Letter

Experimental evidence of the effect of nuclear shells on fission dissipation and time

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Nuclear fission is still one of the most complex physical processes we can observe in nature due to the interplay of macroscopic and microscopic nuclear properties that decide the result. An example of this coupling is the presence of nuclear dissipation as an important ingredient that contributes to drive the dynamics and has a clear impact on the time of the process. However, different theoretical interpretations, and scarce experimental data make it poorly understood. In this Letter, we present the first experimental determination of the dissipation energy in fission as a function of the fragment split, for three different fissioning systems. The amount of dissipation was obtained through the measurement of the relative production of fragments with even and odd atomic numbers with respect to different initial fission energies. The results reveal a clear effect of particular nuclear shells on the dissipation and fission dynamics. In addition, the relative production of fragments with even and odd atomic numbers appears as a potential contributor to the long-standing problem of the time scale in fission.

Introduction. More than 80 years after its discovery [1,2], nuclear fission is still one of the most challenging reactions we can study in the laboratory. While some of its basic properties can be derived by describing the fissioning nucleus in a macroscopic fashion [3], it was soon realized that nuclear structure and single-particle excitations may also have an impact on the process [4,5]. Since then, the interplay between collective and intrinsic degrees of freedom is yet to be fully understood and accounted for in current theoretical fission models [6].

Expected effects of such an interplay are dissipative processes, where part of the energy stored in collective excitations is transferred to single-particle excitations. Nuclear dissipation is a pivotal ingredient that can be found in different phenomena, such as nuclear fusion and the production of superheavy elements [7,8], deep inelastic collisions [9,10], or giant resonances [11]. In all of them, nuclear dissipation

contributes to define the time scale and outcome. The case of nuclear fission should not be different: as the system evolves from an equilibrated system at the barrier [12] to the scission point, intrinsic excitations draw energy from collective deformation, slowing down the process (see early discussions in [13,14]). The actual role of dissipation and its link with the time scale in fission are still to be fully understood; different prescriptions can be found in fission models [6,15–17], while interpretations from experimental data are usually model-dependent [18,19].

An additional consequence of the interplay between collective and intrinsic excitations would be a certain correlation between dissipation and nuclear structure. While the role of closed shells in the fissioning system was studied in the past [20–22], few descriptions explore this dependence with the structure of the fragments [23], which was never observed in experimental data. However, indications of low dissipative fission were assigned in very asymmetric fragment splits [24] and in the survival of nuclear clusters in cold fission [7].

Experimentally, the amount of intrinsic energy produced in fission events can be related with the global even-odd effect δ, which is defined as the relative difference between the total yield of even-Z fragments and that of odd-Z fragments, being Z the number of protons. An example of this relation can be found in fission reactions of even-Z systems with a low initial excitation energy E^*, of the order of the fission barrier height. On their way from the ground-state deformation to the fission

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barrier, the systems transform the initial energy into further deformation in order to overcome the barrier, and the single-particle excitations in the transition states at the fission barrier represent the starting conditions for the dynamical evolution towards scission. In these low-energy cases, only completely paired transition states, with no single-particle excitations, are populated [25]. However, in such reactions, odd-Z fragments were measured [26–29]. This means that, at some point before scission, proton pairs are broken and the single protons are distributed between the prefragments,[1] resulting in odd-Z combinations (an equivalent process happens with neutrons, although neutron evaporation from fragments prevents its direct measurement). The appearance of these single protons in even-Z systems that have started their descent from saddle to scission in completely paired configurations is an evidence that part of the potential-energy release is dissipated into intrinsic excitations [32–34].

The amplitude of δ was found to decrease exponentially with E^{Bf}, the energy above the barrier Bf: $E^{Bf} = E^* - Bf$ [28,29]. Since E^{Bf} and the dissipated energy, E^{dis}, are intrinsic excitations, it is straightforward to assume that δ would have the same dependence on both. A combinatorial analysis led to a direct link between E^{dis} and δ for low-energy fission [35,36]:

$$E^{\mathrm{dis}} + E^{Bf} \approx -4\ln(\delta). \tag{1}$$

Similarly, it is also possible to compute the *local* even-odd effect, δ_Z, which is the magnitude of the even-odd effect as a function of the fragment Z [37–39]. The study of δ_Z revealed a systematic increase of the effect with the asymmetry of the split, explained with the influence of the relative level densities of the fragments [40,41]. In a recent model [34], δ_Z is obtained from the statistical breaking of pairs and their distribution between the prefragments, according to the energy-sorting mechanism identified in [42]. However, this model does not describe explicitly the generation of the energy dissipated, assuming it as constant fraction of the potential energy gained in the process, following the prescription of [43].

Despite the close relation between δ_Z and the intrinsic energy, no data set on the evolution of δ_Z with E^* was available, so far. We present here the first set of such measurements. From their analysis, we can estimate E^{dis} as a function of the fragment Z, and reveal the role of spherical and deformed shells in the generation of dissipation. These results also identify different stages of energy sharing through nucleon exchange, and hint at a potential observable to compare the fission time of fragment splits.

Local even-odd effect and fission energy. We have measured the local even-odd effect δ_Z in the fragment distributions of three fissioning systems as a function of the fission excitation energy E^*. The data were obtained from transfer-induced fission reactions between a ^{238}U beam and a ^{12}C target, measured in inverse kinematics with the large-acceptance magnetic spectrometer VAMOS++ at GANIL

(France) [44,45]. The main experimental details can be found in [46–48]. In the present work, the computation of the local even-odd effect is based on the widely used prescription from [37], in which δ_Z corresponds to the fraction that the elemental yields of groups of four consecutive fragment Z deviate from a Gaussian behavior (see Supplemental Material [49] for details).

The data presented here include fission from ^{238}U, ^{239}Np, and ^{240}Pu, each with its corresponding measured distribution of E^* [46] detailed in the Supplemental Material [49]. In addition, we have included data of thermal-neutron-induced fission of ^{240}Pu and ^{239}Np from [50,51]. Figure 1 shows the evolution of δ_Z as a function of the fragment Z and the average E^{Bf}.

The expected systematic increase of δ_Z with the asymmetry of the split is observed in Fig. 1 in ^{238}U and ^{240}Pu. As discussed in the previous section, unpaired protons are drawn to higher level densities, thus their probability of ending up in heavier fragments increases with the ratio of the fragment densities, closely related to the asymmetry of the split. This mechanism is clearly seen in the evolution of δ_Z in the odd-Z system ^{239}Np: the heavy fragment is more likely to receive the unpaired proton, resulting in negative δ_Z.

The measured δ_Z also shows a clear maximum around the spherical closed shell $Z = 50$ in the three systems. However, the fact that the maximum in ^{239}Np corresponds to positive δ_Z, instead of negative values, reveals a mechanism other than the effect of level density. While it is tempting to explain it with an inversion of the energy and proton flux due to the reduced level density at $Z = 50$, it is not supported by experimental data, which show that the energy flux towards heavy fragments is maintained along the fragment distribution [53]. The presence of this maximum suggests that heavy prefragments with atomic number $Z = 50$ are formed preferably without unpaired protons. The data from ^{239}Np show then two ways in which intrinsic energy contributes to δ_Z in two different time frames: with the distribution of unpaired protons at the very formation of prefragments close to the barrier and with the transfer of unpaired protons between prefragments right until scission.

Figure 1 also hints at the relation between the intrinsic energy and the magnitude of δ_Z: as the initial E^{Bf} increases, $|\delta_Z|$ decreases. The total intrinsic energy available is the sum of the initial energy above the barrier and the dissipation generated in the process, $E^{\mathrm{int}} = E^{Bf} + E^{\mathrm{dis}}$. In order to describe the dependence of δ_Z with E^{int}, we use a phenomenological generalization of the prescription discussed in Eq. (1):

$$E^{\mathrm{dis}}(Z) + [E^{Bf} - \Delta] = G(Z)\ln(|\delta_Z|) \quad E^{Bf} > \Delta,$$
$$E^{\mathrm{dis}}(Z) = G(Z)\ln(|\delta_Z|) \quad E^{Bf} \leqslant \Delta. \tag{2}$$

Here, the minimum energy needed to break proton pairs,[2] Δ, is subtracted from E^{Bf} (see Supplemental Material [49] for

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[1]Prefragments are still joined through the neck but they already display basic individual properties [30,31].

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[2]Similar to the Bardeen-Cooper-Schrieffer formalism, Δ is twice the energy to produce a quasiparticle, i.e., a free proton (see Supplemental Material [49] for details).
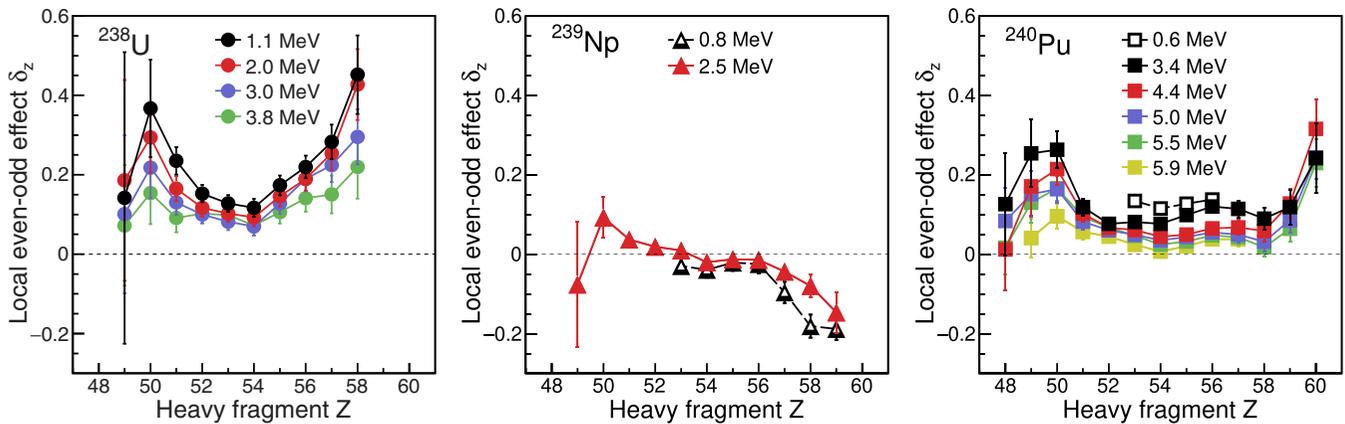
FIG. 1. Local even-odd effect δ_Z as a function of the heavy-fragment Z for ^{238}U (left), ^{239}Np (middle) with data from [51] (dashed line with empty symbols), and ^{240}Pu (right) with data from [50] (dashed line with empty symbols). (We only include values of δ_Z calculated with complete isotopic distributions [52].) Each line and color correspond to the $\langle E^{Bf} \rangle$ listed in the figures.

details). Below this value, pairs are only broken through dissipation. This formula has two free parameters that depend on the fragment split: $E^{\mathrm{dis}}(Z)$ and $G(Z)$. The former corresponds to the intrinsic energy for initial E^{Bf} below Δ. Such intrinsic energy is necessarily released during the process and thus, identified as dissipation energy. The parameter G corresponds to the slope of the correlation between E^{Bf} and $\ln(|\delta_Z|)$. In other words, G can be interpreted as the sensitivity of the δ_Z to changes in excitation energy. As a reference, $G = -4$ MeV in Eq. (1) for the case of the global even-odd effect δ [36].

In Eq. (2), as in previous works, it is assumed that E^{dis} has a negligible dependance on E^{Bf}, that Δ is constant[3] with respect to E^{int}, and Bf is the same for all fragment splits. In order to compare it directly with experimental data, Eq. (2) is folded with the measured probability of E^* (see Supplemental Material [49] for details). The parameters E^{dis} and G are obtained by fitting the resulting functions to the measured data. Figure 2 shows the evolution of $|\delta_Z|$ as a function of E^{Bf} and the corresponding fit for each heavy-fragment Z. In the case of ^{239}Np and ^{240}Pu, the data from thermal-neutron-induced fission shown in Fig. 1 [50,51] are included. Left panel of Fig. 3 displays $E^{\mathrm{dis}}(Z)$ and $G(Z)$ for the three systems explored.

Dissipation energy and nuclear shells. Octupole-deformed shells $Z = 52$ and 56 were recently proposed as responsible for asymmetric fission in actinides [54,55], thus it is not completely unexpected that dynamic features and dissipation may have a dependence on these shells. Indeed, the left top panel of Fig. 3 shows that the deduced E^{dis} follows a similar pattern in ^{238}U and ^{240}Pu that seems to react to the shell structure of the fragments. The most prominent feature is a peak that reaches beyond 10 MeV close to the deformed shell $Z = 52$. This maximum suggests that, in the path to form this shell and the corresponding light fragment, the system goes through a relatively large number of level-crossings where fission is slowed

down [16,56], increasing the amount of one-body dissipation and breaking additional pairs through the Landau-Zener effect [5,32]. Around the octupole-deformed shell $Z = 56$, E^{dis} falls to ≈ 5 MeV in the three systems; an indication that they seem to endure less one-body dissipation and/or level crossing than in the case of $Z \approx 52$.

Around the spherical closed shell $Z = 50$, E^{dis} drops to a minimum of ≈ 2 MeV, suggesting that most of the excitation energy gained in the process is used in deforming the light fragment, while the $Z = 50$ heavy fragment is kept mostly spherical and with low intrinsic energy. This is consistent with the maximum of δ_Z seen in Fig. 1 and with the low neutron evaporation measured around $Z \approx 50$ (see [57] for a review). The low E^{dis} also suggests that the system splits in a shorter time, which is consistent with the relation between the production of $Z = 50$ fragments and the development of a short neck in the scission configuration of ^{240}Pu and ^{239}U reported in [55,58].

For more asymmetric splits, ^{240}Pu and ^{239}Np display a sudden increase of E^{dis}, while ^{238}U data seem to stop short of a similar increase. This behavior is addressed later.

As it was mentioned, E^{dis} was estimated in previous works as a constant fraction of the potential energy gained up to scission [25,34,43,59], which shows a smooth evolution with the fragment split, with no dependence on specific shells [55,60,61]. With a typical fraction of 0.35, this approximation gives an average E^{dis} around 7 MeV for ^{238}U and 9 MeV for ^{240}Pu, slightly larger than our results, which average to 5.4 and 6.4 MeV, respectively. Another recent calculation, based on strongly damped shape evolution on potential-energy surfaces, also gives a larger value of 11.3 MeV for ^{240}Pu [62].

The bottom left panel of Fig. 3 also shows the evolution of G as a function of the heavy-fragment Z. The average values are $G \approx -3$ and -3.4 MeV for ^{238}U and ^{240}Pu, respectively; not dissimilar to the $G = -4$ MeV estimated in [36]. Around the octupole-deformed $Z = 52$, we observe a clear deviation, as in the case of E^{dis}, that makes δ_Z less sensitive to changes in E^{Bf}. For more asymmetric splits, beyond $Z = 58$ in ^{240}Pu, the magnitude of G increases nearly a factor 10, rendering δ_Z almost constant and thus insensitive to changes in E^{Bf}. The

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[3]Following the prescription of [25], the change in Δ along the range of energy in this work is less than 10%, and the effect on the fitted parameters falls to $\approx 1\%$.
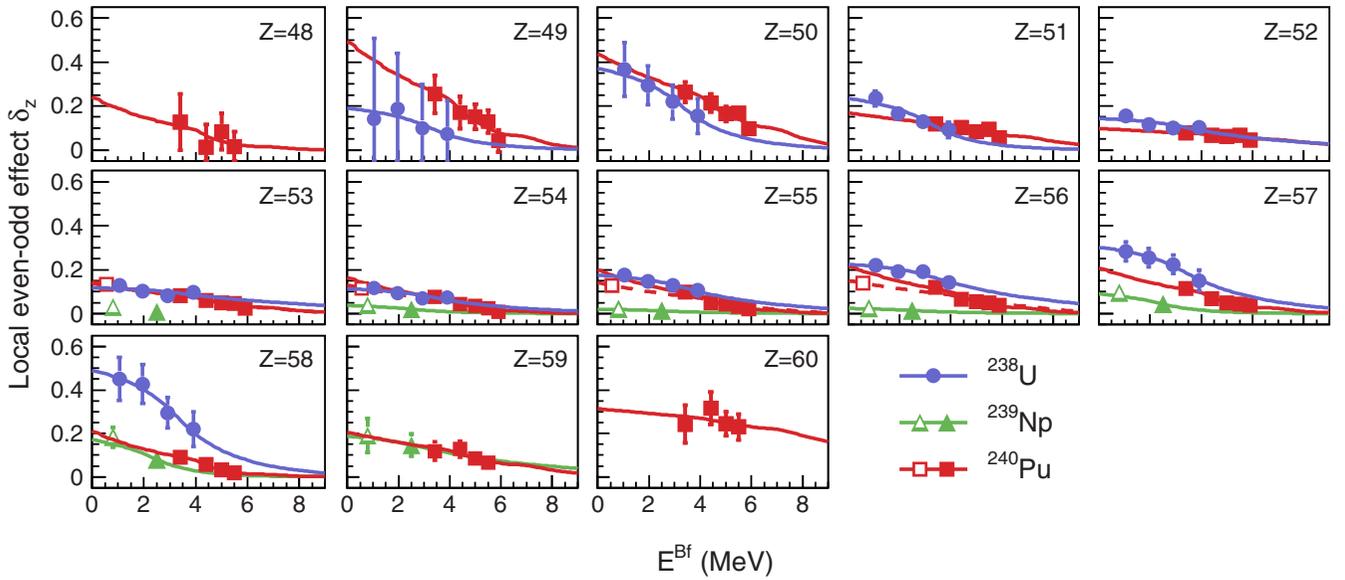
FIG. 2. Evolution of δ_Z as a function of the average $E^{\rm Bf}$ for each split in ^{238}U (blue dots), ^{239}Np (green triangles), and ^{240}Pu (red squares). In the case of ^{239}Np, $-\delta_Z$ is plotted. Solid symbols are measured data and lines are fits to Eq. (2) folded with the measured E^* detailed in the Supplemental Material [49]. Empty symbols are data from [50,51]. Red dashed lines show fits when data from [50] are included.

data on ^{238}U and ^{239}Np stop right before, but they follow a similar trend.

Threshold asymmetry in the local even-odd effect. As it was discussed, the energy dissipated when breaking proton pairs is reflected in the measured δ_Z in two ways: with the redistribution of these protons during the formation of the prefragments, and with their transfer between prefragments along the process. While the former is mainly ruled by stochastic breaking and the structure of the prefragments, the latter is dominated by the level densities and the temperature difference between prefragments, which also depend on the asymmetry of the split [34,63]. In addition, the transfer of protons needs a certain time, $t_{\rm tr}$, to be completed. In average, this time increases

with the amount of intrinsic energy stored in the fragment and decreases with the mass asymmetry [63].

The exchange of protons through the neck is possible until the Coulomb repulsion between the two nascent fragments approaching scission hinders it. If, for a sufficiently large asymmetry, the time until this point, $t_{\rm sc}$, is much longer than $t_{\rm tr}$, having more initial $E^{\rm int}$ would not affect δ_Z: the few unpaired protons that remain in the light fragment at the final stage of the energy sorting would have time enough to be all transferred [34,63]. The result is that, for fragment asymmetries beyond a threshold point, the measured δ_Z loses its sensitivity on $E^{\rm int}$, that is, the amplitude of G increases very fast. The rapid evolution of G for large asymmetry in ^{240}Pu,
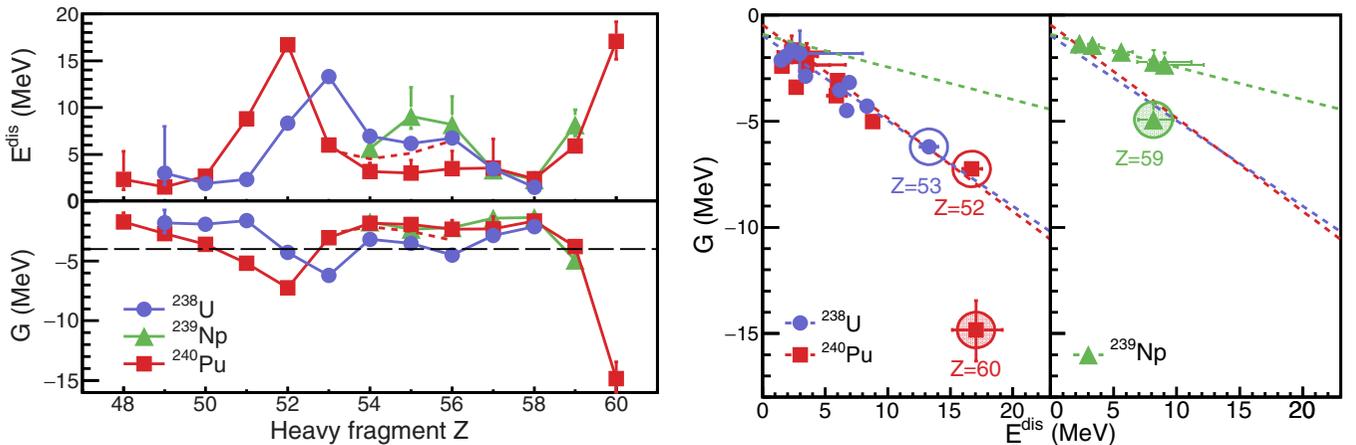

FIG. 3. Left: Deduced $E^{\rm dis}$ (top) and G (bottom) as a function of the heavy-fragment Z. The red dashed lines show the results of ^{240}Pu when including data from [50]. In the bottom panel, the horizontal dashed line shows $G = -4$ MeV (see text). Right: Correlation between G and $E^{\rm dis}$ for ^{238}U and ^{240}Pu (left), and ^{239}Np (right). The lines are fits to guide the eye. Circled points correspond to splits where the influence of the $Z = 52$ shell was identified (empty enclosing), and to threshold splits (shaded enclosing). In all panels, blue dots correspond to ^{238}U, green triangles to ^{239}Np, and red squares to ^{240}Pu.

shown in the left bottom panel of Fig. 3, suggests that the threshold point for this system is around $Z = 60$. Assuming the same t_{sc} [63] would result in similar threshold asymmetries for ^{238}U and ^{239}Np, which is consistent with the parallel evolution of G in the three systems for very asymmetric splits. Since $t_{tr} \approx t_{sc}$ at the threshold asymmetry, the estimation of t_{tr} from microscopic models would inform directly on the magnitude of t_{sc}, adding another piece to the long-standing problem on the time scale in fission.

Since there is a certain correlation between dissipation and the time to scission, for high E^{dis}, it is more probable to transfer all protons from light to heavy prefragments, which, in turn, decreases the sensitivity of δ_Z or, equivalently, increases the amplitude of G. This must result in a correlation between E^{dis} and G. Right panels of Fig. 3 show that such a correlation appears for the three systems. ^{238}U and ^{240}Pu seem to have the same trend while ^{239}Np, probably due to its odd-Z nature, follows a different but also well-defined correlation with a slope that is approximately half of the one corresponding to ^{238}U and ^{240}Pu. The measured data show two clear deviations from these correlations in ^{239}Np and ^{240}Pu at the splits corresponding to $Z = 59$ and $Z = 60$, respectively, already discussed as threshold asymmetries. These deviations support the interpretation that, beyond these asymmetries, the few unpaired protons stored in the light fragment have time enough to be transferred to the heavy one, degrading the sensitivity of δ_Z to E^{int} and losing the underlying dependence on time. For splits with a heavy-fragment Z beyond these threshold points, the dissipation deduced with Eq. (2) is probably no longer reliable. The observation of these threshold points also indicates that the evolution of δ_Z as a function of E^{int} in highly asymmetric splits is a potential observable to compare the fission time of different systems.

In summary, high-precision data on fission fragment yields from different fissioning systems at different excitation energies have been used to extract the evolution of the dissipation energy as a function of the fragment split. The analysis of the relative production of even- and odd-Z fragments shows a clear influence of nuclear structure in the dissipation process and fission dynamics: fission is faster when producing fragments with spherical shell $Z = 50$, but slower and more dissipative when fragments are shaped around octupole-deformed shell $Z = 52$.

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