Fission process of low excited nuclei with Langevin approach

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Fragment mass distributions from fission of U and Pu isotopes at low excitation energies are studied with a dynamical model based on the fluctuation-dissipation theorem formulated as Langevin equations. The present calculations reproduced the overall trend of the asymmetric mass distribution without parameter adjustment for the first time in terms of the Langevin approach. The Langevin trajectories show complicated time evolution on the potential surface which causes time delay of fission, showing that dynamical treatment is vital. It was found that the shell effects of the potential-energy landscape has the dominant role to determine the mass distribution, while it is rather insensitive to the strength of dissipation. Still, it is indispensable to include the effect of dissipation since it gives a crucial role to give “fluctuation” to Langevin trajectories as well as to explain multiplicities of pre-scission neutrons as excitation energy increases. Therefore, the present approach can serve as a basis for more refined analysis.

I. INTRODUCTION

Discovery of nuclear fission [1, 2] opened an important chapter not only in study of nuclear physics, but also in technology of energy supply. After the nuclear power plant accident at Fukushima in 2011, further understanding of the fission process became to be required to predict quantitatively the amount of heavy elements and radioactive fission products remaining as “debris”, or melted spent nuclear fuel, which is still present in the remnant of the power plant today. Moreover, such information is also important to make emerging nuclear power plants all over the world much safer. Therefore, further study of the nuclear fission process has been necessitated.

Just after the discovery, nuclear fission was interpreted in analogy with that of a charged liquid drop; fission occurs as a result of a competition between the disrupting effect of Coulomb repulsion and the stabilizing influence of surface tension. Bohr and Wheeler founded this idea and invoked the liquid drop model to describe the process [3]. However, that concept could not explain the asymmetric mass splitting which is the dominant mode of fission in the region of nuclear fuel, namely, U and Pu nuclei.

A fission model which used the level densities of the nucleus at the ground state and the saddle point was developed, namely statistical model. It was possible with this model to explain the qualitative feature of the mass-asymmetric splitting by introducing several phenomenological parameters. This model, however, did not include dynamics of fission process, that, starting from an almost spherical nucleus, nucleus overcomes the fission barrier and passes through the scission point. In the meantime, pre-scission particles, especially the neutrons and $\gamma$-rays, may be emitted, which alters the excitation energy and potential-energy landscape of the fissioning system. In 1980’s, many experimental data indicated that the pre-scission neutron multiplicities from highly excited nucleus exceeded the expectation based on the statistical model [4]. To explain this observation, the concept of dissipation, which was suggested by Kramers in 1940 [2], was recalled.

By taking account of nuclear friction, which is the coupling between fission degree freedom (collective motion) and other degrees of freedom such as nucleon single-particle motions, it was expected that there exist a time delay necessary for deforming the system from spherical shape to that of elongated saddle, which deserves a time for nucleons (mostly neutrons) to escape from the fissioning hot nuclei. It was concluded that the pre-scission neutron multiplicities by this picture obviously exceeded the prediction of statistical model and in good agreement with observations [6]. On the other hand, the mass asymmetric fission, e.g., those produced by thermal-neutron induced fission of Th, U and Pu nuclei, might be linked to the microscopic structure of fissioning nucleus or fragments. However, the origin and the mechanism of asymmetric fission have not been clear so far. Recently, asymmetric fission of $^{180}$Hg was discovered following electron capture of $^{180}$Tl [5]. It was expected that a symmetric fission should occur from the statistical model picture due to the strong shell effects of the half magic nucleus $^{90}$Zr. The fission paths, however, seem to be terminated before the system reaches to apparently dominating configuration of populating two $^{90}$Zr nuclei. The dynamics, therefore, should play a key role in understanding fission.

To clarify this contradiction and give a possibly unified picture of the fission process, we need to introduce a dynamical model of fission starting from a nearly spherical shape to the scission region through the fission saddle point. Such a shape evolution proceeds in competition with pre-scission particle emissions, so dynamical treatment is essential. As such an approach, the method of Langevin equations based on the fluctuation-dissipation theorem has been applied to nuclear fission process by several groups to describe the process, including friction, inertia mass and multi-dimensional potential-energy surfaces [8–15]. These past investigations focused on systems having high excitation energy. The calculation resulted in symmetric mass distribution of fission fragments.
(MDFF) in good agreement with experimental data arising from relatively high-excitation energy. It reflects the properties of potential-energy surface of the liquid drop model. On the contrary, the dynamical calculation with Langevin equation has not been applied so much to fission process at low excitation energy [21], due to difficulties to obtain the shell correction energy of configurations in multi-dimensional space of collective coordinates and a huge computing time. The computation time was, however, reduced drastically with the recent advancement of computer technologies and utilizing parallel computing. Moreover, we can calculate the shell correction energy at each configuration by using the two-center shell model.

In this paper, we present the possibility to calculate dynamically the fission process at low excitation energy using Langevin equations, including the shell effects, pairing effects, dissipation and fluctuation. Using this model, we calculate the mass distribution of fission fragments (MDFF) of $^{236,234}$U and $^{240}$Pu at the low excitation energy and they were compared with experimental data. In this way, we can investigate the fission mechanism including the origin of mass asymmetric fission.

The paper is organized as follows. In Sec. II, we detail the framework of the model. In Sec. III, we show the results for MDFF for $^{236,234}$U and $^{240}$Pu at the excitation energy $E^* = 20$ MeV. In Sec. IV, we present a summary of this study and further discussion.

II. MODEL

We use the fluctuation-dissipation model and employ Langevin equations [21] to investigate the dynamics of the fission process. The nuclear shape is defined by the two-center parametrization [22,23], which has three deformation parameters, $z_0$, $\delta$, and $\alpha$ serving as collective coordinate: $z_0$ is the distance between two potential centers, while $\alpha = (A_1 - A_2)/(A_1 + A_2)$ is the mass asymmetry of the 2 fragments, where $A_1$ and $A_2$ denote the mass numbers of heavy and light nuclei, respectively [21]. The symbol $\delta$ denotes the deformation of the fragments, and is defined as $\delta = 3(R_{\|} - R_{\perp})/(2R_{\|} + R_{\perp})$, where $R_{\|}$ and $R_{\perp}$ are the half length of the axes of an ellipse in the $z_0$ and $\rho$ directions of the cylindrical coordinate, respectively, as shown in Fig. 1 in Ref. [22]. We assume in this work that each fragment has the same deformation. This constraint should be relaxed in the future work since the deformations of the heavy and light fragments in the fission of U region are known to be different from each other. The deformation parameters $\delta$ and $\beta_2$ are related to each other as

$$\beta_2 = \frac{\delta}{\sqrt{\frac{5}{16\pi} (3 - \delta)}}.$$

Notice that $\delta < 1.5$ since $R_{\|} > 0$ and $R_{\perp} > 0$. In order to reduce the computational time, we employ the coordinate $z$ defined as $z = z_0/(R_{CN} B)$, where $R_{CN}$ denotes the radius of a spherical compound nucleus and $B$ is defined as $B = (3 + \delta)/(3 - 2\delta)$. We use the neck parameter $\epsilon = 0.35$, which is recommended in Ref. [23] for the fission process. The 3 collective coordinates may be abbreviated as $q$, $q = \{z, \delta, \alpha\}$.

For a given value of a temperature of a system, $T$, the potential energy is defined as a sum of the liquid-drop (LD) part, a rotational energy and a microscopic (SH) part:

$$V(q, \ell, T) = V_{LD}(q) + \frac{\hbar^2}{2I(q)} + V_{SH}(q, T),$$

$$V_{LD}(q) = E_S(q) + E_C,$$

$$V_{SH}(q, T) = E_{shell}(q)\Phi(T),$$

$$\Phi(T) = \exp\left(-\frac{aT^2}{E_\delta}\right).$$

Here, $V_{LD}$ is the potential energy calculated with the finite-range liquid drop model, given as a sum of the surface energy $E_S$ [24] and the Coulomb energy $E_C$. $V_{SH}$ is the shell correction energy evaluated by Strutinski method from the single-particle levels of the two-center shell model. The shell correction have a temperature dependence expressed by a factor $\Phi(T)$, in which $E_\delta$ is the shell damping energy chosen to be 20 MeV [25] and $a$ is the level density parameter. At the zero temperature ($T = 0$), the shell correction energy reduces to that of the two-center shell model values $E_{0}^{SH}$. The second term on the right hand side of Eq. (2) is the rotational energy for an angular momentum $\ell$, with a moment of inertia at $q, I(q)$.

The multidimensional Langevin equations [21] are given as

$$\frac{d\dot{q}_i}{dt} = (m^{-1})_{ij} p_j,$$

$$\frac{dp_i}{dt} = -\frac{\partial V}{\partial q_i} - \frac{1}{2} \frac{\partial}{\partial q_i} (m^{-1})_{jk} p_j p_k - \gamma_{ij} (m^{-1})_{jk} p_j p_k + g_{ij} R_{ij}(t),$$

where $i = \{z, \delta, \alpha\}$ and $p_i = dq_i/dt$ is a momentum conjugate to coordinate $q_i$. The summation is performed over repeated indices. In the Langevin equation, $m_{ij}$ and $\gamma_{ij}$ are the shape-dependent collective inertia and the friction tensors, respectively. The wall-and-window one-body dissipation [26,27] is adopted for the friction tensor which can describe the pre-scission neutron multiplicities and total kinetic energy of fragments simultaneously [8]. A hydrodynamical inertia tensor is adopted with the Werner-Wheeler approximation for the velocity field [28]. The normalized random force $R_{i}(t)$ is assumed to be that of white noise, $i.e., \langle R_{i}(t) \rangle = 0$ and $\langle R_{i}(t_1) R_{j}(t_2) \rangle = \delta_{ij} \delta(t_1 - t_2)$.
FIG. 1. (Color online) The potential energy surface $V = V_{LD} + E_{\text{shell}}^0$ with $\epsilon = 0.35$ in (a) the $z - \alpha$ space at $\delta = 0.2$ and (b) the $z - \delta$ space at $\alpha = 0.0$ for $^{236}$U. The arrow indicates the fission valley which corresponds to $A = 140$.

$2\delta_{ij}\delta(t_1 - t_2)$. The strength of the random force $g_{ij}$ is given by Einstein relation $\gamma_{ij} T = \sum_k g_{ij} g_{jk}$.

The temperature $T$ is related with the intrinsic energy of the composite system as $E_{\text{int}} = aT^2$, where $E_{\text{int}}$ is calculated at each step of a trajectory calculation as

$$E_{\text{int}} = E^* - \frac{1}{2} (m^{-1})_{ij} p_i p_j - V(q, \ell, T = 0).$$

The excitation energy of the compound nucleus $E^*$ is given by $E^* = E_{\text{cm}} - Q$, where $Q$ denotes the $Q$-value of the reaction.

The fission events are determined in our model calculation by identifying the different trajectories on the deformation space. Fission from the compound nucleus is defined as the case that a trajectory overcomes the scission point on the potential energy surface. As an example, the potential $V_{LD} + E_{\text{shell}}^0$ with $\ell = 0$ and $\epsilon = 0.35$ in the $z - \alpha$ space at $\delta = 0.2$ and in the $z - \delta$ space at $\alpha = 0.0$ for $^{236}$U are shown in Figs. 1(a) and (b), respectively. In Fig. 1(a), the arrow indicates the fission valley which corresponds to $A = 140$. We do not take into account both the pre-scission and post-scission neutron emission in our calculation as our first step.

FIG. 2. (Color online) Sample trajectory projected onto the $z - \alpha$ plane at $\delta = 0.2$ (a) and $\delta = -0.2$ (b) of $V_{LD} + E_{\text{shell}}^0$ for $^{236}$U. The trajectory starts at $z = 0.65$, $\delta = 0.2$, $\alpha = 0.0$ at $E^* = 20$ MeV, where is corresponds to the second minimum of the potential energy surface, due to reduce the calculation time.

FIG. 3. (Color online) The trajectories in Fig. 2 is projected onto the $z - \delta$ plane at $\alpha = 0.0$.
FIG. 4. (Color online) Mass distribution of fission fragments of $^{236}$U at $E^* = 20$ MeV. Calculation and experimental data are denoted by histogram and circles, respectively.

FIG. 5. (Color online) Mass distribution of fission fragments of $^{234}$U at $E^* = 20$ MeV. Calculation and experimental data are denoted by histogram and circles, respectively.

III. MASS DISTRIBUTION OF FISSION FRAGMENTS

Figure 2 shows a sample trajectory that is projected onto the $z - \alpha$ plane at $\delta = 0.2$ (a) and $\delta = -0.2$ (b) of $V_{LD} + E_0^{\text{shell}}$ for $^{236}$U. The trajectory starts at $z = 0.65, \delta = 0.2, \alpha = 0.0$ at $E^* = 20$ MeV, where corresponds to the second minimum of the potential energy surface, due to reduce the calculation time. The trajectory stays around the pocket where is located at $\{z, \delta, \alpha\} \sim \{1.35, -0.2, 0.0\}$ with thermal fluctuations. Then, it escapes from the second minimum and moves along the valley which corresponds to $A \sim 140$. We project the trajectory of Fig. 2 onto the $z - \delta$ plane at $\alpha = 0.0$, as shown in Fig. 3. The trajectory moves in the pocket around the second minimum and it stays around $z \sim 1.35$ and $\delta \sim -0.2$ on the this plane, which corresponds to the pocket in Fig. 2(b). We discuss more precisely the fission dynamics analyzing the behaviors of trajectories in the forthcoming paper.

Figures 4 and 5 show the calculated (histograms) mass distribution (MDFF) for $^{236,234}$U together with the corresponding experimental data (dots) for neutron induced fission leading to the same compound nuclei at $E^* = 20$ MeV, respectively. The dots were taken from JENDL Fission Yield Data File 2011[30] to represent experimental data concisely. In the present calculation, we prepared 10,000 trajectories. It is equivalent to the number of trajectories of fission normalized to all the fission events of experimental data. Here, we assume $\Phi(T) = 1$ in Eq. (4), that corresponds to the full shell correction energy, to simulate the low-excitation energy introduced by thermal neutrons. The effects of $\Phi(T)$ will be discussed later. For these nuclei, the present approach yields results consistent with measured data without any adjustment of parameters in the Langevin calculation, showing predictive power of the present model. The widths and positions of the peaks are reproduced to a good accuracy. We consider that the trajectories move along the fission valley in Fig 1(a), which is indicated by the arrow. However, in more detail, the peak of light fragments in the calculation locates at heavier position by few mass unites. The discrepancy is caused partly by the the neutron emission from fission fragments, which is not included in our model.

We also calculate the MDFF of $^{240}$Pu at $E^* = 20$ MeV, which is shown in Fig. 6 together with the correspond-
ing experimental data. The results are obtained using the same parameters as the calculations for $^{234,236}$U. It agrees with experimental data quantitatively, and the tendency of the difference between calculated results and experimental data is similar to the cases of $^{234,236}$U.

The shell correction energy depends on the excitation energy of the nucleus $E^*$, or the nuclear temperature $T$. We discuss the temperature dependence of the shell correction energy and how the fission process and the MDFF are affected. Considerable effort has been made to investigate the temperature dependence of the level density parameter $^{234}$ and applied to the calculation of the statistical model for the fission process $^{234}$. The temperature dependence of the potential energy surface has been investigated in Refs. $^{234,234}$. Here, we assume that the temperature dependence of shell correction energy is described by Eq. $^{234}$ with the factor Eq. $^{234}$. The value of the shell damping energy has uncertainty still now $^{234}$, even it was suggested to use 20 MeV by Ignatyuk $^{234}$. Using the several values of the shell damping energy, we investigate the affection of the MDFF. Figure 7 show the MDFF of $^{236}$U at $E^* = 20$ MeV with $E_d = 16.3, 20, 30$ and 40 MeV. In the case of $E_d = 16.3$ MeV, the effect of shell correction energy is weaker than the case of $E_d = 40$ MeV, in this system. So, the MDFF with $E_d = 16.3$ MeV shows that mass symmetric fission events are larger than the other cases, due to the effects of the potential energy surface of the liquid drop model. However, the gross features of each case do not change so much. Here, we plot the MDFF using $V_{LD}$ which is denoted by the dark-blue line (full LDM). It shows the mass symmetric fission, which follows the structure of $V_{LD}$.

The MDFF is essentially governed by the dynamics of the trajectories in the potential energy surface and it is affected by the friction and inertia mass. We investigate the MDFF of $^{236}$U at $E^* = 20$ MeV by varying the strength of the friction tensor. Figure 8 shows the MDFF for the frictions $\gamma$ multiplied by the various factors, 0.1, 0.5, 1, 2 and 5. Here, we assume $\Phi(T) = 1$ in Eq. $^{234}$. The present results are rather insensitive to the strength of the fraction, because the excitation energy is low. In the case at the low excitation energy, the fluctuation of trajectories is not so much, and the trajectories are mostly affected by the landscape of the potential energy.

The great benefit of the dynamical calculation with
Langevin equations is that we can investigate time scale of the fission process. Time-dependent decay rate is governed by nuclear collective dynamics, including fluctuation, dissipation, etc. The study of fission time scale is also important in the nuclear engineering since emission of pre-scission neutrons as a process competing to fission alters excitation energy of the fissioning system, therefore it affects many phenomena such as MDFF, number of prompt neutrons, their energy spectra and also number of β-delayed neutrons.

The time evolution of the MDFF of $^{236}$U at $E^* = 20$ MeV shows Fig. 9. Till $t = 1.0 \times 10^{-20}$ sec, the trajectories do not reach at the scission point. Then, the fission events increase as time goes by. Figure 10 shows the time evolution of number of fission events for $^{236}$U at $E^* = 20$ MeV with logarithmic scale for time. Here, in the Langevin calculation, we prepare 10,000 trajectories. We can see the almost all of fission events occur dynamically till $t = 1 \times 10^{-18}$ sec. After $t = 1 \times 10^{-18}$ sec, the trajectories within the fission barrier are strongly or deeply trapped into the pockets, which are presented in Fig. 11. It needs a lot of time to escape from the pockets by the fluctuation. Till $t = 1 \times 10^{-18}$, 29% of all trajectories reach at the scission point. It means that the 71% of all trajectories are still trapped into the pockets around the ground state at this time. Such trajectories need more time to escape from the pockets or will decay by neutron emission, which is not treated in the present model. It should be an important future subject.

**IV. SUMMARY**

In this article, we investigate the fission process at a low excitation energy by Langevin equations. We calculate the MDFF of $^{234,236}$U and $^{240}$Pu at $E^* = 20$ MeV, and the results show the mass asymmetric fission which is in good agreement with the experimental data without any parameter adjustment. It is the first time to obtain the MDFF by the Langevin calculation with shell effects and to compare it with experimental data. In the present model, we use three collective variables to describe the fissioning nuclear shape. We discuss the origin of the mass asymmetric fission by analyzing sample trajectories. The trajectory analysis allows us to directly observe the time evolution of the dynamical process.

The calculated results depend slightly on the shell damping energy. However, dependence to the strength of the friction tensor was weak. It does not mean that friction is not important in the study of fission since variation of observables due to change in the excitation energy is important, and, at higher energy, friction plays a very important role. Therefore, inclusion of the effect of friction is important for a unified treatment of fission in a consistent manner and to apply the method for nuclei like minor actinides for which experimental data are extremely scarce. At the same time, it became clear that dynamical treatment is vital since the Langevin trajectories show rather complicated time evolution on the potential-energy surface. Especially, they spend long time in the first and second potential minima, and exhibits “fission time delay”, during which competition to neutron evaporation may occur.

The reproduction of the experimental MDFF in this model can be the ground to support the investigation of fission dynamics at the low excitation energy. Furthermore, the generalized formula proposed in this model has a potential to simulate any kind of nuclear fission which appears in the nuclear engineering fields. Such prediction is becoming particularity necessary after the accident at Fukushima in 2011 to handle nuclear waste safely and make the emerging nuclear power plants much more safer.

It should be noted that, a random walks method on the potential energy surface including shell correction energy was proposed and applied to fission process at the low excitation energy [30]. This method gives also good reproduction of experimental mass yields of $^{240}$Pu and $^{236,234}$U. It seems to be a quite useful calculation tool, although it could not discuss the time-scale of fission process and the dynamics.

For further study, we plan to improve the model to decrease the differences between calculated results of the MDFF and experimental data. We need to increase the number of variables, at least to introduce independent deformation parameters of each fragment. Moreover the neutron emission from fissioning system and also from fission fragments should be included in the model. Microscopic treatment of the transport coefficients may also be important especially at low excitation energies as treated in this paper. Since it takes more computing time to solve the Langevin equation by incorporating such improvements, we have to modify the computing algorithm to suit to the high performance computers and utilize
parallel computing. Still, the present approach can serve as a basis for these more refined analysis aiming at a realistic description of the whole process of fission, starting from the compound nuclei at various excitation energies reaching to the fission products populated after $\beta$-decay, which is supported financially by MEXT through JST.

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