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Description of isotopic fission-fragment distributions within the Langevin approach

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Dynamical calculations based on the four-dimensional stochastic Langevin approach are extended to the description of fragment isotopic distributions in fission at moderate-to-high excitation energy. Benefiting from previous theoretical work on charge-asymmetry relaxation times, and existing experimental charge-dispersion curves from low-energy fission, charge fluctuations are introduced at the scission point in an effective way. Comparison with recent suited measurements shows a good description over the full fragment production. The development proposed in this work can easily be implemented in other codes used in the field.

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Introduction. Fission, the dynamical process that results in the division of a compact nucleus into two distinct fragments, depends on the intricate interplay between the potential energy landscape of the system, its inertia, and the strength of dissipative forces inside the nuclear medium.

From a theoretical point view, no unified model exists for fission over a wide range of system mass, excitation energy, and angular momentum. Powerful models have been developed for specific branches in the field. With rising temperature, dynamical effects take an increasingly important role, and it has become customary to model the fission process within a transport theory, which distinguishes between collective and intrinsic degrees of freedom [1]. In recent decades, the stochastic approach based on the integral Langevin classical equation of motion for the collective coordinates has been widely developed [2]. Codes involving a high-dimensionality phase-space [3] and suited down to low excitation energy [4] are now available. An approach in terms of a Metropolis random walk [5] was recently proposed for low-energy fission, and has shown powerful for specific purposes.

To date, existing dynamical models of fission have concentrated on fragment mass A, angle θ, and total kinetic energy TKE distributions, as well as the coincident light particles evaporated either before or after scission. Widespread experimental information exists on these observables, and comparison with predictions by Langevin models permitted a considerable progress in the understanding of fission dynamics at moderate and high excitation energies (see Ref. [2] and therein). Measurements on fission-fragment nuclear charge Z are, in contrast, very scarce, due to the difficulty of identifying in experiment atomic numbers typical for fission fragments ($20 \leqslant Z \leqslant 60$). Some data obtained from direct measurement in a heavy-ion spectrometer exist for fissioning actinide systems at low energy (see, e.g., Refs. [6,7]), but only Z of the light fragment could be determined. Data using indirect techniques [8–15], based on radiochemistry, or on γ or x-ray spectroscopy (prompt or delayed), can give access to the charge of the heavy fragment as well. But, according to the identification method itself, a restricted part of the fragment production is covered, only. A new approach, based on fission induced in inverse kinematics, was introduced by Schmidt *et al.* [16] in the late 1990s at GSI, Darmstadt. It permitted for the first time determining with high resolution the nuclear charge of the whole range of fragments. Unfortunately, fragment masses were not available in this setup. Inspired by the GSI experiment, a further step was recently done at GANIL, Caen [17]. Fission of ^{250}Cf at an excitation energy $E^* = 45$ MeV was induced in inverse kinematics using ^{238}U (6.1 MeV/nucleon) + ^{12}C collisions. The large-acceptance VAMOS spectrometer with a dedicated detection system was employed to identify one of the fragments, and determine its mass and charge with high resolution [18]. For the first time, complete and high-quality isotopic fission-fragment distributions became available for fission around the Coulomb barrier.

According to the limited experimental information available till recently, dynamical models of the Langevin type tried neither to model, nor to investigate, fragment nuclear charges in detail. Most of the models focus on proper determination of mass division. Atomic number partition is derived, using a "sharp" univocal correlation between A and Z once scission is reached. That is, charge fluctuations are completely neglected. It is the goal of this work to benefit from the recent data set [17] on complete (A,Z) fragment production yields, to improve upon the capability of current Langevin models to describe *isotopic* distributions in moderate-to-high excitation energy fission.

In the following a brief description of the theoretical framework is given, while implementation of charge fluctuations in the model is developed in detail. Results are discussed in comparison with experimental data, followed by a summary and conclusions.

Theoretical framework and development. The model used in this work is based on the stochastic classical approach of fission dynamics [1]. Dynamical calculations were performed with the four-dimensional Langevin code developed recently by Nadtochy and collaborators [3]. We restrict here to a presentation of the main ideas and ingredients of the model, and refer to Ref. [3] for further details.

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a. Four-dimensional Langevin dynamics. Fission is modeled considering four collective coordinates: Three variables describe the shape of the nucleus, and a fourth coordinate corresponds to the orientation of its angular momentum relative to the symmetry axis. The evolution of these variables is computed, time step by time step, from the solution of the multidimensional classical equation of motion. Deexcitation of the system by evaporation of light particles prior to scission, and by the fragments after scission, is simulated along the time evolution by employing the Monte Carlo approach.

It is emphasized that the theoretical framework used in this work does not account for microscopic effects. All ingredients are based on either classical or macroscopic concepts. The model is therefore not suited for low-energy fission. In contrast, it has been shown to be impressively powerful in describing a wide set of experimental observables for fission at moderate to high E^* (above about 30 MeV). See Refs. [2,3] and therein.

b. Charge fluctuations. Once the scission point is reached, mass division is determined according to the volume of the nascent fragments. In any Langevin code, whenever the dynamics of the charge asymmetry degree of freedom is not treated by means of an explicit collective coordinate, an assumption has to be made about partitioning of protons between the fragments. A number of empirical hypotheses have been proposed by various authors [19,20]. In the code used in this work, proton partitioning is based on the unchanged charge density (UCD) assumption, which is a reasonable approximation for the most probable charge at fixed mass at medium to high E^* [21]. It implies conservation of A/Z between the fissioning nucleus and the fragments at scission. For a specific mass split, the atomic number of the fragments is univocally defined by

$$Z_{\mathrm{FFi}}^{\mathrm{UCD}} = \frac{A_{\mathrm{FFi}} Z_{\mathrm{fiss}}}{A_{\mathrm{fiss}}}, \tag{1}$$

where A_{FFi} (Z_{FFi}) is the mass (charge) of the ith fragment before post-scission evaporation, and A_{fiss} (Z_{fiss}) is the mass (charge) of the fissioning nucleus. Sharing of neutrons straightforwardly follows from mass and charge. In such a sharp Z partitioning scheme, charge fluctuations are nonexistent: the isobaric charge variance σ_Z, which corresponds to the width of the fission-fragment Z distribution at fixed A (often referred to as charge-dispersion [22]), is zero. To our knowledge, the absence of charge fluctuations is common to all available Langevin codes used at day, although the most probable charge may be derived from one or another hypotheses [2–4,23–29].

The consequences of disregarding charge fluctuations could not be investigated so far in medium-to-high energy fission, since suitable experimental information was scarce. The recent experimental progress [17,18] mentioned in the Introduction permits us now to investigate this point, and possibly develop the model in this respect. According to the simplified $\sigma_Z = 0$ assumption, it seems reasonable to expect that the calculated isotopic fission-fragment distributions would be too narrow. This conjecture was already suspected in our previous work [30].

In Refs. [31,32] the authors for the first time proposed to consider charge asymmetry as a collective coordinate. Having introduced the possibility of different repartitions of the neutrons and protons for any shape, they found that statistical considerations are applicable to the charge-asymmetry degree of freedom. In Ref. [33], Karpov and Adeev went a step further by investigating the dynamics of the charge-asymmetry coordinate, explicitly including it in the equation of motion. They observed that the relaxation time of charge asymmetry does not exceed a few 10^{-21} s, independent of viscosity. That is, it is smaller than the time characteristic of collective modes responsible for deformation. Charge asymmetry can therefore be assumed to be equilibrated at any point along the trajectory, and its explicit dynamical treatment can reasonably be replaced by statistical considerations close to scission. Within the statistical limit applied at scission, the charge variance is given by $\sigma_Z^{\mathrm{stat}} = \sqrt{T_Z(q_{sc})/C_Z(q_{sc})}$, where T_Z and C_Z correspond, respectively, to the effective temperature and stiffness in the charge-asymmetry direction. Karpov and Adeev [33] obtained that σ_Z^{stat} varies slightly with E^*, from 0.6 to 0.8 for $10 \leqslant E^* \leqslant 100$ MeV. These estimates of σ_Z^{stat}, and its near constancy, are in very good agreement with the charge variances extracted in low-energy fission experiments [6,7,34,35]. Guided by these results, in the present work,

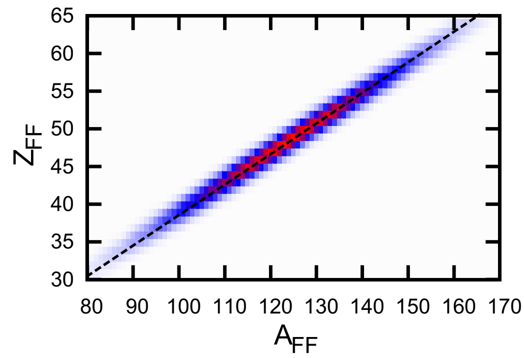

FIG. 1. (Color online) Calculated (A_{FF}, Z_{FF}) production for fission of ^{250}Cf at $E^* = 45$ MeV and with $\sigma_Z = 0.8$. The result assuming $\sigma_Z = 0$ is shown with a thick dashed line.

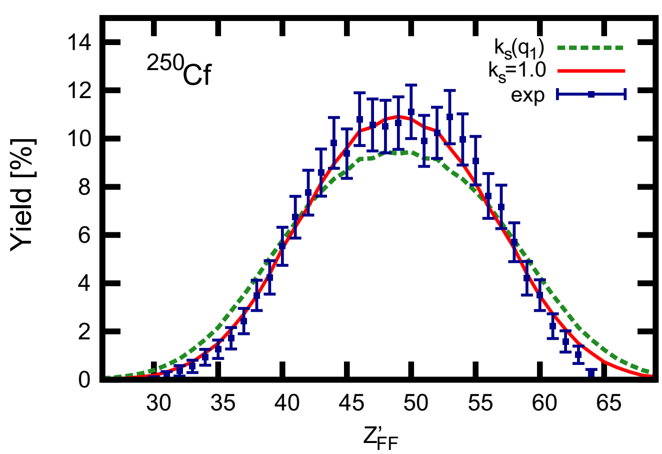

FIG. 2. (Color online) Fragment charge distribution for fission of ^{250}Cf at $E^* = 45$ MeV: Experimental data [17] (full squares) are compared to calculations obtained with $k_s = 1$ (full red line) and the $k_s(q_1)$ (dashed green line); see text. In both cases, $\sigma_Z = 0.8$.

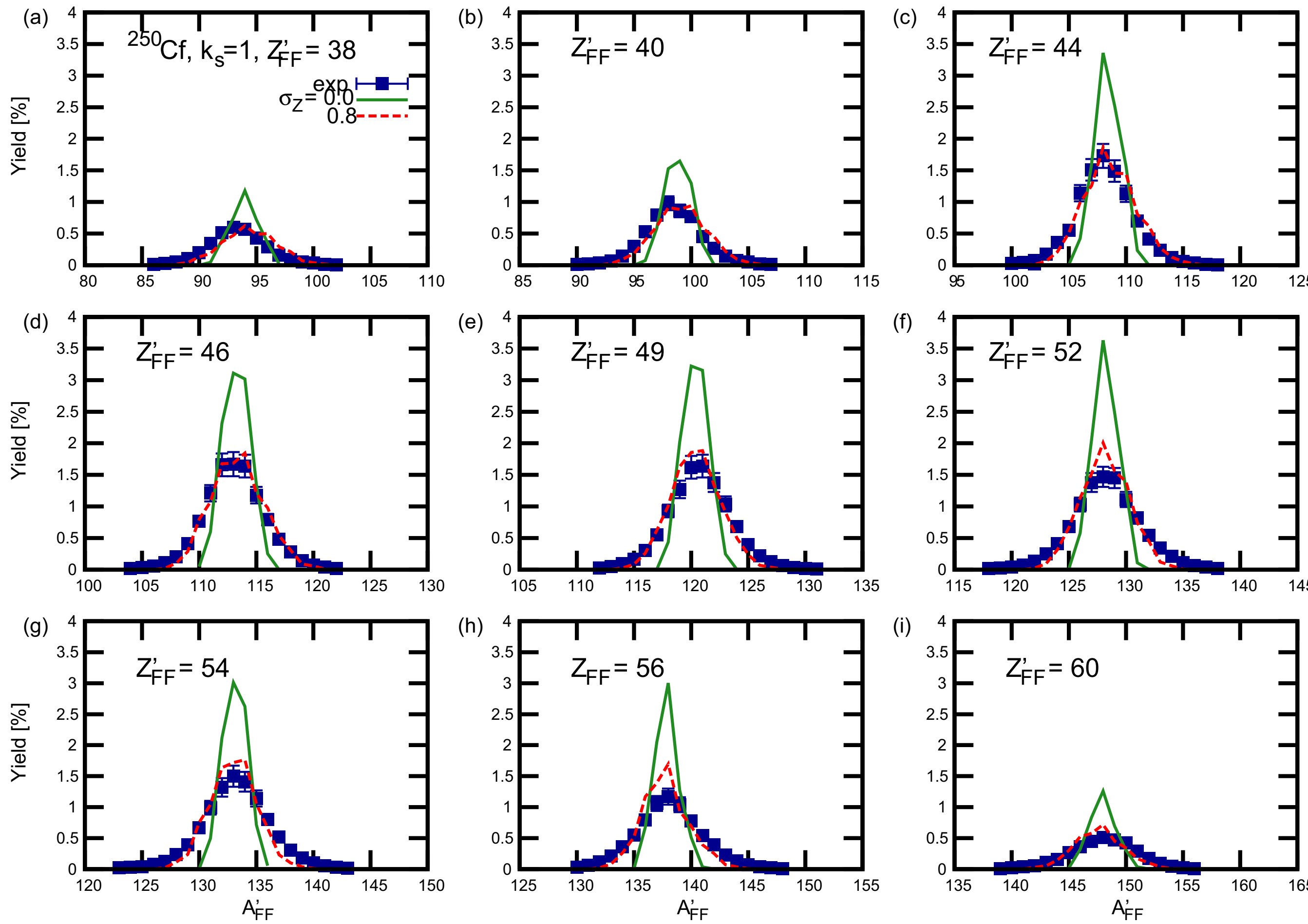

FIG. 3. (Color online) Fragment isotopic distribution for fission of ^{250}Cf at $E^* = 45$ MeV for elements between Sr and Nd. Experimental data [17] (full squares) are compared to calculations obtained without (full green line) and with (dashed red line) account of charge fluctuations with $\sigma_Z = 0.8$.

we adopt the following prescription for modeling charge dispersion. Once scission is reached, mass partitioning is determined by the volume of the nascent fragments, and the mean Z of the fragments is computed from the UCD assumption, similarly to the procedure used so far [3]. Charge dispersion is next introduced by sampling the atomic number of one the fragments, e.g., Z_{FF1}, from a Gaussian distribution centered around $Z_{\mathrm{FF1}}^{\mathrm{UCD}}$ and of finite width σ_Z. The charge of the second fragment follows from $Z_{\mathrm{FF2}} = Z_{\mathrm{fiss}} - Z_{\mathrm{FF1}}$. In the present work, according to the above discussion, σ_Z is assumed to be a constant, and its numerical value is borrowed from the aforementioned low-energy fission experiments and theoretical predictions. Refinements may be introduced as a next step.

Results. Calculations were performed for two values of the isobaric charge variance, $\sigma_Z = 0.6$ and 0.8, which cover the range of theoretical [33] and low-E^* experimental [6,7,34,35] results. The effect of the modification implemented in the model is illustrated in Fig. 1 for $\sigma_Z = 0.8$. The fragment production, before post-scission evaporation, is shown in the (A_{FF}, Z_{FF}) plane. For a given mass, several charges are populated. In contrast, under a sharp partitioning, given here by the UCD assumption and represented with a dashed line, only one charge is observed for a specific mass.[1]

In the following, the calculation is compared to the experiment of Ref. [17] in detail. The latter gave access to the mass and charge of the cold fragments. Hence, unless specified, we restrict hereafter to post-scission evaporation masses and charges. These are denoted A'_{FF} and Z'_{FF}, respectively, to distinguish them from the same quantities before fragment deexcitation. Since fission fragments evaporate almost exclusively neutrons, $Z'_{\mathrm{FF}} = Z_{\mathrm{FF}}$ applies in most cases.

The measured and calculated integral fission-fragment element distributions are shown in Fig. 2. The calculation is displayed as obtained using two prescriptions for friction in the shape variables: the wall-and-window formula [36,37] with nominal strength, $k_s = 1$, and the chaos-weighted wall formula [38], $k_s(q_1)$. In both cases, σ_Z is taken equal to 0.8. The calculation with $k_s = 1$ rather finely follows the experimental distribution, while the prediction by $k_s(q_1)$ yields a slightly too broad distribution [3]. The result for $\sigma_Z = 0.6$ is very similar to that for $\sigma_Z = 0.8$, and thus it is not shown.

[1] Rounding of Z_{FF} can yield two populated charges for one mass.

Introduction of charge fluctuations leads to a redistribution of the fragment isotopic composition, while leaving the integral Z'_{FF} distribution nearly unchanged. The description of the data can be improved further with adjustment of the model ingredients. Such a tuning is not the purpose of this communication. Apart from σ_Z which is the focus of the work and which we are varying, we adopted all along a unique set of model parameters borrowed from previous investigations [3]. Figure 2 thus gives an idea about the theoretical uncertainty range. In the remainder of this paper, we will restrict to results obtained with the wall-and-window formalism with $k_s = 1$.

Light-particle multiplicities and fragment *TKE* could not be measured with the setup of Ref. [17]. We have compared the presently calculated values with data on nearby systems [39] and systematics [2,3]. Good achievement is observed for pre- and post-scission neutron multiplicities, as well as for the *TKE*.

Having ensured that the calculation describes bulk observables, we can turn to a detailed survey of the isotopic content of the fragments. For a selection of elements, from Sr to Nd, measured and calculated isotopic distributions are compared in Fig. 3. While the calculation neglecting charge fluctuations, $\sigma_Z = 0$ (full green line), yields much too narrow distributions as anticipated, a pretty good description is obtained with the predictions accounting for charge fluctuations and assuming $\sigma_Z = 0.8$ (dashed red line). As for fine tuning, another (other than UCD) prescription of most probable charge division could further make the calculated peak positions perfectly match the experimental ones [29].

The first and second moments of the fragment isotopic distributions are known as relevant tools to get insight into the fission process [6,7,34,40]. They are affected by both the influence of structural effects in the primary fragment production, and subsequent neutron evaporation. The mean $\langle N'_{FF}\rangle/Z'_{FF}$ ratio, characterizing the neutron excess of the fragment, and the isotopic width $\sigma(N'_{FF})$ are considered, respectively, in Figs. 4 and 5, as functions of fragment nuclear charge. The same quantities as calculated prior to post-scission evaporation

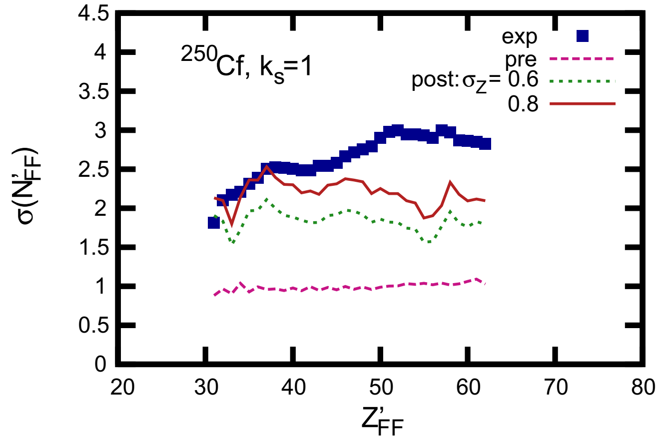

FIG. 5. (Color online) Identical to Fig. 4 for the isotopic width $\sigma(N'_{FF})$.

are displayed as well. Note that, while the isobaric width σ_Z characterizes the Z distribution at fixed A, the isotopic width considered in Fig. 5 is related to the N (equivalently, A) distribution at fixed Z. The neutron-richness $\langle N'_{FF}\rangle/Z'_{FF}$ predicted for the final fragment follows the experimental constant trend. The ratio $\langle N_{FF}\rangle/Z_{FF}$ before post-scission evaporation is related to the ratio of the fissioning nucleus ($\approx$1.55), according to the UCD assumption. Thus, the difference between $\langle N'_{FF}\rangle/Z'_{FF}$ and $\langle N_{FF}\rangle/Z_{FF}$ is a direct measure of the total neutron multiplicity. The average calculated value is around 9, in good agreement with the information extracted for this system from prompt γ-ray measurements [41]. Figure 5 shows that the broadening of the fragment isotopic distribution due to post-scission evaporation is predicted to be large (see the substantial difference between the dashed pink and the full red or dotted green curves). The $\sigma(N'_{FF})$ predictions are in reasonable agreement with experiment for $\sigma_Z = 0.8$. Nevertheless, while the measurement shows a tendency of $\sigma(N'_{FF})$ to increase with increasing Z'_{FF}, the calculation exhibits a flat trend. The calculated distribution is slightly too narrow for the heaviest fragments (see also Fig. 3). This discrepancy may be attributed to the oversimplified assumption of a constant isobaric variance [7,33].

Summary and conclusions. Dynamical calculations based on the four-dimensional stochastic Langevin approach are extended to the description of fragment isotopic distributions in fission at moderate-to-high excitation energy. Charge fluctuations are introduced in an effective way, by sampling from a Gaussian distribution, centered at the UCD predicted charge and of a width σ_Z borrowed from previous theoretical and experimental results. The predictions by the update model are compared with recent measurements on isotopic yields from fission of ^{250}Cf at $E^* = 45$ MeV. The achievement is impressively good, keeping in mind the simplicity of the procedure. Remaining discrepancies are ascribed to the dependence of the isobaric width σ_Z on excitation energy, fissioning system fissility, and fragment charge, which can be improved on.

The procedure developed in this work for simple but efficient implementation of charge fluctuations can readily be

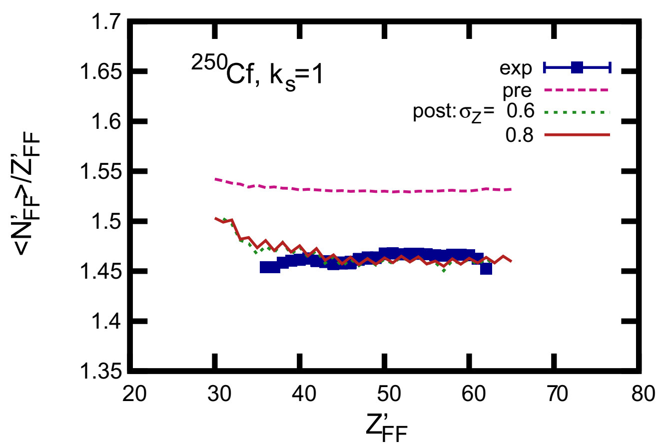

FIG. 4. (Color online) Fragment neutron excess as a function of fragment charge for fission of ^{250}Cf at $E^* = 45$ MeV. Experimental data [17] (full squares) are compared to calculations assuming $\sigma_Z = 0.6$ (dotted green line) and $\sigma_Z = 0.8$ (full red line). The result before post-scission evaporation is also shown (dashed pink line).

plugged in other stochastic models used in the field. It will permit the analysis of correlations between more observables, enabling us to constrain model parameters, and getting insight into the underlying dynamics of fission.

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