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A dynamical model calculation to reconcile the nuclear fission lifetime from different measurement techniques

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The pre-scission particle multiplicities suggest a lifetime of $\sim 10^{-20}$ s for the nuclear fission to occur which is in contrast to the fission lifetime $\sim 10^{-18}$ s as predicted by atomic probe. This long standing ambiguity, arising due to the orders of magnitude differences among the fission lifetime measured from the nuclear and atomic probes, has been addressed within a dynamical model which includes the contributions from the nuclear shell effects. We show that, at lower excitation energies, these two probes decouples as the fissioning system survives for a long time without any particle evaporation. We also consider a wide range of reactions to study the impact of the excitation energy of compound nucleus on the fission dynamics in general. Prescission neutron multiplicity is found to be an inappropriate probe to measure fission lifetime at low excitation energy. In addition, our model predicts the average fission life time of superheavy nucleus $^{302}120$, to be more than 10^{-18} s which is in reasonable agreement with the recent experiments.

Introduction – Nuclear fission is a fundamental decay mode for very heavy atomic nuclei. The formation and survival of superheavy elements [1–4] is strongly governed by the associated reaction dynamics and, in particular, the fission probability. Moreover, the fission rate critically influences the origin of elements heavier than iron [5–7]. Therefore, a precise understanding of the fission lifetimes is of extreme importance.

Substantial effort has been made to measure the fission lifetime. Traditionally, the pre-scission charged-particles [8–12], neutron [9, 11–20], and γ -ray [21–24] multiplicities are often used as a clock to estimate the fission time. The transient time also can be estimated by measuring the fission fragment distributions [25]. In general, all these nuclear techniques indicate that the fission process is fast enough with an upper bound in average fission time: $\langle\tau_f\rangle \leq 10^{-19}$ s. These nuclear probe encompass different variants of dissipative model to bridge the experimental observables related to the fission lifetime. Often a simplified statistical model is assumed to mimic the actual dynamics [26, 27]. On the other hand, the blocking technique in single crystals, which is considered to be a direct probe, leads to scission time scales much longer than the ones inferred from the nuclear methods [28, 29]. This contradiction is intensified after several recent crystal-blocking measurements [30–33] that indicate attosecond (10^{-18} s) time delay in heavy-ion induced fission. It is shown recently that K X-ray emission prior to fission can be used to measure fission lifetimes [34, 35]. This method measures the long fission-time component in

agreement with the crystal-blocking technique. Both the atomic clocks are used to explore the survival of superheavy element with $Z = 120$ [32, 35]. A detailed review on the study of fission lifetime can be found in Ref. [19].

Theoretical modeling of fission is extremely challenging as it involves many-body quantum dynamics. Recently, the self-consistent density functional theory is successfully applied for the studies of the spontaneous fission and thermally induced fission. The spontaneous fission half-lives are calculated using advanced energy density functionals [7, 36, 37]. Along this direction, the importance of nuclear pairing in spontaneous fission half-life is demonstrated [38–40]. In case of thermal fission, the time-dependent generator coordinator method assuming the Gaussian-overlap approximation is used successfully to study the fission fragment distributions [41, 42]. A thorough review of all these works is given in Ref. [43]. On the other hand, time-dependent density functional theory (TDDFT) methods made substantial progress to describe low-energy thermal fission [44]. Recently, density fluctuations are incorporated within TDDFT approach to generate spontaneous-fission yield distributions and kinetic energy distributions of fission fragments[45]. However, the situation becomes more complicated for induced fission from excited states, where pairing is quenched, compound system is formed with a high angular momentum, and dynamics becomes strongly dissipative and non-adiabatic [46]. The use of a complete microscopic theory is prohibitively expensive to simulate the fission process of a rotating hot nucleus in coincidence with light particles and γ -ray evaporations. In this regime, stochastic transport theories are successfully applied to describe the energy transfer between the collective and intrinsic degrees of freedom of the fissioning nucleus [26, 47, 48]. Among such theories, dynamical approaches based on the Langevin equation and its derivatives have been success-

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ful in reproducing fission dynamics [27, 49–53].

In the present work, an explicit simulation of large amplitude collective dynamics is performed, without incorporating any statistical model description, to extricate the long standing ambiguities in the fission lifetime measurements. Over the years, different theoretical propositions are attempted to resolve the fission lifetime ambiguity [19, 54, 55]. Particularly, an unrealistically large dissipation strength is used to reproduce the atomic-clock results [26, 56]. The present work, for the first time, reproduce the atomic-clock data using the dissipation strength calculated from a realistic model. Moreover, the applicability of the nuclear probe is carefully scrutinized over a wide range of excitation energy.

Theoretical framework – We have implemented a state-of-the-art model based on stochastic Langevin equation to study the full dynamical evolution of an excited compound system starting from the ground-state configuration up to the scission. The Funny-Hills shape parameter c [57], which represents the elongation of a nucleus, is used as the collective coordinate. The one dimensional Langevin equation [26, 47, 58] in terms of c is given as,

$$\begin{aligned} \frac{dp}{dt} &= -\frac{p^2}{2} \frac{\partial}{\partial c} \left(\frac{1}{m(c)} \right) - \frac{\partial F}{\partial c} - \eta(c)p + g\Gamma(t), \\ \frac{dc}{dt} &= \frac{p}{m(c)} \end{aligned} \quad (1)$$

where, p is the momenta conjugate to c ; the strength of the random force, g , is related to the friction coefficient η through fluctuation-dissipation theorem: $g = \sqrt{\eta T}$ [47]. The shape-dependent collective inertia $m(c)$ is extracted using Werner-Wheeler approximation [59, 60] for the irrotational flow of incompressible nuclear fluid. The chaos-weighted wall friction model [60, 61] is used to calculate the friction coefficient $\eta(c)$ as it seems to be most suitable for the present purpose [62–64]. The $\Gamma(t)$ represents the random force with the time correlation property $\langle \Gamma(t) \rangle = 0$ and $\langle \Gamma(t_1) \cdot \Gamma(t_2) \rangle = \delta(t_1 - t_2)$. The Helmholtz free energy F is used as the driving force for the collective motion. Specifically, we assumed the Fermi gas model [65] to define: $F = V - (a - a_0)T^2$, where V and a are the deformation dependent potential energy and level density parameter [66], respectively, a_0 being the value of a at the ground-state deformation. The energy and deformation dependent shell-correction is incorporated in a following Ignatyuk's prescription [67]. The temperature T is obtained from the ground-state excitation energy E^* as: $T = \sqrt{E^*/a_0}$. The average liquid-drop part of V is calculated following the double-folding Yukawa-plus-exponential model [48] and the associated shell-correction energy is obtained by applying the Strutinsky's method [57, 68] of shell-correction to the nucleonic levels generated with the two-centered Woods-Saxon mean field [69]. We use the BCS pairing to account the nuclear super-fluidity [57, 69]. Calculated potential barriers are found to be in good agreement with the existing results [70]. Langevin equations are solved numerically using finite difference method [47]. For this purpose, we

performed a second order expansion of Eq. 1 in terms of small time increment δt . We consider $\delta t = 10^{-25}$ s in the present work. Large scale computing is executed for an ensemble of 10^6 Langevin events to generate the results for a single macrostate. Each of the Langevin trajectories is allowed a maximum dynamical time of 10^{-15} s. For each event, the initial angular momentum of the compound nucleus is sampled from the corresponding fusion spin distribution. We consider the scission to occur when the neck radius of the compound nucleus becomes equal to $0.3R_0$ [57, 59], R_0 being the spherical radius.

Evaporation of light particles - n , p , α and statistical γ -rays are sampled at each time step of the dynamical evolution by using the Monte-Carlo technique [26]. In case of any evaporation, the compound system and the associated energy and angular momentum are modified accordingly. Therefore, in advance, we calculate all the inputs for a total of 48 daughter nuclei that leaves open the possibility of fifteen neutron (n) evaporations in combination with either a proton (p) or an α evaporation. It ensures the scope of all the feasible evaporation channels for the present study. For each fission event, we record the average n -evaporation time τ_n , the time

FIG. 1. (Color online) Distributions of τ_f , τ_{nl} , and τ_n as labelled for initial excitation energy (a) $E^* = 37$ MeV, (b) $E^* = 97$ MeV, and (c) $E^* = 187$ MeV. Inset in the panel (a) depicts the changeover of τ_f -distributions with E^* . The peak of τ_f -distribution for $E^* = 37$ MeV is indicated by arrow.

τ_{nl} when the last n is evaporated, and the scission time τ_f . According to the neutron-clock [15], the product: $\langle \tau_{n1} \rangle = n_{pre} \langle \tau_n \rangle$ gives the average fission lifetime, where n_{pre} is the average pre-scission neutron multiplicity. In practice, n -decay width, which is directly related to τ_n , is calculated and combined with a suitable fission-decay model to reproduce the experimental n_{pre} . Effectively, the measured n_{pre} determines the experimental fission lifetime [8, 9, 15]. Equivalently, the n -clock can be devised using τ_{nl} with the underlying assumption that scission occurs immediately after the last neutron is evaporated. Of course, it may be violated in practice and the present work investigates the faith of this assumption viz-a-viz the applicability of n -clock. We compare these two neutron probes with the actual scission time τ_f . In addition, for a comprehensive understanding of the dynamical evolution, we calculate the average deformation $\langle d_f \rangle$ of the fissioning system by taking the time average of the collective coordinate for each event.

The present model avoids a major approximation of the existing CDSM codes. In case of CDSM [26, 56, 62, 71, 72], Langevin dynamics is performed up to an initial transient time (10^{-19} s in [26, 56, 62]) and then a suitable statistical model is used to decide the faith of fission events. These statistical models usually consist approximate stationary fission width that neglects the details of deformation dependence of the input quantities. In contrast, dynamics is followed in the present work till the system bifurcate or an evaporation residue is formed. Further, the deformation dependent shell effect is accounted coherently within the potential energy and nuclear level density parameter.

Results and Discussion – We first consider the $^{16}\text{O} + ^{208}\text{Pb} \rightarrow ^{224}\text{Th}$ reaction since this system is well-studied experimentally [15, 73]. The normalized yields corresponding to τ_n , τ_{nl} , and τ_f calculated for different values of initial excitation energy E^* are shown in Fig. 1. Evidently, at large E^* (Fig. 1(b)-(c)), the distributions of τ_{nl} and τ_f almost coincide except for the long-time tail in τ_f . This behavior of τ_f is reported in Refs. [26, 71] using CDSM calculations. In contrast, for the lowest E^* (Fig. 1(a)), the shape of τ_f becomes very broad with the peak at $\tau_f > 10^{-18}$ s, whereas the shapes of τ_n and τ_{nl} remain almost unaffected. This decoupling of τ_f from τ_{nl} and τ_n appears somewhere between $E^* = 67$ MeV and 37 MeV (inset of Fig. 1(a)). It emphasizes the fact that, at a lower energy, the fissioning system survives for a long time without any particle evaporation as the available excitation energy falls below the particle emission threshold. Additionally, we found that the long fission-time component is further enhanced by the nuclear shell effects as conjectured in [26, 71].

For a deeper understanding of the nature of τ_f , we calculated the correlation between τ_f and $\langle d_f \rangle$. The corresponding two-dimensional distribution of fission events are plotted in Fig. 2(a) and Fig. 2(b), respectively, for the lowest and highest E^* considered in Fig. 1. Also, the free energy F for different values of T are shown in

FIG. 2. (Color online) Distribution of fission yields on the $\tau_f - \langle d_f \rangle$ plane for (a) $E^* = 37$ MeV and (b) $E^* = 187$ MeV. (c) The variation of free energy F with deformation d_f for different temperature as indicated.

Fig. 2(c). Clearly, the events with a long fission-time predominantly stay around the ground state deformation ($0.95 \leq d_f \leq 1.1$). Here, $d_f = 1$ corresponds to the spherical configuration. On the other hand, the average deformation increases for the high energy fission events as the free energy profile becomes flatter. This observation clarifies the ambiguity related to the role of deformation-dependent dissipation in escalating fission lifetime. Since majority of the long-time events roam around the ground state deformation, these are hardly affected by the dissipation near scission.

We have calculated the average fission time $\langle \tau_f \rangle$, $\langle \tau_{nl} \rangle$, and $\langle \tau_{n1} \rangle$ associated to the distributions of τ_f , τ_{nl} , and τ_n , respectively. These are plotted in Fig. 3(a) along with n_{pre} in Fig. 3(b). As expected, $\langle \tau_{nl} \rangle$ and $\langle \tau_{n1} \rangle$ remain very close to each other at all energies. For higher E^* , $\langle \tau_f \rangle$ is comparatively large due to the presence of long-time tail. Where as, at low E^* , it is one order of magnitude higher than those for the other two distributions due to the decoupling of τ_f from τ_{nl} and τ_n as shown in Fig. 1(a). Moreover, the absolute value of $\langle \tau_f \rangle$ reaches the attosecond time-scale in agreement with atomic measurements. One experimental data of fission lifetime is available for the present system from neutron multiplicity measurement and it matches well with our calculated

FIG. 3. (Color online) (a) Average fission lifetime $\langle \tau_f \rangle$ (dash-dotted line), $\langle \tau_{n1} \rangle$ (dashed line), $\langle \tau_{nl} \rangle$ (solid line) as a function of E^* . The symbol indicates the experimental data [15]. (b) Comparison of experimental [15, 73] n_{pre} with the calculated values.

$\langle \tau_{nl} \rangle$ and $\langle \tau_{n1} \rangle$. It is clear from Fig. 3(b), the experimental neutron multiplicities are reproduced simultaneously without any free parameter.

After the benchmark study on ^{224}Th , we computed the fission lifetime for several other reactions. Specifically, we considered the reactions: (1) $p + ^{238}\text{U}$, (2) $^4\text{He} + ^{232}\text{Th}$, and (3) $^{19}\text{F} + ^{181}\text{Ta}$ covering a wide mass range relevant to fission. The excitation energy dependence of $\langle \tau_f \rangle$ for these reactions are demonstrated in Fig. 4. The $\langle \tau_f \rangle$ for both the actinides vary similarly and become slower than a attosecond for $E^* \leq 90$ MeV. This behavior is in compliance with the predictions from the atomic probe. The upper limit of $\langle \tau_f \rangle$ for the reaction (1) is measured [28] for the lowest E^* (indicated by down arrow in Fig. 4). As evident in Fig. 4, our calculation follows this limit. For reaction (3), $\langle \tau_f \rangle$ remains more than 10^{-17} s even at a very large E^* . This system has been studied extensively [74] around $E^* = 100$ MeV and, subsequently, analyzed theoretically in Ref. [56]. In their analysis, a comparatively large reduced friction was required within the dynamical model to delay fission. However, our calculation reproduces the lifetime of more than 10^{-17} s without recouring to the tuning of any input parameter. To determine the origin of large fission lifetime, we extracted the distributions of τ_f , τ_{nl} , and τ_n (similar to Fig. 1) in Fig. 5. Interestingly, the second peak in τ_f has substantial contribution between $10^{-17} - 10^{-15}$ s that results in a large $\langle \tau_f \rangle$. It appears because of strong ground state shell correction in ^{200}Pb that hinders the system to over-

FIG. 4. (Color online) Average fission lifetime $\langle \tau_f \rangle$ as a function of excitation energy for the reactions (1)-(3) as mentioned in text.

come the barrier. Although this effect should disappear with excitation energy, the evaporation of neutrons helps it to persist even at high excitation energy. The presence of this broad second peak is consistent with the crystal-blocking data in Ref. [74].

FIG. 5. (Color online) Distributions of τ_f , τ_{nl} , and τ_n for the reaction $^{19}\text{F} + ^{181}\text{Ta}$.

Finally, we address the fission lifetime for the $^{238}\text{U} + ^{64}\text{Ni}$ reaction which is proposed to be a possible candidate for the discovery of $Z = 120$ isotopes. Several studies have been made to this end [32, 35, 54]. Crystal blocking measurement [32] predicts that the $^{302}120$ nuclei can survive more than 10^{-18} s. In this experiment, the initial excitation energy of the compound system was uncertain due to the large target thickness. We found, as shown in Fig. 6, that the $\langle \tau_f \rangle \geq 10^{-18}$ s is possible for this system only if the excitation energy $E^* \leq 10$ MeV

FIG. 6. (Color online) Average fission lifetime as a function of excitation energy calculated with full (dashed line) and reduced (solid line) energy dependence in shell correction. Vertical arrows indicates the E^* corresponding to $\langle \tau_f \rangle = 10^{-18}$ s.

(dashed lines). Therefore, the long-lived component in the experiment may be contributed from very low energy events. The stability of superheavy elements is very much sensitive to nuclear shell effects. Thus, an accurate estimation of nuclear potential energy surface is essential for an reliable prediction. In our calculation, the average fission barrier for $Z = 120$ isotopes is 9 MeV which is close to the microscopic density functional prediction [75]. The multidimensional macro-micro prediction [76] is 2 MeV smaller than this value. Although, the fission lifetime is strongly influenced by the fission barrier, the temperature dependence of the potential surface apparently dilutes this effect. So, a proper modeling of the finite-temperature potential is crucial. Within the Fermi-gas model, the shell effects washes out much faster, as shown in Fig. 2(c), compared to the finite-temperature shell model predictions [77]. To explore the effect of shell-washing in the present system, we performed an-

other set of calculation with a modified shell damping factor. Specifically, the energy dependence of the level density parameter a is reduced to 50% of its unaffected value.

The corresponding $\langle \tau_f \rangle$ is plotted in Fig. 6 and it shows that, with this reduction, the attosecond time can be reached at a higher $E^* \sim 30$ MeV (solid line). This study reveals the importance of an appropriate microscopic calculation as far as superheavy elements are concerned.

Conclusions – A state-of-the-art calculation of fission dynamics is performed for excited compound systems where - (i) the dynamics is followed from the ground state deformation to scission including all possible evaporation channels, (ii) the energy and deformation dependent shell-effect is properly accounted, and (iii) no parameters are tuned to reproduce a specific observable. The discrepancy between the atomic and nuclear probes for the fission lifetime measurement is resolved by comparing the scission time with the indirect predictions from evaporated neutrons. The neutron multiplicity probe is found to be inappropriate to estimate fission lifetime at low excitation energies. Our results comply with the crystal-blocking measurements for actinides. Also, we have shown that both the measurement methods should agree at sufficiently high excitation energy where crystal-blocking data are still missing. Moreover, the attosecond fission timescale around mass ~ 200 region is reproduced with the same model parameters. Finally, for the generalization of our finding, calculation is performed for superheavy $Z = 120$ nucleus. The attosecond lifetime for $Z = 120$ nucleus is found to be consistent with our theoretical calculations.

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