Possible ternary fission of super-heavy nuclei

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Abstract. In super-heavy nuclei, we may expect to discover new phenomena because of the strong Coulomb force. One example is the true ternary fission. We study the sequential two binary fission of 300120 and 252Cf that produces three fragments. We use the Metropolis method to estimate the probability of the second fission. The probability of the second fission is $10^{-4}$–$10^{-3}$ for super-heavy nuclei while it is $10^{-7}$–$10^{-6}$ for heavy actinide nuclei. The most probable mass division is almost symmetric in the case of 300120. We also demonstrate the applicability of the Metropolis method to a non-equilibrium process by comparing it with the Langevin equation.

1 Introduction

In super-heavy nuclei, because of the large atomic number, one may expect to discover new phenomena that we could not observe in lighter nuclei. One example is a new fission such as the true ternary fission where a massive nucleus decays into three fragments of not very different masses [1]. For actinide nuclei, ternary fission means the light-charged-particle-accompanied binary fission. It is characterized by the angular distribution of the light charged particle that is peaked around the direction perpendicular to the fission axis. It is interpreted that the light particle is emitted from the neck region so that it is accelerated by the Coulomb fields of the two heavy fragments. The possibility that a heavy excited nucleus might split into three fragments of comparable mass was considered soon after the discovery of fission. Such decays of low excited heavy nuclei have not been unambiguously observed. For thermal neutron fission of U and Pu, a relative frequency of ternary to binary events is reported to be the order of one in $10^6$ [1]. There have been attempts to predict the possible combination of the three fragments for super-heavy nuclei [2, 3]. Zagrebaev et al. [2] found that ternary decays with the formation of two heavy and a lighter third fragment are possible for super-heavy nuclei. By assuming symmetric ternary fission, they obtained $^{132}$Sn-$^{32}$S-$^{132}$Sn colinear configuration for $^{296}$116. Later, Vijayaraghavan et al. [3] calculated the yield of “colinear cluster tripartition” of $^{252}$Cf (measured by Pyatkov et al. [4]) within the framework of three cluster model. They obtained $^{88}$Se+$^{82}$Ge+$^{82}$Ge as one probable fragment combination. These works are essentially based on the argument of the final Q-value of the reaction. In order to obtain more quantitative estimates of ternary fission in very heavy and super-heavy nuclei, we need a dynamical approach that takes account of the fission saddle heights.

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It is known, especially for spontaneous fission, that shell correction energy plays an important role in determining the mass distribution of the fragments. Recently, Carjan et al. \[5\] calculated the pre-scission configuration for the binary fission of super-heavy nuclei using the microscopic-macroscopic approach. They suggested that the $N = 82$ neutron shell plays a dominant role in determining the most probable mass separation. The light fragment has the mass number around 136 and has almost spherical shape. On the other hand, the heavy fragment is found to be extremely elongated. It is possible that the elongated heavy fragment undergoes a subsequent binary fission thus producing three fragments. In this paper, we will estimate the probability of ternary fission in comparison with that of the binary fission. In Sect. 2, we will describe our framework and in Sect. 3, the results are given with discussions. In Sect. 4, we will give the summary.

2 Framework

First, we select the most probable binary mass separation and determine the nuclear shape just before scission by the pre-scission model \[6\]. The shape of the mother nucleus is expressed by the generalized Cassinian oval parameterization \[7\]. In the original Cassinian oval parameterization, the parameter $\alpha$ specifies the elongation of the nucleus; $\alpha = 0$ corresponds to the spherical shape and $\alpha = 1$ corresponds to the zero-neck shape \[7\]. In the generalized Cassinian oval parameterization, nuclear shapes are described in terms of Cassinian ovals generalized by the inclusion of additional shape parameters: $\alpha_1$, $\alpha_3$, $\alpha_4$ and $\alpha_6$. The pre-scission shape is calculated by minimizing the deformation energy with respect to $(\alpha_1, \alpha_3, \alpha_4, \alpha_6)$ for $\alpha_1 = 0.98$. The deformation energy is calculated with the microscopic-macroscopic approach \[8\]. We show, in Fig. 1, the pre-scission shape of a super-heavy nucleus $^{300}120$. As can be seen, the light fragment of mass 136 is almost spherical while the heavy fragment is strongly deformed. Since we are interested in the probability of the second fission of the deformed fragment, the shape of the deformed fragment is fitted by the generalized Cassinian oval parameterization as shown by the red curve in Fig. 1.

![Figure 1. Pre-scission shape of the mother nucleus $^{300}120$.](image)

The development of the shape of the deformed fragment is calculated by means of the Metropolis method. The specific algorithm introduced by Metropolis \[9\] has two parts: 1. Each configuration $\chi$ has a specified set of “neighbours” $\{\chi'\}$. 2. When the system is at the configuration $\chi$, the next configuration in the sequence is sampled as follows: first, a tentative next configuration, $\chi'$, is selected randomly from the set of neighbours and it is then decided
whether the system makes a “step” to that new configuration $\chi'$ or remains at the current configuration $\chi$. The Metropolis step probability is given by

$$P(\text{step}) = \begin{cases} 1/W(\chi')/W(\chi) & (W(\chi') > W(\chi)) \\ 1 & (W(\chi') < W(\chi)). \end{cases}$$

(1)

Namely, if the statistical weight of the tentative configuration is greater than that of the current configuration then the step probability is one, and in the opposite case, then the probability of taking the tentative configuration is $W'/W$ and the probability of staying in the current configuration is $1 - W'/W$. In the present work, we assume that the statistical weight is proportional to $\exp(-V(\chi)/T)$, where $T$ is the temperature of the system, then the step probability is given as

$$P(\text{step}) = \begin{cases} 1 & (V(\chi') < V(\chi)) \\ \exp\left(-\frac{V(\chi')-V(\chi)}{T}\right) & (V(\chi') > V(\chi)). \end{cases}$$

(2)

Now, for the calculation of the second fission, we need the potential energy surface of the deformed fragment in the two-dimensional configuration space $(\alpha, AL)$, where $AL$ denotes the mass number of the (future) light fragment in the second fission. We start from an initial configuration and let the samples move according to the Metropolis method. For the current position $(\alpha, AL)$, we take the neighbours as $(\alpha, AL\pm\Delta A)$ and $(\alpha\pm\Delta \alpha, AL)$. In the actual calculation, we take $\Delta A = 0.5$ and $\Delta \alpha = 0.0025$. For simplicity, we use the fixed temperature $T$ to calculate the step probability. We continue the random walk until the sample reaches either $\alpha = \alpha_{\text{MAX}} = 1.0$ or $\alpha = \alpha_{\text{MIN}} = 0.85$. Fission probability is calculated by dividing the number of samples that reach $\alpha_{\text{MAX}}$ by that of all samples. The number of samples ranges from $10^7$ to several $10^8$ depending on the fission probability.

![Figure 2. Potential energy surface of the deformed fragment: left panel is for Tb and right panel is for Pd.](https://example.com/figure2.png)

**3 Results and discussions**

In this work, we calculate the probability of sequential binary fission with respect to the first binary fission to estimate the probability of observing “ternary” fission in which we observe three fragments of significant mass. We choose two systems: a super-heavy nucleus $^{300}\text{Pd}$ and a heavy actinide nucleus $^{252}\text{Cf}$. According to the pre-scission model, the most probable mass separation is $A_1 = 136$ and $A_2 = 164$ (Tb, $Z = 65$) for the fission of $^{300}\text{Pd}$ and $A_1 = 132$ and $A_2 = 120$ (Pd, $Z = 46$) for $^{252}\text{Cf}$. In both cases, one fragment is found to be almost
spherical while the other fragment is fairly deformed as shown in Fig. 1. In order to estimate the probability of the second fission with the Metropolis method, we calculate the energy surface of the deformed fragment (\(^{164}\)Tb or \(^{120}\)Pd) by minimizing the energy with respect to \(\alpha_3\) and \(\alpha_4\) for each set of \((\alpha, A_L)\). Figure 2 shows the contour plot of the deformation energy in the two-dimensional configuration space \((\alpha, A_L)\) for \(^{164}\)Tb (left panel) and \(^{120}\)Pd (right panel). The point marked with a star corresponds to the most probable initial configuration which is obtained by refitting the shape obtained with the pre-scission model. The red rectangle denotes the area from where we take the points to start the Metropolis random walk.

3.1 Probability of the second fission with respect to the first binary fission

In this subsection, we will discuss the probability of the second fission. Table 1 shows the probability of the second fission with respect to the first binary fission for nine initial configurations around the most probable configuration for the case of a super-heavy nucleus \(^{300}\)\(^{120}\). The most probable initial configuration is \(\alpha = 0.905\) and \(A_L = 24\), as is shown by a star symbol in the left panel of Fig. 2. In the case of \(T = 1.0\) MeV, the fission probability is found to be in the order of \(10^{-4}\) and it is in the order of \(10^{-3}\) for \(T = 1.5\) MeV. Table 2 is for the case of \(^{252}\)Cf. The most probable initial configuration is \(\alpha_1 = 0.89\) and \(A_L = 26\) in this case. The probability of the second fission of \(^{252}\)Cf is found to be in the order of \(10^{-7}\) in the case of \(T = 1.0\) MeV and in the order of \(10^{-6}\) in the case of \(T = 1.5\) MeV. In all cases, the fission probability depends significantly on the initial position. Nevertheless, we can conclude that the fission probability will be larger by a few orders of magnitude in the case of super-heavy nuclei than in the case of actinide nuclei (Cf).

Table 1. Fission probability of the fragment \(^{164}\)Tb calculated for several initial configurations.

| \(^{164}\)Tb | \(T = 1.0\) MeV | \(T = 1.5\) MeV |
|---|---|---|
| \(A_L = 22\) | \(A_L = 24\) | \(A_L = 26\) | \(A_L = 22\) | \(A_L = 24\) | \(A_L = 26\) |
| \(\alpha = 0.910\) | \(1.2\times10^{-4}\) | \(4.8\times10^{-4}\) | \(9.4\times10^{-4}\) | \(1.3\times10^{-3}\) | \(3.2\times10^{-3}\) | \(5.4\times10^{-3}\) |
| \(\alpha = 0.905\) | \(5.5\times10^{-5}\) | \(2.3\times10^{-4}\) | \(5.1\times10^{-4}\) | \(7.5\times10^{-4}\) | \(1.9\times10^{-3}\) | \(3.4\times10^{-3}\) |
| \(\alpha = 0.900\) | \(2.4\times10^{-5}\) | \(1.0\times10^{-4}\) | \(2.8\times10^{-4}\) | \(4.2\times10^{-4}\) | \(1.1\times10^{-3}\) | \(2.2\times10^{-3}\) |

Table 2. Fission probability of the fragment \(^{120}\)Pd calculated for several initial configurations.

| \(^{120}\)Pd | \(T = 1.0\) MeV | \(T = 1.5\) MeV |
|---|---|---|
| \(A_L = 24\) | \(A_L = 26\) | \(A_L = 28\) | \(A_L = 24\) | \(A_L = 26\) | \(A_L = 28\) |
| \(\alpha = 0.895\) | \(6.0\times10^{-7}\) | \(9.9\times10^{-7}\) | \(2.2\times10^{-6}\) | \(5.4\times10^{-6}\) | \(8.0\times10^{-6}\) | \(1.2\times10^{-5}\) |
| \(\alpha = 0.890\) | \(2.8\times10^{-7}\) | \(6.2\times10^{-7}\) | \(1.1\times10^{-6}\) | \(3.2\times10^{-6}\) | \(4.9\times10^{-6}\) | \(7.6\times10^{-6}\) |
| \(\alpha = 0.885\) | \(1.6\times10^{-7}\) | \(2.9\times10^{-7}\) | \(5.9\times10^{-7}\) | \(1.9\times10^{-6}\) | \(3.1\times10^{-6}\) | \(4.7\times10^{-6}\) |

In order to understand what determines the probability of the second fission, we perform the calculation starting from many points in \((\alpha, A_L)\) space. The calculation area is marked by a red rectangular in Fig. 2. The results are plotted as contour maps in Figs. 3 and 4. We plot the common logarithm of the probability, namely \(\log_{10}(P_f(\alpha, A_L))\) where \(P_f(\alpha, A_L)\) is the probability of the second fission with the starting point being \((\alpha, A_L)\). Figure 3 is for \(^{164}\)Tb and Fig. 4 is for \(^{120}\)Pd. The left panels show the results for \(T = 1.0\) MeV and the right panels show those for \(T = 1.5\) MeV. Obviously, the fission probability is larger for \(T = 1.5\) MeV than that is for \(T = 1.0\) MeV. On the other hand, as is seen from the figures, the structure of the contour plot is the same for \(T = 1.0\) MeV and for \(T = 1.5\) MeV. The temperature
dependence comes from the statistical weight \( \exp(-\Delta V/T) \) at each step of the Metropolis method. The feature of the plot can be understood with the two factors: the statistical weight \( \exp(-(V_{\text{sad}} - V(\alpha, A_L))/T) \) and the distance between the initial and the saddle point. Here, \( V_{\text{sad}} \) denotes the potential energy at the saddle point. The saddle point lies around \((\alpha, A_L) = (0.98, 30)\) for \(^{164}\text{Tb}\) and \((0.99, 40)\) for \(^{120}\text{Pd}\). The fission probability depends on the distance, because it is a non-equilibrium diffusion process.

![Figure 3](image3.png)

**Figure 3.** Fission probability \( \log_{10} P_f \) of the deformed fragment \(^{164}\text{Tb}\) as a function of the initial configuration \((\alpha, A_L)\). Left panel is for \(T = 1.0\) MeV and right panel is for \(T = 1.5\) MeV.

![Figure 4](image4.png)

**Figure 4.** Fission probability \( \log_{10} P_f \) of the deformed fragment \(^{120}\text{Pd}\) as a function of the initial configuration \((\alpha, A_L)\). Left panel is for \(T = 1.0\) MeV and right panel is for \(T = 1.5\) MeV.

When we start the calculation from the points close to the most probable initial configuration, the mass of the light fragment of the second fission (at \(\alpha = 1.0\)) is found to be close to that at the saddle point. Thus, we predict the most probable mass divisions in the ternary fission to be 136+30+134 for \(^{300}\text{120}\) and 132+40+80 for \(^{252}\text{Cf}\). Hence, in the case of super-heavy nuclei, the heavy outer fragments have almost equal masses.
3.2 The Metropolis method and the Langevin approach

The Metropolis method was introduced as a means for exploring available configurations according the associated statistical weight. In order to demonstrate the applicability of the Metropolis method in calculating a non-equilibrium process, we solve a simple one-dimensional over barrier process with three methods: the Metropolis method, the reduced Langevin equation in q-space, and the Langevin equation in (p, q)-space. The Langevin equation in (p, q)-space is written as

\[
\frac{dp}{dt} = -\frac{dV}{dq} - \frac{\gamma}{m} p + R(t),
\]
\[
\frac{dq}{dt} = \frac{p}{m}.
\]

(3)

And the reduced Langevin equation in q-space is written as

\[
\frac{dq}{dt} = \frac{1}{\gamma} \left(-\frac{dV}{dq} + R(t)\right).
\]

(4)

Here \( m \) denotes the inertia mass, \( \gamma \) the friction coefficient, and \( V \) the potential. \( R(t) \) denotes the random force which has the following properties;

\[
\langle R(t) \rangle = 0,
\]
\[
\langle R(t) R(t') \rangle = 2\gamma T \delta(t - t'),
\]

(5)

where \( \langle \rangle \) denotes the ensemble average. We assume a constant mass and friction. The potential energy is given as a connection of parabola, inverse parabola, and parabola shape which is drawn in a small panel in Fig. 5. We put a sample particle at \( x = 0.5 \) and calculate the development using the three methods. In the case of the Langevin equation in (p, q)-space, we consider two cases: zero initial momentum case (p=0) and thermal initial momentum distribution case (p=thermal). We set the mass \( m = 1 \) and the friction coefficient \( \gamma = 5 \). We assume a fixed temperature of \( T = 1 \) in this calculation.

![Figure 5. Comparison of the three methods to calculate the diffusion process over a barrier. Black line denotes the result with the Metropolis method, red dashed line that of the reduced Langevin equation, blue line that of the Langevin equation with zero initial momentum, and green line denotes that of the Langevin equation with thermal initial momentum distribution.](image)

In Fig. 5, we plot the distribution at the moment when we observe the largest probability in the left potential pocket. In the left panel, we show the probability distribution in coordinate space and in the right panel, we magnify it so that we can see the difference in the left pocket region. It should be noted that if we continue the calculation for a long time, all three methods
give the same distribution and the thermal equilibrium is accomplished. One can see in the left panel of Fig. 5 that the big peak in the right potential pocket is almost the same in all cases. On the other hand, in the left potential pocket, as you can see in the right panel of Fig. 5, the height of the peak differs slightly among the methods. The Metropolis method gives the largest peak and the Langevin equation with zero initial momentum gives the smallest peak. The reduced Langevin equation and the Langevin equation with thermal initial momentum distribution give almost the same result. In the previous calculation of the fission probability of the deformed nuclei, the left peak corresponds to the “ternary” (sequential binary) fission process and the right peak corresponds to the normal binary fission process. From the results, we can say that the use of the Metropolis method should be justified.

4 Summary

It is predicted that one of the fragment is extremely deformed in the binary fission of super-heavy nuclei. We expect that the deformed fragment may undergo the second fission thus producing three fragments. Starting from the pre-scission shape of the fragment of the first binary fission, we calculate the probability of the second fission by using the Metropolis method. Results are given for the fission of a super-heavy nucleus $^{300}120$ and a heavy actinide nucleus $^{252}Cf$. The probability of the second fission is in the order of $10^{-4}$ for $^{300}120$ and $10^{-7}$ for $^{252}Cf$, when we assume the constant temperature of $T = 1.0$ MeV. We also show that the ternary mass division is almost symmetric for the case of super-heavy nuclei. It should be mentioned that this work is just a simple, preliminary calculation of a new phenomenon that could be observed in the fission of super-heavy nuclei. More detailed and quantitative study is definitely necessary.

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