Efficient method for estimation of fission fragment yields of \( r \)-process nuclei

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The knowledge of fission fragment yields of hundreds of nuclei inhabiting very neutron-rich regions of the nuclear landscape is crucial for the modeling of heavy-element nucleosynthesis. In this study, we propose a novel model for a fast calculation of fission fragment yields based on the concept of shell-stabilized prefragments calculated within realistic nuclear density functional theory. The new approach has been benchmarked against experimental data and advanced calculations reaffirming the dominant role of shell effects in the pre-scission region for forming fission yields.

Introduction – The predictive power of \( r \)-process network calculations could be improved by providing sound predictions of fission fragment yields, including yields from spontaneous fission (SF), beta-delayed fission, and neutron-induced fission. In particular, it has been long established that fission fragment distributions play a crucial role in shaping the final \( r \)-process abundances [1–6], and recent studies suggest that uncertainties in fission yields should be reduced in order to properly constrain the contribution of binary neutron star mergers to the chemical evolution of \( r \)-process elements [7]. When it comes to realistic predictions, the Langevin approach [8–10] has proven to be extremely successful in terms of quantitative reproduction of fragment yields. Unfortunately, full-fledged Langevin calculations in a multidimensional collective space, based on the microscopic nuclear density functional theory (DFT) input, are computationally expensive when it comes to large-scale theoretical fission surveys. The same is true for the time-dependent DFT approaches [11–14]. Given the computational cost of microscopic methods and the large number of fissioning nuclei that are expected to contribute during the \( r \)-process nucleosynthesis, network calculations have mostly relied on simple parametrizations or highly phenomenological models to determine fission yields.

To date, the models that aim at global surveys of fission yields can be grouped into three main categories. The first group comprises the so-called Brownian motion models, wherein the fission process is described as an overdamped motion across potential energy surfaces obtained from microscopic-macroscopic calculations [15–17]. These models take into account the dissipative dynamics required to describe the full distribution of fission yields. To deduce charge yields, additional assumptions are made such as a linear scaling of mass distributions. Methods belonging to the second category, the scission-point models [18, 19], are free from this limitation because they treat charge asymmetry as a collective degree of freedom. On the other hand this approach (which recently has been combined with the DFT input [20, 21]) relies on a static description of the fragments along the scission configuration, thus neglecting fission dynamics. Finally, the third category are semi-empirical models, such as GEF [22] and ABLA [23], which have been widely used in \( r \)-process calculations [3–5, 24, 25]. These models turned out to be extremely successful in describing data close to stability, since their parameters have been fine-tuned to reproduce observed fission fragments distributions. However, their applicability to extrapolate far from stability is questionable and the related uncertainties are difficult to assess.

To overcome these shortcomings, the present paper presents an efficient and predictive model for fission fragment yields that is based on the microscopic DFT input. There are two main assumptions behind our model: (i) the formation of fission fragments is governed mainly by shell effects of the prefragments that develop in the pre-scission region; and (ii) the final fragments are produced in the scission region by a rapid distribution of neck nucleons, a process that is statistical in nature. Both assumptions are based on results of previous microscopic calculations [8, 26, 27]. Indeed, by studying the nucleonic localization function (NLF) for deformed configurations of fissioning nuclei, we observe that two distinct prefragments form and then separate well before scission is reached. This early development of the prefragments is a manifestation of the freeze-out of single particle energies along the fission path [28–31] as the system tries to maintain its microscopic configuration to avoid level crossings. The suggestion that the prefragment particle numbers play essential roles in determining the fission fragment distribution can be traced back to strong shell effects in prefragments in the semiclassical periodic-orbit theory [32, 33].

Thus, a deformed fissioning nucleus can be thought of as two well-formed prefragments connected by a “glue” of nucleons in the neck. This concept is illustrated in Fig. 1, which shows the neutron and proton prefragments, and the neck nucleons. Within this concept, it is not necessary to calculate the full potential energy surface (PES) up to the scission configuration, since a simplified estimation of the PES is sufficient to obtain...
the most-probable prefragment configuration(s). Then, final fragment yields are generated by distributing the neck nucleons into prefragments according to a statistical prescription wherein the frequency of a particular fragmentation channel is decided with an appropriate microcanonical probability \[18, 34\]. Our model is described in the next section, followed by benchmark calculations performed for several nuclei. We then study two representative cases of $r$-process nuclei and compare our results with those obtained with more phenomenological models.

\[\text{FIG. 1. Density distributions of prefragments (bottom) and neck-nucleons (top) in the configuration } C, \text{ indicated in Fig. 2, of } ^{254}\text{Fm obtained using UNEDF1}_{HFB}.\]

**Theoretical framework** – Our method of estimating primary fission yields is based on NLFs computed at a point on the most probable fission path that corresponds to a compact configuration at the outer turning point well before scission. This configuration, obtained by minimizing the collective action between the ground state and the outer turning line, determines the peak position of the fragment yield distribution. As discussed earlier, NLFs are related to the underlying shell structure and remain almost unaffected along an effective fission path \[26, 35, 36\]. Moreover, multiple nuclear configurations may contribute in the yield distribution if several static fission pathways are close in energy (multimodal fission) \[37\]. For example, both mass-symmetric and mass-asymmetric structures must be accounted for when the corresponding fission valleys are close. For neutron-induced fission, the PES is obtained using the finite-temperature Hartree-Fock-Bogoliubov (HFB) formalism, in which the compound nucleus is described using the grand-canonical ensemble \[38–40\].

Given these general considerations, our approach to fission-yield estimates can be summarized as follows. First, for a given nucleus and fission decay (spontaneous, neutron-induced or fusion-fission) we find the point where the total energy drops down from the fission barrier to its ground state value ($E_{GS}$) along the most-probable fission path. In case of SF, this point lies on the outer turning line. For this configuration, called $C$ in the following, the two prefragments are identified using nucleonic densities and NLFs. The remaining nucleons are assumed to belong to a neck region, see Fig. 1. In the second step, the neck nucleons are distributed among the two prefragments with all possible combinations sampled. For each combination, binding energies of the resulting fragments ($E_{b1}$ for the fragment $(A_1, Z_1)$ and $E_{b2}$ for the fragment $(A_2, Z_2)$) are calculated using the isospin-dependent liquid drop model \[41\] (it is to be noted that the minimum mass/charge of a fragment is that of a corresponding prefragment). The energy of each fragment combination is $E_r = E_t - (E_{b1} + E_{b2} + E_C)$, where $E_t$ is the calculated energy of the fissioning nucleus at $C$ (which is equal to $E_{GS}$) and $E_C$ is the electrostatic repulsion energy between the fragments (for simplicity, assumed to be that of two point charges). In the third step, each pair of fragments is associated with a microcanonical probability distribution \[18\] (see Eq. 2 in Supplemental Material \[42\]). Finally, we fold the probabilities with a Gaussian smoothing function of width $3$ for $A$ and $2$ for $Z$ \[8\].

The prefragments are determined using the following algorithm. Along the nuclear symmetry axis, we identify the center of the prefragment as the NLF maximum (or minimum) placed at the center of the NLF’s concentric shell rings (see Fig. S1 in Supplemental Material \[42\] for NLF plots). In this way, the equatorial plane passing through this center delimits the prefragment’s hemisphere. Proton and neutron numbers of prefragments are obtained by integrating the nucleon density over such hemispheres and doubling the resulting number. As shown in Fig. 1, the remaining nucleons are assumed to constitute the neck. This method is a generalization of the previous definition \[26\].

The microscopic DFT input has been generated using the HFB solver HFODD \[43\]. To test the robustness of our predictions to the choice of effective interaction, we applied two Skyrme energy density functionals (EDFs) in the particle-hole channel: SkM* \[44\] and UNEDF1$_{HFB}$ \[45\]. In the pairing channel, we took the mixed-type density-dependent delta interaction \[46\].

**Results** – To benchmark our model, we consider several nuclei ($^{178}\text{Pt}$, $^{240}\text{Pu}$, $^{254}\text{Cf}$, and $^{254,256,258}\text{Fm}$) for which experimental fission yield distributions are available. For each of these systems, an appropriate configuration $C$ is identified to extract the prefragments. In case of thermal fission ($n_{th}$, $f$), we calculate the PES at the thermal excitation energy ($\approx 6 \text{ MeV}$) and we select the configuration $C$ by taking the point on the static fission path that has zero energy with respect to the ground state minimum obtained at $E^* = 6 \text{ MeV}$. For pre-actinide nuclei such as $^{178}\text{Pt}$, the potential barrier is fairly flat and broad, and it is crossed by the fusion valley \[47, 48\]. Therefore, we have arbitrarily chosen a configuration on the static path of $^{178}\text{Pt}$ \[48\] at $Q_{20} = 220 \text{ b}$. We checked that the prefragment-structure does not change beyond this point.
The isotopes of Fm are of special interest as the corresponding mass yields show a transition from asymmetric to a symmetric distribution with increasing parent mass \([37, 49–51]\). Calculated PESs are shown in Fig. 2. For all the systems except \(^{258}\)Fm, fission fragment distributions are expected to be asymmetric because of a large outer fission barrier. However, the potential surface of \(^{258}\)Fm is rather flat at large \(Q_{20}\) and we expect contributions from both symmetric and asymmetric fission pathways.

Figure 3 shows the calculated prefragments for different fissioning systems. We notice that for the actinides the heavy prefragment is very close to the doubly-magic nucleus \(^{132}\)Sn. This result suggests an early development of shell effects along the fission path, in agreement with the previous NLF-based study on the formation of \(^{240}\)Pu fission fragments \([26]\). On the other hand, both mass and charge of the lighter fragment are subjects to appreciable fluctuations; this supports the need for microscopic calculations in the prefragment determination. As already pointed out in Ref. [26], the main advantage of using NLFs is that the localization patterns of the prefragments defined at the outer turning line closely resemble those at scission. This is confirmed by the fact that both proton and neutron single particle levels show a smooth pattern in their evolution from the outer turning point to the scission point, indicating the stabilization of shell effects (see Supplemental Material \([42]\) for more discussion).

Since the notion of a prefragment is a purely theoretical concept, their properties cannot be measured experimentally. Moreover, alternate prefragment definitions exist \([31, 52]\). Consequently, the validity of a prefragment-based description is checked by the ability of a model to predict and reproduce experimental observables. To this end, in Fig. 4, we benchmark our approach by predicting the fission fragment mass and charge distributions for selected nuclei. In general, very good agreement with experiment has been obtained for SF. For thermal fission, one does not expect a drastic change compared to SF as the excitation energy is quite low. Indeed, for \(^{240}\)Pu\((n_{th}, f)\) both experiment and predictions overlap with the SF fragment yields. For \(^{256}\)Fm\((n_{th}, f)\) we predict broader mass and charge distributions than for SF. We note that the \(^{254}\)Fm configuration of \(^{254}\)Fm and \(^{290}\)Fm are predicted to fission symmetrically; hence, only one symbol is shown.
In the present exercise, we extended our calculations to \( ^{254}\text{Pu} \) and \( ^{290}\text{Fm} \) as two representative fissioning nuclei that are expected to significantly contribute to the \( r \)-process nucleosynthesis occurring in neutron star mergers \[5\]. A one-dimensional fission trajectory is calculated for each isotope up to the fission isomer by constraining \( Q_{30} \) and leaving the other degrees of freedom unconstrained. In the region between the fission isomer and the outer turning line, the collective space is expanded to include \( Q_{30} \) in order to account for mass asymmetry. The resulting PESs and yield distributions are shown in Fig. 5 and the yields are compared with predictions given by the semi-empirical GEF \[22\] and ABLA \[23\] models.

Our method and the GEF model give similar results for \( ^{254}\text{Pu} \), which is predicted to split asymmetrically, while ABLA fission yields are shifted towards a more symmetric distribution. Conversely, for the \( ^{290}\text{Fm} \) our approach predicts a symmetric fission centered around \( ^{142}\text{Sn} \), but with a wider fragment distribution compared to GEF and closer to the width predicted by the ABLA model. This result suggests a weakening of \( Z/N = 50/182 \) shell stabilization for very neutron-rich nuclei, in agreement with a general trend found in recent calculations \[17\] that would extend the range of \( r \)-process elements affected by fission cycling \[6\]. It is important to notice that while in both GEF and ABLA the effect of shell stabilization determining the fission modes is introduced \textit{ad hoc} using parameters finely tuned to reproduce experimental data, in our approach they naturally emerge from the underlying EDF. This is a crucial feature in the study of nuclei placed far from stability, where shell closures can differ from those found close to stability. A recent example is the predicted cluster decay of \( ^{294}\text{Og} \), where fission fragment distributions are dominated by the heavy fragment of doubly-magic \( ^{208}\text{Pb} \) \[27, 61\]. We benchmarked our model with the full-fledged Langevin predictions of Ref. \[27\] and found a good agreement between the two calculations (see discussion in the Supplemental Material \[42\]). This again confirms the ability of our model to capture the relevant physics. Finally, we have tested that our results are insensitive to the particular realization of the liquid drop model and other parameters defining the microcanonical probability.

**Conclusions** — We developed a method for estimating fission fragment yields based on nuclear DFT with realistic energy density functionals and simple statistical...
assumptions governing the redistribution of neck nucleons at scission. This method is inspired by recent microscopic Langevin calculations suggesting an early formation of the prefragments along effective fission paths and a rapid distribution of neck nucleons around the scission point [26], which is reminiscent of the separability principle proposed in previous phenomenological studies [62]. The new approach is fast and computationally-expensive compared to microscopic models that require the knowledge of the full multi-dimensional PES and related quantities (such as the collective inertia) all the way to scission, while retaining at the same time the relevant physics determining the main structure of fission fragment distributions. A sound agreement with experimental charge and mass distributions is obtained for a wide range of fissioning nuclei.

In the next step, we intend to use the new method to carry out global calculations of fission fragment distributions across the $r$-process region. Such input could be combined with systematic calculations of fission rates [63–66] to consistently study the impact of fission on the $r$-process nucleosynthesis.

Acknowledgements — The authors are grateful to G. Martínez-Pinedo and N. Vassh for providing the ABLA and GEF data, respectively, used in Fig. 5. This work was supported by the U.S. Department of Energy under Award Numbers DE-DE-NA0002574 (NNSA, the Stewardship Science Academic Alliances program) and DE-SC0008511 (Office of Science, Office of Nuclear Physics NUCLEI SciDAC-3 collaboration). Part of this research was also performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Computational resources were provided through an INCITE award “Computational Nuclear Structure” by the National Center for Computational Sciences (NCCS), and by the National Institute for Computational Sciences (NICS). Computing support for this work also came from the Lawrence Livermore National Laboratory (LLNL) Institutional Computing Grand Challenge program.

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