On current ambiguity in the interpretation of fission at intermediate excitation energy

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A B S T R A C T

Various approaches are currently used to interpret experimental data on fission. We critically examine a wide set of observables measured for fission of ^{206,210}Po nuclei at medium excitation energy, and illustrate the ambiguity in current analysis. Dynamical calculations based on the four-dimensional Langevin equation using a macroscopic potential energy landscape are performed, and found to consistently describe available measurements. This observation calls into question the robustness of recent analysis based on statistical-model calculations and concluding, on the contrary, to substantial shell effects at the fission saddle point in ^{206,210}Po. The inconsistency in interpretation reached by the two approaches shows that, depending on the system, the conclusion can be strongly model-dependent. Although this may not be surprising, it emphasizes the today still limited reliability of firmly extracting fundamental nuclear properties from customary analysis.

1. Introduction

The complex process of re-arrangement of a compact compound nucleus into two separated fragments remains a puzzle for current understanding, even 75 years since the discovery of nuclear fission. A major difficulty is to un-fold the influence of the various aspects that enter into play, and whose strength is system-dependent. At the same time, the fission mechanism constitutes a rich laboratory for investigating fundamental nuclear properties, and it is of key importance in applied science. Therefore, much effort was, and is still being, devoted to understand and model fission.

Reaching a robust understanding of the fission process depends on the respective magnitude of the various effects. Low-energy fission of actinides is primarily determined by static considerations based on the potential energy landscape as crucially modulated by microscopic shell effects. Fission of low-fissility compound nuclei populated at high excitation energy (E^*) and/or angular momentum (L) depends on the macroscopic Liquid-Drop-like part of the potential energy, and is usually strongly influenced by dynamical effects. With rising excitation energy and decreasing fissility, one thus goes from one regime of fission to another. In this context,

fission of compound nuclei with $A \approx 200$ at intermediate excitation energy ($E^* \approx 30$–60 MeV) is most challenging, lying somewhere in the transition region. The progressive (more or less controlled) washing out of some effects, and the appearance of (more or less controlled) other influences, can lead to subtle compensation effects, adding to the difficulty of singling out un-ambiguously specific aspects.

Statistical-model analyses are most often used to interpret experimental fission observables [1]. This framework gathers a variety of models, using a variety of ingredients and parameters. Thus, discordant results as obtained with different statistical-model codes are not rare (see discussion in Ref. [2] for recent status). Another approach consists in a dynamical treatment based on the stochastic solution of the classical Langevin equation of motion [3]. This (more time-consuming) time-dependent approach goes beyond some limitations of the statistical model, taking consistently into account the simultaneous action of the driving potential, inertia and dissipation effects, along the evolution of the system. During the last decade, elaborate Langevin codes have been developed, involving a high-dimensionality phase-space [4] and suited down to low E^* [5]. Yet, as in the statistical model, the knowledge about some ingredients is limited. A hybrid approach in terms of a Metropolis random walk [6] was recently proposed and has shown powerful for specific purposes.

As a consequence of the aforementioned uncertainty, caution is the order when attempting to interpret experimental data guided

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by model calculations. Among most critical parameters in the description of fission are [2]: the fission barrier B_f, and connected to it, the magnitude of shell effects and the rate of their washing out with E^*, the level density parameter in the ground state (a_n) and at the saddle point (a_f), the influence of L, and the influence of the dynamics. Depending on the sensitivity of the available experimental information, it is impossible to discriminate between different interpretations, and the reliability of the conclusion drawn about nuclear properties is questioned. Analyses within the statistical model are particularly subject to such ambiguities. A dynamical approach of the Langevin type, although it is not free of variable ingredients, has the advantage that it implicitly includes several of the features which have, either to be mocked up, or to be treated as fitting parameters, in the statistical model.

As noted above, fission of medium-fissility compound nuclei at intermediate excitation energy is among most difficult to interpret un-ambiguously. In a recent work dedicated to fission of ^{206,210}Po at $E^* \approx 49$–63 MeV, and based on a careful statistical-model analysis, Golda et al. [7] concluded to substantial shell effects at the fission saddle point. This result is surprising. A recent study of the topographical properties of fission barriers [8] shows that, in the considered mass region, the topographic theorem [9] of close-to-zero saddle-point shell energies does indeed not strictly apply. Yet, the deviation from the theorem remains small, and the macroscopic saddle-point energy is conjectured to be a good approximation [8]. According to the significance of the result of Ref. [7], we propose in this work to examine the robustness of the conclusion. We perform dynamical calculations for the reaction systems measured in Ref. [7] solving a four-dimensional (4D) set of Langevin equations, which use for the potential energy landscape a purely macroscopic prescription. We find that, within this approach, no shell effects at the saddle point are required to be introduced for a consistent description of the experimental data. This observation suggests that the demonstration of Golda et al. [7] is not an irrefutable proof for substantial shell corrections at the fission saddle point.

The paper is organized as follows. In Section 2 a brief description of the theoretical framework is given. Results of the model calculations are gathered in Section 3, and compared with experiment. The conclusions drawn in previous publications within the statistical-model approach are discussed in Section 4 in the light of the conclusion from the Langevin calculation. Summary and concluding remarks are given in Section 5.

2. Description of the model

The dynamical calculations discussed in the next section were performed with the advanced 4D Langevin code recently developed in Refs. [10,4]. We restrict here to a presentation of the main ideas and ingredients of the model, and we refer to the quoted references for further details.

In the stochastic approach fission is modeled considering the most relevant degrees of freedom as collective coordinates. Their evolution with time is treated as the motion of Brownian particles, which interact stochastically with the larger number of internal degrees of freedom constituting the surrounding "heat bath". In the present model, four collective coordinates are considered: Three variables describe the shape of the deforming nucleus, and the fourth one corresponds to the orientation of its angular momentum relative to the symmetry axis. The shape coordinates (q_1, q_2, q_3) are derived from the well-know (c, h, α) parametrization [11], and the projection K of the total angular momentum onto the symmetry axis of the fissioning nucleus is chosen for the fourth so-called tilting mode [12].

The evolution of the shape coordinates is computed from the coupled Langevin equations of motion:

$$\frac{dq_i}{dt} = \mu_{ij} p_j,$$
$$\frac{dp_i}{dt} = -\frac{1}{2} p_j p_k \frac{\partial \mu_{jk}}{\partial q_i} - \frac{\partial F}{\partial q_i} - \gamma_{ij} \mu_{jk} p_k + \theta_{ij} \xi_j(t) \qquad (1)$$

where $\mathbf{q}$ is the vector of collective coordinates, and $\mathbf{p}$ is the vector of conjugate momenta. The driving potential is given by the Helmholtz free energy $F(\mathbf{q}, K) = V(\mathbf{q}, K) - a(\mathbf{q})T^2$, with $V(\mathbf{q}, K)$ being the potential energy, $a(\mathbf{q})$ the level-density parameter and T the temperature of the system. The quantity $\|\mu_{ij}(\mathbf{q})\| = \|m_{ij}(\mathbf{q})\|^{-1}$ refers to the tensor of inertia, and $\gamma_{ij}(\mathbf{q})$ is the friction tensor. The last term of the right-hand side of Eq. (1) describes the stochastic nature of the process, which is assumed to be Markovian. It is related to friction with $\sum \theta_{ik} \theta_{kj} = T \gamma_{ij}$. The potential energy $V(\mathbf{q}, K)$ is calculated within the framework of the macroscopic model with a finite range of the nuclear forces [13], and the prescription of Ignatyuk [14] is used for the level-density parameter. Calculation of inertia uses the Werner–Wheeler approximation of an incompressible irrotational flow [15], and friction is derived assuming the chaos-weighted one-body dissipation formalism [16].

The dynamics of the K coordinate is obtained from the solution of an over-damped Langevin equation as proposed in Ref. [12], and solved in parallel with Eq. (1):

$$dK = -\frac{\gamma_K^2 I^2}{2} \frac{\partial V}{\partial K} dt + \gamma_K I \sqrt{\frac{T}{2} dt} \xi(t) \qquad (2)$$

where $\xi(t)$ is as in Eq. (1), and γ_K determines the strength of the coupling between K and the heat bath.

De-excitation of the system by evaporation of light particles prior scission, as well as by the fragments after scission, is taken into account employing the Monte-Carlo approach. Particle-decay widths were calculated within the Hauser–Feshbach theory.

We emphasize that the theoretical framework used in this work does not account for microscopic shell effects, neither in the calculation of the potential energy, nor for the level density. All ingredients are restricted to macroscopic concepts. That implies that the model is not suited at low E^* (below about 30 MeV). It has shown impressively powerful in explaining a large variety of observables over a wide range in compound nucleus mass, excitation energy and angular momentum for fission at moderate to high excitation energy (see Refs. [10,4] and references therein). Its recent development including the K degree of freedom has shown crucial to achieve a consistent description over a wide range of systems.

3. Results

The outcome of the 4D dynamical calculation for the reactions ^{12}C + ^{194,198}Pt → ^{206,210}Po is summarized in Figs. 1 and 2 for ^{206}Po and ^{210}Po, respectively, and compared to available experimental data. Panels (a), (b), (c) and (d) display, respectively, the neutron pre-scission multiplicities M_{pre}, neutron post-scission multiplicities M_{post}, fission cross sections σ_{fiss} and fission-fragment anisotropies A_{FF}, as a function of compound-nucleus excitation energy. Theoretical statistical error bars do not exceed 2% in all cases. Before coming to comparison with experiment, we note that, due to the somehow uncertain E^* and L values at which shell effects have completely vanished, the purely macroscopic model used here may approach the lower-limit of its applicability range for the lowest beam energy (equivalently, E^*) measured in Ref. [7].

Having in mind that there is basically one free parameter in the model, i.e. γ_K (with the additional possibility to use different prescriptions for the level density and the friction tensor), the
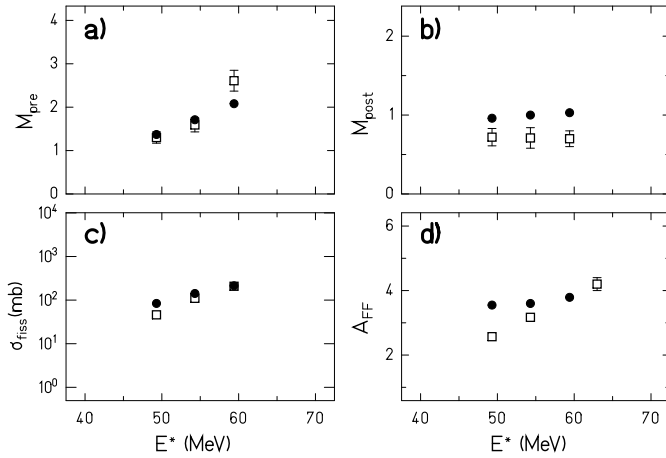
Fig. 1. Neutron pre-scission multiplicity M_{pre} (a), neutron post-scission multiplicity M_{post} (b), fission cross section σ_{fiss} (c) and fission-fragment anisotropy A_{FF} (d), as a function of compound-nucleus excitation energy for $^{12}C + ^{194}Pt \rightarrow ^{206}Po$. Model calculations (full circles) are compared to experimental data (open symbols) from Refs. [7,17].

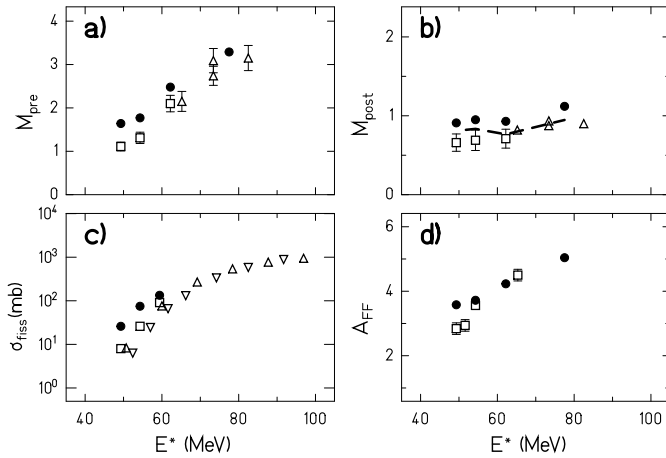**Fig. 2.** Similar to Fig. 1 for $^{12}C + ^{198}Pt \rightarrow ^{210}Po$. Experimental data are from Refs. [7, 17–19] (different symbols refer to different experiments). In (b) the dashed line corresponds to a variant of the calculation using a constant level-density parameter $a_n = A/12$.

agreement between calculation and experiment is very satisfactory overall. No attempt was made to adjust parameters beforehand: γ_K is set to the prescribed value of 0.077 $(MeV\,zs)^{-1/2}$ in the mass region of interest [4], the level density follows Ignatyuk's prescription and the chaos-weighted formula is used for friction in the shape coordinates. The overestimation of M_{pre} and σ_{fiss} for ^{210}Po is attributed, for the lowest E^*'s, to the remaining ground-state shell effects around neutron number $N = 126$, as well as to the possible shell-structure dependence of dissipation [20]. A better description of the experimental M_{post} values for both systems can be achieved by adjusting the level-density parameter. This is illustrated in Fig. 2(b) for ^{210}Po as an example. The description of the anisotropies can be improved with adjustment of the deformation-dependence of friction and the γ_K strength. Such a tuning is not the purpose of this letter.

Within the stochastic multi-dimensional approach of the Langevin type, a wide variety of observables can be calculated. In particular, with the code of Refs. [10,4], in addition to the observables shown in Figs. 1 and 2, fission-fragment mass and kinetic energy, light-charged particle multiplicities, angular distributions and energies, γ-ray spectra, can be investigated. As the available
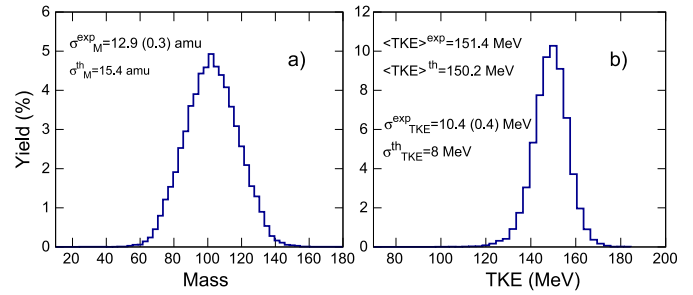

Fig. 3. Fission-fragment mass (a) and *TKE* (b) distribution for $^{12}C + ^{198}Pt \rightarrow ^{210}Po$ ($E^* = 77$ MeV). Model calculations are displayed only. The numerical values of most relevant moments of the distributions are indicated in insert and compared to experiment [21].

experimental data set is limited for the systems studied here, we can consider in Fig. 3 only the calculated fission-fragment mass (a) and total kinetic energy (b) distribution for ^{210}Po at $E^* = 77$ MeV, and compare in insert the extracted relevant moments with the measured values [21]. The calculated width σ_M of the mass distribution and mean total kinetic energy *TKE* are in reasonable agreement with experiment, while the width σ_{TKE} is underestimated. This deficiency of the model is well-known, and was attributed to the limited dimensionality of the shape parameterization [22], what can be cured in future developments.

4. Discussion

The results of the 4D dynamical modeling demonstrate that a macroscopic model, neglecting microscopic shell effects, and in particular shell corrections at the saddle point, is able to describe the experimental data available for fission of ^{206}Po and ^{210}Po above $E^* \approx 50$ MeV. The discrepancy that is observed for the neutron-magic ^{210}Po compound nucleus is attributed to the persistence of part of ground-state shell effects. The special behavior of ^{210}Po was already inferred in Ref. [7] from the experimental data, which do not exhibit the expected increase of M_{pre} relative to ^{206}Po based on mere N/Z considerations.

At variance with the dynamical calculation, Golda et al. [7] required to introduce in their statistical-model calculations a sizeable shell correction energy (several MeV) at the saddle point, for both ^{206}Po and ^{210}Po, in order to reproduce the experimental M_{pre}. This result may have an important impact, since previous studies of topographical properties of fission barriers suggest the saddle-point shell correction energy to be small, although not inexistent in this mass region [9,8]. The demonstration of the present work and the ambiguities in statistical-model analyses [12,23] balance the robustness of the conclusion of Ref. [7].

For the purpose of developing a framework devoted to predictions for synthesis of very heavy elements, Siwek-Wilczynska et al. [23] performed an analysis similar to Ref. [7], considering the shell correction energy at saddle as a free parameter. Using a compilation of experimental B_f values for nuclei above Pb, they found that the shell correction at the fission barrier is usually close to zero, in agreement with Ref. [9,8]. In Ref. [17], Shrivastava et al. focused on fission-fragment anisotropies for $^{12}C + ^{194,198}Pt \rightarrow ^{206,210}Po$ over an energy range similar to the one investigated here. They found that only for ^{210}Po are shell corrections required to be invoked, presumably due to its closed neutron shell. They did not attempt to figure out where these structural effects should be applied. On the contrary, the analysis of σ_{fiss} by Mahata et al. [24] for the $^{12}C + ^{194,198}Pt$ data set concluded to sizeable saddle-point shell effects, but of different magnitude than those extracted in Ref. [7]. That shows the non-univocal character of the statistical-model parameter set (see also Fig. 5 of Ref. [7]). For the nearby

^{19}F + $^{194-198}$Pt → $^{213-217}$Fr reactions, the statistical-model calculations by Singh et al. [25] introduced an overall scaling of the finite-range rotating Liquid-Drop fission barrier to explain measurements of evaporation-residue excitation functions. Various interpretations of similar data, involving also different parameterizations of the level-density parameter and/or the time-dependence of the fission-decay width, exist (see *e.g.* discussion in Refs. [26,27]).

As soon as in the seventies, Moretto et al. [28,29] and Ignatyuk et al. [14] performed a meticulous analysis of fission cross sections for several tens of proton- and α-particle-induced reactions, leading to compound nuclei with $A \approx 175$–215 and covering a wide excitation-energy range $E^* \approx 20$–120 MeV. These comprehensive studies showed that a consistent description of experimental cross sections can be obtained with the statistical model provided that the ground-state shell effects are adequately accounted for in the fission barrier. No indication for the need of including shell effects at the saddle point was found. The work and systematics of Refs. [28,29,14] are particularly relevant in the present discussion. Apart from the wide variety of the data set, they involve excitation energies down to very close to the fission barrier. Such E^* are best suited to extract more reliable, less-model dependent, estimates of the barrier height. In addition, as compared to heavy-ion induced reactions, the angular momentum is small with light-particle projectiles. That further limits complex interplay effects.

As already mentioned, the dynamical approach used in the present work is, of course, not free of assumptions. Different variants of its implementation exist, and inconsistent conclusions have been drawn depending on the specific model used [30]. However, the demonstration of Section 3 is sufficient to consider with caution the definitive statement on substantial shell effects at saddle in the $A \approx 200$ and $E^* \approx 30$–60 MeV critical region.

Although the statistical model has limitations [31,32], it shall not be concluded from this work that the difference between the results of Ref. [7] and the present ones is caused by dynamical effects. Indeed, the two sets of calculation differ certainly already in some statistical-model parameters (level densities, particle-decay widths, etc.). In this sense, our aim is all but to claim that we evidenced a manifestation of the *sole* dynamics. There is certainly much more behind all this, and the discussion is out of scope of the present letter. However, the demonstration of this work clearly suggests that dynamics plays a non-negligible role for moderately fissile systems at excitation energies close or slightly above the barrier. Fluctuations may be crucial in determining the fate of the system.

5. Summary and conclusions

Experimental data on fission observables are analyzed within an advanced dynamical approach based on the solution of a set of 4D Langevin equations. The model which neglects microscopic shell effects describes reasonably well pre- and post-scission neutron multiplicities, fission cross sections, fission-fragment anisotropies, mass and energy distributions, for the heavy-ion collisions ^{12}C + ^{194,198}Pt → ^{206,210}Po at above barrier energies. A small discrepancy observed for ^{210}Po is attributed to the persistence of shell effects from the spherical ground state. The conclusion of the dynamical results is at variance with recent statistical-model analysis [7,24] which concluded to the necessity of introducing sizeable shell effects at the fission saddle point. The good achievement by the purely macroscopic dynamical model presented in this work, as well as the discussion about alternative statistical-model-based investigations using various procedures and parameters, demonstrate that it is uncertain to claim that strong saddle-point shell effects have been firmly evidenced.

The present demonstration is not meant to stand for an evidence of purely dynamical effects. It nonetheless points to the need of dedicated studies with inclusion of both static shell effects and the influence of the dynamics, and this, in particular in the $A \approx 200$ region at moderate excitation energy. To improve the robustness of the conclusion, suited experimental observables have to be made available in parallel. These involve cross-bombardments with light and heavy probes, leading to similar compound systems populated in various E^* and L conditions, as well as the measurement of as various as possible observables *and* their correlation.

The present letter finally makes a point on the limited reliability of firmly extracting fundamental nuclear properties from customary analyses. Although this conclusion may be considered well-known and trivial, at the present, one can find numerous inconsistent conclusions. Poorly-constrained model-prescriptions and parameters can have important impact on the interpretation of the underlying physics.

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