probability. Therefore, it is not a surprise that 73 the uncertainty analysis performed in Refs. [5, 6] has led to 74 pin down the fission width as a key parameter that needs to 75 be assessed in order to improve the predictive power of the 76 models.

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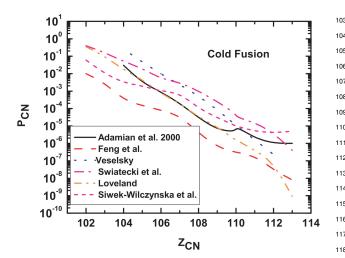


Fig. 1. Formation probability for cold fusion reactions as a function 120 of the charge of the compound nucleus calculated by various models. Figure reproduced from Ref. [3]. See references therein for the models.

₇₈ the survival probability and propose strategies to constrain the ₁₂₅ Drop-Model and ΔE_{shell} is the shell correction energy at the formation probability that accounts for the fusion hindrance.

UNCERTAINTIES IN THE SURVIVAL PROBABILITY

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Various predictions

Compound nuclei formed during the collision decay through neutron evaporation or fission. Thus, the 1n survival 84 probability is nothing else than a branching ratio

$$P_{1n} = \frac{\Gamma_n}{\Gamma_n + \Gamma_f},\tag{2}$$

where Γ_n and Γ_f stand for the neutron evaporation width and the fission width respectively. The latter dominates. They can be calculated by a standard statistical model; see e.g. Ref. [7]. 89 Uncertainty in the survival probability is easily deduced from 90 the uncertainties in the widths, using the usual propagation 141 duce uncertainties in the calculated physical quantities. There 91 formula:

$$\left[\frac{u(P_{1n})}{P_{1n}}\right]^2 = \left[\frac{\Gamma_f}{\Gamma_n + \Gamma_f}\right]^2 \left(\left[\frac{u(\Gamma_n)}{\Gamma_n}\right]^2 + \left[\frac{u(\Gamma_f)}{\Gamma_f}\right]^2\right).$$

93 Here, we have assumed that Γ_n and Γ_f are independent from 147 each other. When the ratio Γ_f/Γ_n is large, the relative uncerfission width will affect much the survival probability.

100 sion width contributes equally. However, Γ_n mainly depends 154 hundreds of keV. on the neutron separation energy, whereas the fission width 155 mainly depends on the fission barrier, the damping energy of 156 measurement of the fission barrier is not an easy task. The

103 the shell correction energy and the friction coefficient when taken into account. None of these parameters can be directly 105 measured and are not well determined, inducing large uncertainties in the fission width and then, survival probability. As shown in Refs. [5, 6], the fission barrier is the dominating quantity among these three parameters.

Fission barriers can be calculated by various microscopic or macroscopic-microscopic models, while the other two parameters are generally adjusted to reproduce experimental data. Comparison between different calculations [8, 9] has shown that predictions in fission barriers, which is the most sensitive parameter, span few MeV. Consequently, calculated survival probabilities can differ by several orders of magni-

Even if one restricts the comparison to fission barriers that share the same shell correction energy, the predicted values span 1.5 MeV and can induce two orders of magnitude differences in the fusion-evaporation cross-sections [5, 6]. In one case, the fission barriers were estimated by the old method,

$$B_f = B_{LDM} - \Delta E_{shell},\tag{4}$$

In this article, we shall focus on the uncertainty analysis of 124 where B_{LDM} is the fission barrier calculated with a Liquid-126 ground state. The other case corresponds to fission barriers calculated directly by the same macroscopic-microscopic

> Although the survival probability is the best understood 130 part of the reaction leading to the synthesis of superheavy el-131 ements, the ambiguities in the fission barrier lead to discrep-132 ancies that span several orders of magnitude. This is more than for the formation probability that is supposed to be less understood.

Thus, reliable predictions require accurate fission barrier predictions. How well can we predict such a sensitive parameter?

B. Best estimate

Assuming that the model predicting the fission barrier is 140 correct, there are still ambiguities in its parameters that in-142 are few uncertainty analyses of the theoretical evaluation of the fission barrier. In Ref. [10], a very simple micro-macro model was used and leads to an uncertainty in the fission barrier of about 0.5 MeV. Another estimate based on a Bayesian analysis of a DFT model leads to a similar value for the uncertainty in the fission barrier of ²⁴⁰Pu [11].

All these estimates rely on a fitting procedure on nuclear tainty in the survival probability reaches its maximum value. 149 masses and give an uncertainty proportional to the RMS of This is very intuitive: as the fission decay mode dominates, 150 the fit [12]. Whatever the model, this RMS has remained over evaporation events are very rare. Thus, a small change in the 151 few hundreds of keV this last decade. There is little hope to 152 get an order of magnitude better. Thus, the uncertainty in Uncertainty in the neutron evaporation width and in the fis- 153 the fission barrier for a given model will also remain at few

Constraint from experiments is possible although direct

157 heaviest nucleus which fission barrier has ever been measured 209 the cross-section due to the fusion hindrance to the synthesis 158 is ²⁵⁴No using the gamma multiplicities. Uncertainty at spin 210 of superheavy elements. For a review, see Ref. [22]. 159 0 is estimated to be about 0.9 MeV [13–15]. This value is 211 160 extrapolated from measurements at higher spins. The fission 212 the fusion hindrance [23], associated fluctuations were not inbarrier can also be extracted by inverting a statistical decay 213 cluded so far. The Langevin equation that has been used ex-162 model, namely the Kewpie2 code [7]. The result is model 214 tensively to study the fluctuation-dissipation dynamics [24] in another model. In particular, the extracted fission barrier 216 conditional saddle that lies between the contact and the comdepends on the value of the other parameters such as the fric- 217 pound configurations [25–29]. This leads to more realistic tion coefficient. An analysis based on Bayesian inference was 218 dependence of the fusion cross section as a function of energy performed to deduce the fission barrier from invented exper- 219 and a better estimate of the extremely low cross-sections. imental data [16]. The uncertainty in the barrier depends on 220 see Ref. [16] for details. 171

Consequently, it seems that there is an incompressible 224 have large uncertainties. The friction coefficient is not better 227 ons [31–33]. Thus, it is contradictory with the models men-176 known than 20 years ago. The damping energy of the shell 228 tioned above based on the evolution of collective degrees of correction energy has not been carefully investigated since 229 freedom. the Ignatyuk's prescription [17] in 1975. 178

estimate the cross-section of rare events, it is hardly conceivfor hot fusion reactions as we need fission barriers of several isotopes.

NECESSITY TO CONSTRAIN THE FUSION HINDRANCE

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For the less known part of the reaction, i.e. the formation 243 188 189 step responsible for the hindrance of the fusion, there is no 244 power of the models describing the whole reaction leading to 190 consensus on the model.

Common understanding of the phenomenon

Understanding and modelling fusion hindrance is a long 193 standing problem. Back in the 1960s, the first predictions 248 of fusion-evaporation cross-sections were far too optimistic 249 to assess the formation mechanism because of the survival [18]. Nix and Sierk showed for symmetric reactions that the 250 probability that is also not well constrained as explained in dynamical trajectory for the fusing system must pass over a 251 the previous section. To get rid of the decay step, one should conditional saddle point in a multidimensional space in or- 252 focus the step before in the reaction, i.e, the fusion crossder to form a compound nucleus, in contrast to light systems 253 section. 199 for which the conditional saddle point lies outside the point 254 of hard contact in heavy-ion reactions [19]. Later, Świątecki 255 quasifission process. It consists in reseparation into two frag-[20, 21] first introduced a dynamical model that broke with 256 ments without reaching the compound state. On an experi-202 the idea that the capture process can be understood in terms 257 mental point of view, of course, understanding the competiof the static interaction of two spheres. The model empha- 258 tion between quasifission and fusion is very important. Howsises the role of rapid dynamical deformations away from the 259 ever, it is very difficult to distinguish the quasifission frag-205 configuration of two spheres in contact. It also includes dis- 260 ments from the fission ones as they have similar mass distri-206 sipation that leads to the requirement of even higher energies 261 butions. The two processes differ by their time scale and thus 207 to fuse. Soon, experimental evidences confirmed the model 262 angular distribution [34]. Consequently, one lacks of reliable 208 and a lot of efforts were devoted to quantify the reduction in 263 experimental fusion cross-sections.

Although dissipation plays a crucial role in understanding dependent and one has to be careful when using the value 215 has been adopted to study the dynamical diffusion over the

Very recently, we showed that the initial condition of the the experimental uncertainty and the number of data points. 221 formation step slips due to the elimination of the fast vari-Values range from 34 keV for quite accurate data to 0.4 MeV; 222 ables in a multidimensional description and this affects sig-223 nificantly the formation probability [30].

This point of view is challenged by so-called DNS model value of the uncertainty in the fission barrier that cannot be 225 that was developed later. It is based on a very different conovercome. Although less important, other parameters also 226 cept, assuming a frozen configuration and exchange of nucle-

Although progress has been made in understanding the fu-Because of these uncertainties in key parameters together 231 sion hindrance, there is still no consensus on the dynamical with the amplification effect due to the fact that we want to 232 treatment. Thus, it is almost impossible to perform an uncer-233 tainty analysis in such conditions. Moreover, many quantiable to produce predictions with an accuracy lower than about 234 tative ambiguities remain. What is the barrier height of the one order of magnitude for cold fusion reactions. It is more 235 conditional saddle that has to be overcome? It is clear that a 236 small change in its value will have a great impact on the for-237 mation probability. What is the dissipation strength? What is the role of structure effects?

> A comparison between models can give some insights on the amplitude of the discrepancies. Figure 1, reproduced from Ref. [3], shows that the formation probability for cold fusion reactions calculated by various models spans two or three orders of magnitude. Consequently, to improve the predictive 245 the synthesis of superheavy elements, one must constrain the 246 formation probability responsible of the fusion hindrance.

B. Strategies to constrain the formation probability

Fusion-evaporation residue cross sections are of no help

The channel competing with the formation is the so-called

One idea is to consider that reaction dynamics models must 292 els might simply be wrong. For example, large fission barri-265 strive to reproduce experimental data on quasifission. How- 293 ers appearing in the comparison of Refs. [8, 9] would mean 266 ever, constraining models on the dominating channel leads 294 almost stable nuclei that would be easy to synthesise as the 267 to unavoidable uncertainties. And, as for fission-evaporation 295 cross-section would benefit from the large survival probabilcompetition, these uncertainties in quasifission modelling 296 ity. Second, assuming that the model is correct, we can perwill have a large impact on the rare fusion events. Thus, re- 297 form an uncertainty analysis of the predictions in order to estiproducing quasifission data will definitively help to constrain 298 mate the dispersion of the results due to the lack of constraints models but might not allow to get a better agreement in the 299 on many parameters. Such an analysis is too optimistic as it 271 272 formation probability predictions.

273 274 ing reactions leading to the same compound nucleus, one be-275 ing hindered and the other not, in order to get rid of the am- 303 However, for the sole survival step, there is no hope to get 276 biguities in the decay phase of the reaction. Such studies, 304 predictions more accurate than an order of magnitude. This is 277 currently under development, should naturally include uncer- 305 a very pessimistic finding in the case of superheavy elements 278 tainty analysis.

IV. CONCLUSION

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Although it is a difficult task, predictions of reaction crosssections are useless without an uncertainty analysis [35]. 312 282 Moreover, uncertainty analysis is a powerful tool to built up a 313 quires a careful evaluation of the covariances. This will be hierarchy of the various sloppy points of a model. This leads 314 one of the priorities of our future research. to show that predictions of superheavy production crosssections mainly suffer from the large discrepancies in the hindrance phenomenon modelling and fission barriers that can 316 both affect the results by orders of magnitude.

289 First is a comparison between various models. This leads 319 (LIA FJNSP). We thank the RCNP of Osaka University, 290 to an estimate of the error in modelling. However, such a 320 GANIL and Huzhou University for their warm hospitality and 291 comparison can appear to be too pessimistic as some mod- 321 supports.

300 assumes that the model is correct. Therefore, it is not pos-Alternatively, hindrance could be constrained by compar- 301 sible to apply it to the whole reaction as there are still too that are produced in handful numbers.

> If the absolute value of the predicted cross-sections cannot be accurately estimated, the ratio between two values will benefit from the strong correlation between them. This could be cross-sections at different energies or with different targetprojectile combinations. Thus, the predicted trends would be correct. However, estimating the uncertainty in the ratio re-

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