Letter

Odd-even stagger in dissipative fission of excited nuclear systems

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First-chance fission probabilities (P_{f1}) of various Am, Pu, and Np isotopes are found to display a significant neutron odd-even stagger (OES), and an obvious excitation energy dependence of the OES is presented. Moreover, a prominent dissipation effect on the OES is revealed, and the differences in P_{f1} of two neighboring fissioning isotopes ^{231}Am and ^{232}Am, ^{230}Pu and ^{231}Pu, and ^{225}Np and ^{226}Np, ΔP_{f1} (which denotes the amplitude of the OES), are shown to be a quick decreasing function of the dissipation strength (β). This means that the amplitude of the OES is a sensitive experimental indicator of nuclear dissipation properties, and it is proposed here as a new experimental signature to constrain β. Additionally, we find that P_{f1} of ^{231,233}Am, ^{230,232}Pu, and ^{225,227}Np, whose neutron numbers are even, respectively exhibit an apparently greater sensitivity to β than that of their neighboring isotopes ^{232}Am, ^{231}Pu, and ^{226}Np, whose neutron numbers are odd. The neutron odd-even effect is shown to be responsible for the marked enhancement in the sensitivity of the former, clearly indicating its strong influence on using P_{f1} to probe β. These results suggest that on the experimental side, to more stringently constrain nuclear dissipation by measuring first-chance fission probability, it is optimal to produce those heavy fissioning systems with even neutron numbers.

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Introduction. How to better search for and utilize the prominent role that nuclear structure plays in nucleus-nucleus collision dynamics in entrance channels and in subsequent decay processes has been an important subject in the field of nuclear physics. In this aspect, a typical example is the experimental production of superheavy nuclei, where targets ^{209}Bi (having a neutron shell closure) [1] and projectiles ^{48}Ca (which is a doubly magic nucleus) [2] have been used in cold and hot fusion reactions, respectively. In addition to the shell structure effect, odd-even stagger (OES) in neutron and proton numbers, as a distinct characteristic, has been experimentally confirmed to affect low-energy fission-fragment yields [3–6], the magnitude of fragment cross sections stemming from intermediate-energy reactions [7], projectile fragmentation [8] and spallation reactions [9], and yields of neutron-deficient nuclei produced in heavy-ion collisions [10]. Recently, nuclear charge radii [11] and α-decay energies [12] also display OES while changing neutron or proton number.

The OES is usually attributed to the existence of neutron and/or proton pairing correlations [13,14]. Fragment cross sections [6–10] generated in nuclear reactions go through a multistep evaporation process. As a result, pairing terms in the binding energies of parent and many daughter nuclei generated along the entire decay chain could affect the cross sections complicating the contribution of pairing energies to end products. It is thus ideal to employ those experimental signals that occur at an early stage of a decay chain to exploit OES. Using cross bombardment experiments, first-chance fission probabilities (P_{f1}) [15,16] were extracted through measured prescission neutron multiplicities in symmetric fission of two neighboring fissioning isotopes under matched experimental conditions. This observable can describe first-chance fission properties and does not involve a multiple particle emission. It could thus provide an optimal experimental condition of studying the OES phenomenon.

A number of experiments on prescission emission in symmetric fission processes [17,18] and fission/evaporation residue cross sections [19,20] have revealed their deviations from predictions by standard statistical models (SMs), as more energies are deposited into a compound nucleus (CN). This discrepancy has been established to be caused by dissipation effects that are not accounted for in SMs [21–24]. To date, many works have surveyed dissipation properties, but the presaddle dissipation strength (β) is still quite uncertain and currently under vigorous debate [25]. A precise β is important not only for explaining fission data of highly excited nuclei, but also for accurately calculating survival probabilities of superheavy nuclei [16,26]. Therefore, making using of different types of observables is necessary for stringently constraining β [27]. The magnitude of P_{f1}, dictated by the competition between fission and evaporation in the first step of a de-excitation process, was shown to be a sensitive signature of dissipation effects in fission [15,28].

In these contexts, the aim of the present work is twofold. First, we search for OES through P_{f1}. The second aim is to explore the nuclear structure effect (i.e., OES) in probing dissipative fission properties. For these aims, besides SMs, the dynamical Langevin model is considered. The stochastic

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approach has been demonstrated to be a powerful tool to address various types of fission data, such as cross sections of fission [29] and evaporation residues from light ^{200}Pb to heavy ^{224}Th [30] and prescission particle multiplicities of compound systems ranging from ^{179}Re up to ^{256}Fm [31]. It was recently applied to calculate higher-chance survival probabilities of heavy systems [32]. We here use the same model developed in Refs. [28–32] to calculate P_{f1} of various heavy Am, Pu, and Np isotopes in order to display OES through the proposed observable and moreover, investigate the role of the OES in pinning down dissipation characteristics in fission of excited nuclei.

Theoretical model. In order to describe the driving force of a hot nuclear system, one should use a thermodynamic potential [22,23] rather than a bare potential. While the derivative of free energy with respect to the deformation coordinate at fixed temperature was applied to calculate the driving force [33], the temperature is not constant during evolution. So we here use entropy [34–36], which is more suited to describe the driving force in Langevin equations.

We employ the following Langevin equation to perform the fully dynamical trajectory calculations for symmetric fission:

$$\frac{dp}{dt} = K - \beta p + g\Gamma(t),$$
$$\frac{dq}{dt} = \frac{p}{m}. \tag{1}$$

Here, q is the dimensionless fission coordinate and is defined as half the distance between the center of mass of the future fission fragments divided by the radius of the CN, and p is its conjugate momentum. Like previous publications [17,19,22,27,35,37–41], the reduced dissipation coefficient (also called the dissipation strength) $\beta = \gamma/m$ denotes the ratio of the friction coefficient γ to the inertia parameter m. The m is calculated in the Werner-Wheeler approximation for the irrotational flow of an incompressible liquid [42]. $\Gamma(t)$ is random force satisfying $<\Gamma(t)> = 0$ and $<\Gamma(t)\Gamma(t')> = 2\delta(t-t')$. The strength of the random force is related to the dissipation strength through the fluctuation-dissipation theorem and is $g = \sqrt{m\beta T}$ with T being the temperature.

The driving force K of the Langevin equation is calculated in terms of derivatives of the entropy S at a constant excitation energy:

$$K(q) = T\frac{dS}{dq}. \tag{2}$$

The entropy $S(q)$ is calculated as

$$S = 2\sqrt{a(q)[E^* - V(q) - E_{\rm coll}]}, \tag{3}$$

where E^* denotes the total excitation energy of the fissioning system, and $E_{\rm coll}$ is the kinetic energy of the collective degree of freedom. Equation (3) is constructed from the Fermi-gas expression [34]. The deformation coordinate q is obtained by the method given in Refs. [22,43], where the Funny-Hills shape parameters [44] are used. The potential energy $V(q)$ includes q-dependent surface, Coulomb, and rotation energy terms, which are calculated with a finite-range liquid-drop model [45]. Shell corrections and pairing energies are calculated as prescribed by Möller *et al.* [46], and the level density parameter $a(q)$ and its dependence on deformation and microscopic corrections (i.e., shell-correction energy plus pairing energy) were calculated as described in Ref. [47]. As to the particle emission, a Monte Carlo simulation technique was used, and the emission width of a particle of kind ν ($= n, p, \alpha$) is calculated with Blan's parametrization [48] that was used in many studies [22,33,40,49,50]. After each emission act of a particle, the excitation energy, the potential energy, the entropy, and the temperature in the Langevin equation are recalculated and the dynamics is continued.

When the dynamic trajectory reaches the scission point, it is counted as a fission event. The scission is considered here to occur when the neck radius of the fissioning nucleus is equal to $0.3R_0$ (R_0 is the radius of the initial spherical CN) [23,51,52]. Our calculations allow for multiple emissions of light particles and higher-chance fission. So, the first, second,etc., chance fission probability can be calculated [22] by counting the number of corresponding fission events in which not a single presaddle particle is emitted, only a presaddle particle is emitted.

The first-chance fission probability (P_{f1}) of a compound system is calculated as

$$P_{f1} = \frac{N_{f1}}{N_{\rm tot}}. \tag{4}$$

Here, N_{f1} and $N_{\rm tot}$ denote first-chance fission event numbers and the total simulated trajectory numbers, respectively.

Similar to previous Langevin calculations [22,51,52], the initial conditions for the dynamical Eq. (1) are assumed to correspond to a spherical CN with an excitation energy E^* and the thermal equilibrium momentum distribution. For starting a Langevin trajectory an orbit angular momentum value is sampled from the fusion spin distribution [22]. The final results are weighted over all relevant angular momenta; that is, the spin distribution is used as the angular momentum weight function.

Results and discussions. For isotopes of heavy elements Am, Pu, and Np considered here, their neutron emission is far stronger than their charged particle emission. So, we focus on the competition between neutron evaporation channel and fission channel. We choose Am isotopes to explore the OES phenomenon through their P_{f1}. The calculated results are presented in Fig. 1(a).

We first discuss SM calculations without dissipation effects (denoted by red circles). The most prominent feature observed is that P_{f1} displays a quite clear neutron OES with a change in the neutron number (N) of Am isotopes. Specifically speaking, even-N Am isotopes have a greater P_{f1} than their odd-N neighbors, which is the consequence that these two types of Am nuclei have different neutron binding energies B_n.

Figure 1(b) plots B_n of Am isotopes predicted with the finite-range droplet model (FRDM) [46]. The model has been demonstrated to describe well experimental data on B_n of isotopes of heavy elements including element Am [53]. It is easily seen from this figure that both ^{231}Am and ^{233}Am have an obvious larger B_n than ^{232}Am. The same is found for other neighboring Am isotopes. Furthermore, the differences in neutron pairing energies of two neighboring Am isotopes

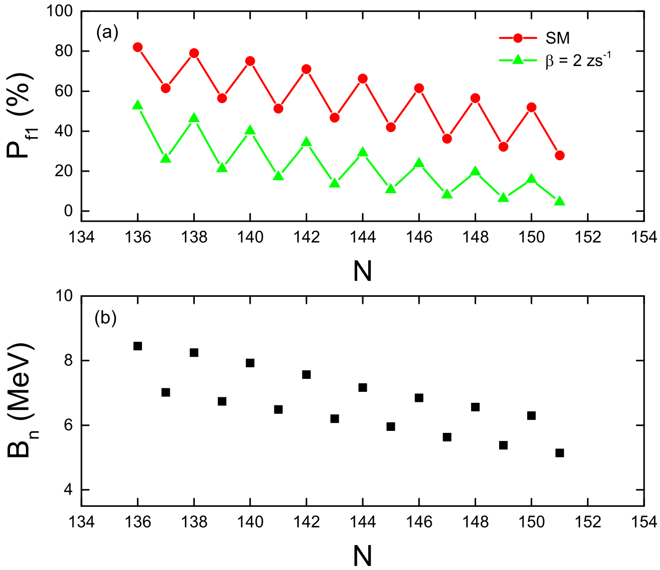

FIG. 1. (a) First-chance fission probability (P_{f1}) of Am isotopes as a function of their neutron numbers N calculated at excitation energy $E^* = 65$ MeV and critical angular momentum $\ell_c = 30\hbar$ for two cases: (i) standard statistical model calculations without dissipation effects and (ii) Langevin calculations with a presaddle dissipation strength of $\beta = 2$ zs^{-1}. Here, 1 zs = 10^{-21}s. (b) Neutron binding energy (B_n) of various Am isotopes predicted by FRDM [46].

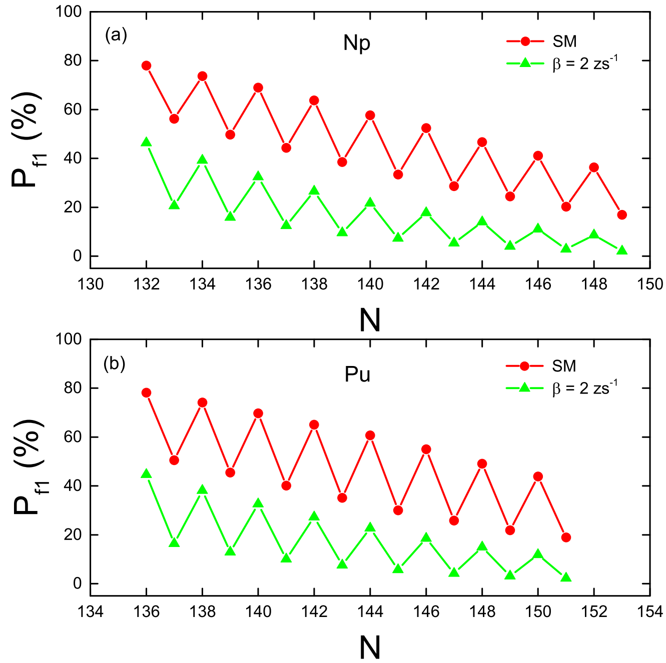

FIG. 2. Same as in Fig. 1(a), but for (a) Np isotopes and (b) Pu isotopes.

calculated by FRDM [46] are found to have a chief contribution to their different B_n.

A high B_n suppresses neutron evaporation, favoring fission. This leads to a larger P_{f1} of ^{231}Am (^{233}Am), ^{235}Am (^{237}Am), ^{239}Am (^{241}Am), and ^{243}Am (^{245}Am) compared to that of ^{232}Am, ^{236}Am, ^{240}Am, and ^{244}Am. Thus, the neutron odd-even effect in B_n significantly affects neutron emission and thereby, its competition with fission in the first-step decay stage of two neighboring Am isotopes. In other words, the neutron OES phenomenon plays an important role in determining the magnitude of P_{f1}. This conclusion is robust with respect to different types of models used, because both statistical models and dynamical Langevin approaches predict an analogous behavior of P_{f1} while changing N of Am isotopes, i.e., the emergence of neutron OES. Also, one can see in Fig. 1(a) that as dissipation is taken into account, the OES is shown to be more pronounced for neutron-poor systems, which is the result of two opposite factors influencing the competition between the neutron evaporation channel and the fission channel. On one hand, with decreasing N of a system, a larger B_n makes neutron emission less competitive with fission. On the other hand, dissipation hinders fission. The two aspects have an impact on the evolution of P_{f1} with N of Am isotopes and correspondingly, the OES.

A picture like Fig. 1(a) is observed for the case of light Np isotopes [Fig. 2(a)]. The same features of curves P_{f1} versus N as those in Fig. 1(a) are also seen for Pu isotopes (which have even proton numbers Z) [Fig. 2(b)], implying that the neutron OES displayed in P_{f1} does not depend on the odevity of Z of a heavy nucleus. One can thus conclude that neutron OES appearing in P_{f1} is a general phenomenon of heavy fissioning systems.

We note that OES reported previously was primary for end products of hot nuclei, for instance, yields of fragment cross sections [7–10], which are produced after multiple particle emissions, i.e., they are formed at the end of a deexcitation process. Different from that, a strong neutron OES displayed through P_{f1} appears at an initial stage of a decay process, where the excitation energy of the fissioning system is not very low. In this aspect, our results on OES as shown in Figs. 1(a) and 2 are complementary to that identified through fission-fragment charge and isotope yields, the magnitude of fragment cross sections coming from intermediate- and high-energy reactions, charge radii, α-decay energies, etc., which were found to show up at a lower energy, see, e.g., Refs. [3–12]. Similar conclusions were also reached by Agostino *et al.* [54], who showed that OES is present in the isotopic fragment distributions when the excitation energy is small. Our finding thus expands the observation of a marked OES phenomenon to a higher energy region through P_{f1} characterizing nuclear fission properties.

Much efforts were made to survey those factors that can affect OES. For example, different descriptions of the density of states at a low energy were shown to be able to influence OES displayed in the charge distribution of the emitted fragments in heavy-ion collisions [7,9]. Figure 1(a) indicates that because of fission hindrance caused by dissipation, neutron evaporation can compete more effectively with fission, leading to a drop in P_{f1}. Moreover, one can see that the differences in P_{f1} of two neighboring Am isotopes, which reflects the amplitude of the OES, vary obviously as calculations are conducted with SMs and with dynamical models, respectively. This comparison clearly demonstrates that dissipation

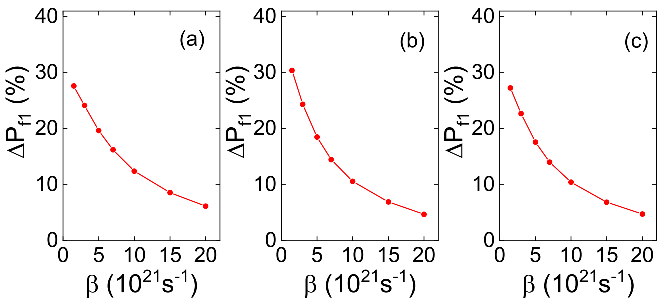

FIG. 3. Differences in first-chance fission probability (ΔP_{f1}) for (a) ^{231}Am and ^{232}Am, (b) ^{230}Pu and ^{231}Pu, and (c) ^{225}Np and ^{226}Np as a function of the presaddle dissipation strength β calculated at excitation energy $E^* = 65$ MeV and critical angular momentum for fusion $\ell_c = 30\hbar$.

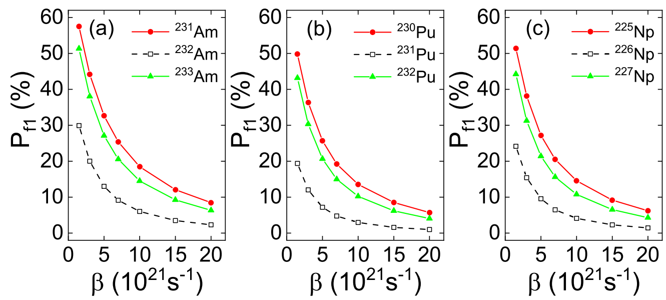

FIG. 4. First-chance fission probability (P_{f1}) as a function of the presaddle dissipation strength (β) for (a) ^{231}Am, ^{232}Am, and ^{233}Am, (b) ^{230}Pu, ^{231}Pu, and ^{232}Pu, and (c) ^{225}Np, ^{226}Np, and ^{227}Np calculated at $E^* = 65$ MeV and $\ell_c = 30\hbar$.

in the fission channel of an excited system is a new factor influencing OES. Meanwhile, it also implies that there could exist a closer correlation between the amplitude of the OES that is denoted here by ΔP_{f1} (which is the difference in P_{f1} of two neighboring fissioning isotopes) and the dissipation strength β.

In order to further explore the correlation, we calculate the evolution of ΔP_{f1} with β for two neighboring isotopes ^{231}Am and ^{232}Am, ^{230}Pu and ^{231}Pu, and ^{225}Np and ^{226}Np. The most distinct feature seen in Fig. 3 is that ΔP_{f1} decreases rapidly with increasing friction. It is mainly because as dissipation becomes stronger, the neutron emission probability is further enhanced, which results in a smaller first-chance fission probability. A low P_{f1} for two neighboring fissioning isotopes generally yields a small ΔP_{f1}; that is, the OES becomes weak at a large β. Also, ΔP_{f1} can be experimentally determined with methods proposed in Refs. [15,16]. Thus, the amplitude of the OES is a sensitive experimental indicator of dissipation effects in fission, suggesting that experimentally, the new observable can be used to accurately probe the dissipation strength.

Figure 1(a) shows that because of the OES, dissipation has an apparently different effect on the magnitude of P_{f1} even if for two neighboring fissioning isotopes. In the following, we investigate the role of the neutron odd-even effect in pinpointing β with P_{f1}. To this end, three neighboring Am isotopes, i.e., ^{231}Am, ^{232}Am, and ^{233}Am are chosen as representatives. The calculated results are depicted in Fig. 4(a). A decreasing P_{f1} with an increase in β is because the stronger the nuclear dissipation, the more a fission process is severely delayed. Apart from that, two typical features are also noticed in the figure. First, both ^{231}Am and ^{233}Am have a larger P_{f1} than ^{232}Am. We explain this below. In the first-step decay stage of a hot heavy nucleus, competition between neutron emission and fission dominates the magnitude of its P_{f1}. So neutron binding energies and fission barriers are crucial factors. These three neighboring Am isotopes have a similar mass number, so their fission barrier heights are also similar [45]. Therefore, it is a difference in B_n between ^{231}Am (^{233}Am) and ^{232}Am [see Fig. 1(b)] that causes their evidently different P_{f1}.

The second feature is that the slope of the curve P_{f1} *vs.* β, which reflects the sensitivity of first-chance fission probability to friction, is steeper for even-N ^{231}Am and ^{233}Am than for odd-N ^{232}Am. This is due to the OES phenomenon, which decreases the magnitude of P_{f1} of the latter and thereby weakens its variation with increasing β. Such a comparison clearly illustrates that on the experimental side, generating those heavy fissioning systems with an even number of neutrons can enhance the sensitive dependence of first-chance fission probability on friction.

We check those observations made from Am isotopes [Fig. 4(a)] for cases of Pu isotopes (whose Z is even) and light Np isotopes and arrive at similar results; that is, P_{f1} of ^{225}Np (^{227}Np) and ^{230}Pu (^{232}Pu) depend more sensitively on β than that of ^{226}Np and ^{231}Pu whose N is odd, see Figs. 4(b) and 4(c). Thus, our calculations demonstrate that when using P_{f1} to constrain β, if taking account of the marked OES phenomenon, then fissioning nuclei can be divided into two categories: One category contains an even N, and the other category contains an odd N. Figure 4 illustrates that experimentally, producing heavy nuclei with even neutron numbers can provide a more stringent constraint on friction by first-chance fission probability.

Finally, we study the influence of excitation energy on OES. In Refs. [3,54,55], it was shown that OES appearing in fragment charge and isotope distributions, produced in intermediate- and relativistic-energy heavy-ion collisions as well as in spallation reactions, seem to be insensitive to the beam energies. Analyses [3,9,10,54–56] suggested that it is because the OES emerges at the end of the evaporation chain of hot nuclei as the excitation energy is close to the nucleon-emission threshold energy, i.e., OES occurs at low excitation energies, which leads to the beam energy independence of the OES.

Unlike fragment cross sections, whose production experiences a sequential decay of a hot source, which can be populated through different reaction mechanisms mentioned above, the first-chance fission probability describes the first-step decay process and hence, it could be affected more strongly by the excitation energy of the hot nucleus. This expectation is confirmed in Fig. 5. There, as an illustration P_{f1} of Am isotopes are calculated at three different excitation energies in the presence of dissipation effects. As seen, P_{f1} decreases with increasing E^*, which is ascribed to an enhanced neutron emission probability at a higher energy. Moreover, it

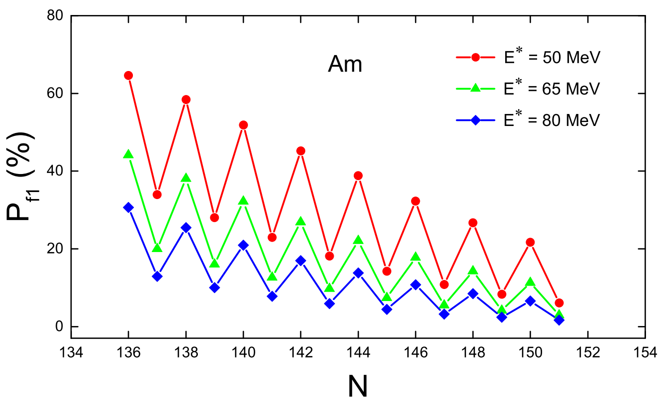

FIG. 5. First-chance fission probability (P_{f1}) of Am isotopes as a function of their neutron numbers N calculated at $\ell_c = 30\hbar$ and a presaddle dissipation strength of 3 zs^{-1} for three excitation energies $E^* = 50$, 65, and 80 MeV.

is noted that a higher E^* also decreases the difference in P_{f1} of two neighboring Am isotopes; that is, the OES is weakened with increasing E^*, clearly showing its dependence on excitation energy. This is because at a higher energy, the competition between neutron evaporation and fission is dominantly controlled by excitation energy, and the neutron binding energy plays a relatively small role, which decreases the influence of the neutron pairing energy on P_{f1} of two neighboring fissioning isotopes and hence reduces the amplitude of OES. This result indicates that in order to better reveal odd-even stagger through first-chance fission probability, it is optimal to yield heavy compound systems with a lower energy.

Conclusions. Using the dynamical Langevin equation that is coupled to a statistical model of particle emission, we have calculated first-chance fission probabilities (P_{f1}) for various Am, Pu, and Np isotopes. We have found P_{f1} of these heavy isotopes to display a prominent neutron odd-even stagger (OES), an apparent dependence of the OES on excitation energy, and a sizable dissipation effect on OES. Apart from that, the amplitude of the OES, denoted by ΔP_{f1} (which is the difference in P_{f1} of two neighboring fissioning isotopes), has been shown to be very sensitive to the dissipation strength (β), illustrating that the new experimental signal is a good probe of nuclear dissipation. Furthermore, it has been shown that P_{f1} of an even-N fissioning nucleus exhibits an obviously stronger sensitivity to β than that of its neighboring odd-N isotope, revealing a significant role of the OES phenomenon in accurately surveying β with P_{f1}. These results suggest that experimentally, to more strictly limit dissipative fission properties through the measurement of first-chance fission probability, it is best to populate heavy compound systems with even neutron numbers.

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