Dynamical calculation for fusion–fission probability in superheavy mass region, where mass symmetric fission events originate

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Received 24 May 2004; received in revised form 9 July 2004; accepted 10 August 2004
Available online 28 August 2004

Abstract

We discuss dynamically the fusion–fission mechanism in the superheavy mass region. By analyzing the mass distribution of fission fragments, we distinguish between the fusion–fission process and the quasi-fission process. We investigate these two processes using the fluctuation–dissipation model. The three-dimensional Langevin equation is employed. We classify the dynamical process by analyzing the trajectory in the nuclear deformation space. In the superheavy mass region, we found that 90–99% of mass symmetric fission events come from the quasi-fission process, in which the system hardly reaches the spherical region. The fusion cross section is also estimated precisely. © 2004 Elsevier B.V. All rights reserved.

Keywords: Superheavy elements; Fluctuation–dissipation dynamics; Fusion–fission process; Quasi-fission process

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doi:10.1016/j.nuclphysa.2004.08.009
1. Introduction

The existence of the “Island of Stability” in the superheavy mass region has been predicted using the nuclear shell model [1]. Recent experiments on the synthesis of superheavy elements have been focused on the investigation of this region and seem to support this hypothesis. Recently, the production of new superheavy elements has been reported by FLNR ($Z = 113, 114, 115, 116$ and $118$) using hot fusion reaction [2–4] and GSI ($Z = 110, 111$ and $112$) using cold fusion reaction [5]. Using the facility in RIKEN, nuclei with $Z = 110$ and $111$ have also been produced by cold fusion reaction and the excitation function of evaporation residue cross section has been measured with high accuracy [6].

Many theoretical studies on the synthesis of superheavy elements have been published recently and the evaporation residue cross section corresponding to the above experiments has been estimated [7–10]. In the theoretical calculation, the evaporation residue cross section is obtained as the product of the fusion probability forming a compound nucleus and its survival probability in the competition with the fission process. The value of the product leads to a pico-barn order and is very small compared with the quasi-fission cross section. Inevitably, a substantial uncertainty is involved: the uncertainty of the potential landscape near the contact configuration of colliding nuclei affects the estimation of fusion probability, and the uncertainty of the fission barrier height of the compound nucleus and the level density parameter used in the statistical calculation affects the estimation of survival probability [11]. In this sense, a theoretical framework of estimating the evaporation residue cross section of such rare events has not been realized yet. In order to estimate the evaporation residue cross section correctly, first we must develop a more accurate estimation of fusion probability. Here, we focus our attention on the precise estimation of fusion probability.

In heavy nucleus collision experiments [12], fusion–fission cross sections are derived from counting mass symmetric fission events. Here, an important question arises, that is, whether all of the mass symmetric fission fragments originate from compound nuclei. Is there any possibility of the composite nuclei splitting into symmetric fragments without forming a compound nucleus?

The main purpose of the present paper is to clarify the origin of the mass symmetric fission fragments. We define the difference between fusion–fission process and the quasi-fission process, and estimate the ratio of the fusion–fission probability to the quasi-fission probability. Finally we would like to obtain the fusion cross section.

To investigate the fusion–fission mechanism, trajectory calculation has been performed under friction force [13–15]. In the superheavy mass region, the mean trajectory cannot overcome the extra potential barrier in the deformation space after contact by only adding the extra bombardment energy. This is due to the substantial energy conversion from the kinetic part to the internal degree of freedom of the composite nucleus. Therefore, to estimate fusion probability, it is necessary to apply fluctuation–dissipation dynamics with the Fokker–Planck equation or with the Langevin equation [16–19].

We investigate whether the trajectory in the deformation space enters the compound nucleus region, and estimate the fusion probability. At the same time, we estimate which part of the mass symmetric events belongs to the fusion–fission cross section.
In Section 2, we explain our framework for the study and the model used. We discuss the mass distribution of fission fragments in Section 3. By analyzing the Langevin trajectories in the three-dimensional deformation space, we discuss the fusion–fission mechanism. We present the theoretical results for the excitation function of the fusion cross section in the reactions $^{48}\text{Ca} + ^{208}\text{Pb}$ and $^{48}\text{Ca} + ^{244}\text{Pu}$. We present a summary and further discussion in Section 4.

2. Model

The evaporation residue cross section $\sigma_{\text{ER}}$ is estimated as

$$\sigma_{\text{ER}} = \frac{\pi \hbar^2}{2\mu_0 E_{\text{cm}}} \sum_{l=0}^{\infty} (2l + 1) T_l(E_{\text{cm}}, l) P_{\text{CN}}(E^*, l) W(E^*, l),$$  \hspace{1cm} (1)

where $\mu_0$ denotes the reduced mass in the entrance channel. $E_{\text{cm}}$ and $E^*$ denote the incident energy in the center-of-mass frame and the excitation energy of the compound nucleus, respectively. $E^*$ is given as $E^* = E_{\text{cm}} - Q$ with $Q$ denoting the $Q$-value of the reaction. $T_l(E_{\text{cm}}, l)$ is the capture probability of the $l$th partial wave, which is calculated with the empirical coupled channel model suggested by Zagrebaev [20]. $P_{\text{CN}}(E^*, l)$ is the probability of forming a compound nucleus in competition with quasi-fission events. $W(E^*, l)$ denotes the survival probability of compound nuclei during deexcitation, which is calculated using a statistical model [21,22].

Our final goal is to accurately estimate $\sigma_{\text{ER}}$. However, since there are many unknown parameters at each stage and the probability is very sensitive to these parameters, here we focus our attention on the precise estimation of the fusion probability $T_l P_{\text{CN}}$ using a dynamical calculation.

To calculate $P_{\text{CN}}$, we use the fluctuation–dissipation model and employ the Langevin equation. We adopt the three-dimensional nuclear deformation space given by two-center parameterization [23,24]. The neck parameter $\epsilon$ is fixed to 1.0 in the present calculation so as to retain the shape of the contact-like configuration more realistically for two-nucleus collision. The three collective parameters involved in the Langevin equation are as follows: $z_0$ (distance between two potential centers), $\delta$ (deformation of fragments) and $\alpha$ (mass asymmetry of the colliding nuclei); $\alpha = (A_1 - A_2)/(A_1 + A_2)$, where $A_1$ and $A_2$ denote the mass numbers of the target and projectile, respectively.

The multi-dimensional Langevin equation is given as

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

$$\frac{d p_i}{dt} = -\frac{\partial V}{\partial q_i} - \frac{1}{2\gamma_{ij}} (m^{-1})_{jk} p_j p_k - \gamma_{ij} (m^{-1})_{jk} p_k + g_{ij} R_j(t),$$  \hspace{1cm} (2)

where summation over repeated indices is assumed. $q_i$ denotes the deformation coordinate which is specified by $z_0$. $\delta$ and $\alpha$, $p_i$ is the conjugate momentum to $q_i$. $V$ is the potential energy, and $m_{ij}$ and $\gamma_{ij}$ are the shape-dependent collective inertia parameters and dissipation tensors, respectively. A hydrodynamical inertia tensor is adopted by Werner–Wheeler.
approximation for the velocity field, and the wall-and-window one-body dissipation is adopted for the dissipation tensor [25–27]. The normalized random force $R_i(t)$ is assumed to be a white noise, i.e., $\langle R_i(t) \rangle = 0$ and $\langle R_i(t_1) R_j(t_2) \rangle = 2\delta_{ij}\delta(t_1 - t_2)$. The strength of random force $g_{ij}$ is given by $\gamma_{ij} T = \sum_k g_{ij} g_{jk}$, where $T$ is the temperature of the compound nucleus calculated from the intrinsic energy of the composite system as $E_{\text{int}} = a T^2$ with $a$ denoting the level density parameter. The temperature-dependent potential energy is defined as

$$V(q, l, T) = V_{\text{DM}}(q) + \frac{\hbar^2 l(l + 1)}{2I(q)} + V_{\text{shell}}(q) \Phi(T),$$

$$V_{\text{DM}}(q) = E_S(q) + E_C(q),$$

where $I(q)$ is the moment of inertia of a rigid body at deformation $q$. $V_{\text{shell}}$ is the shell correction energy at $T = 0$, and $V_{\text{DM}}$ is the potential energy of the finite range liquid drop model. $E_S$ and $E_C$ denote a generalized surface energy [28] and Coulomb energy, respectively. The centrifugal energy arising from the angular momentum $l$ of the rigid body is also considered. The temperature-dependent factor $\Phi$ is parameterized as $\Phi = \exp(-a T^2/E_d)$ following the work of Ignatyuk et al. [29]. The shell dumping energy $E_d$ is chosen as 20 MeV. The intrinsic energy of the composite system $E_{\text{int}}$ is calculated for each trajectory as

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

At $t = 0$, each trajectory starts from the touching point which is defined as $R_{\text{touch}} = R_p + R_t$, where $R_p$ and $R_t$ are the radii of projectile and target, respectively.

### 3. Results

We analyze the mass distribution of fission fragments following heavy nucleus collision [12] and classify them into two components, namely, those coming from the fusion–fission (FF) process and quasi-fission (QF) process. The mechanisms of FF and QF are clarified by analyzing the Langevin trajectory on the three-dimensional potential energy surface.

#### 3.1. Mass distribution of fission fragments and fusion cross section of $^{48}\text{Ca} + ^{208}\text{Pb}$

For the purpose of calibrating our model calculation, first we analyze the fusion cross section of the $^{48}\text{Ca} + ^{208}\text{Pb}$ reaction in which the fusion–fission process is dominant. Fig. 1 shows the potential energy surface of the liquid drop model with the shell correction energy for $^{258}\text{No}$ on the $Z-\alpha$ plane, in the case of $\delta = 0$ and $l = 0$. This potential energy surface is calculated using the two-center shell model code [30,31]. To save computational time, we use scaling and employ the coordinate $z$. The coordinate $z$ is defined as $z = z_0/(R_{\text{CN}}B)$, where $z_0$ and $R_{\text{CN}}$ denote the distance between two potential centers and the radius of the spherical compound nucleus, respectively. The parameter $B$ is defined as $B = (3 + \delta)/(3 - 2\delta)$. In Fig. 1, the position at $z = \alpha = 0$ corresponds to a spherical compound nucleus. Due to the shell structure of the nucleus, we can see a valley on the potential energy surface.
near Pb and Sn fragments. The black arrow indicates the inject direction and the touching point is marked by +. We start to calculate the Langevin equation at this point.

In this model, we have an unknown parameter, that is, how much of the relative kinetic energy of the colliding system dissipates into intrinsic energy during the approaching process. It is an open question in the superheavy mass region.

Using this model, we have discussed this matter in Ref. [32]. As an initial condition of the Langevin calculation, the intrinsic energy and relative kinetic energy of nuclei at touching point are very important. However, our attention is focused to the problem of the clarification of the difference between fusion–fission process and quasi-fission process. For this purpose, i.e., as the first preliminary calculation which is able to reveal difference of the reaction process clearly, we assume that the kinetic energy does not dissipate during the approaching process, as the simple and extreme example. We would like to estimate mainly the ratio of fusion–fission probability to quasi-fission probability in our model.

So, we start to calculate the Langevin equation at the touching point which is located at $z = 1.56, \delta = 0.0, \alpha = 0.62$. All trajectory starts at this point. The initial velocity is directed in only the $z$ direction, that is, both components of the initial velocities along the $\delta$ and $\alpha$ direction are assumed to be zero.
To estimate fusion probability, the definition of the fusion area in the deformation space is very important. It is reasonable to define the fusion area around the pocket near the ground state in the deformation space. Here, we define the fusion area (fusion box) as the inside of the fission saddle point in the system, which is \( |z < 0.8, \delta < 0.3, |\alpha| < 0.3 \).

To reach the spherical region, the trajectories have to overcome the saddle point (S.P.) located at \((z, \alpha) = (1.5, 0.62)\) which is marked by \(\otimes\), or the Businaro–Gallone (B.G.) point [33], which is marked by \(\times\) in Fig. 1. The di-nucleus concept [7] corresponds to the idea that the trajectory overcomes the B.G. point. However, the difference in potential energy between the touching point and the B.G. point is approximately 13 MeV. On the other hand, the difference in potential energy between the touching point and the saddle point is very small due to the shell structures of \(^{208}\text{Pb}\) and \(^{48}\text{Ca}\). We can see the valley leading to the spherical region passing the saddle point. Therefore, almost all the trajectories pass the saddle point and reach the spherical region. Finally the trajectories that come into the spherical region enter the fission region through the mass symmetric fission valley. This situation is shown in Fig. 1 by the arrow located near \(\alpha = 0\).

The mass distribution of the fission fragments in the reaction \(^{48}\text{Ca} + ^{208}\text{Pb}\) at the incident energy corresponding to the excitation energy of the compound nucleus \(E^* = 40\) MeV is shown in Fig. 2(a). Here, in the fusion–fission process the range of the nuclear tem-

![Fig. 2. (a) Mass distribution of fission fragments in reaction \(^{48}\text{Ca} + ^{208}\text{Pb}\) at energy corresponding to excitation energy of compound nucleus \(E^* = 40\) MeV. (b) The cross section given by the calculation and experimental data are denoted by the solid line and open symbols, respectively.](image)
perature is approximately $T = 0.5–1.2$ MeV. The calculations and experimental data are denoted by the black and gray lines, respectively. In this system, the fusion–fission reaction is dominant, so that the yield of fission fragments near mass symmetry is significant, whose mean path is indicated by the two white arrows in Fig. 1. In Fig. 2(a), the calculation results of the mass distribution of fission fragments show good agreement with the experimental data in the mass symmetric region.

The excitation function of the fusion–fission cross section $\sigma_{\text{CN}}$ for $^{48}\text{Ca} + ^{208}\text{Pb}$ is shown in Fig. 2(b). Open triangles and squares denote experimental data [34–36]. The solid line denotes our calculation. The calculation gives a good prediction of the general trend of the experimental data above the Coulomb barrier energy region [37].

### 3.2. Mass distribution of fission fragments and fusion cross section of $^{48}\text{Ca} + ^{244}\text{Pu}$

For $^{48}\text{Ca} + ^{244}\text{Pu}$, the situation changes. The potential energy surface of the liquid drop model with shell correction energy for $^{292}114$ is shown in Fig. 3. The touching point and the saddle points are marked by + and $\otimes$, respectively. In this case, most of the trajectories enter the fission region passing through the QF path, that is to say, the QF process is dominant. One of the fission fragments distributes around Pb, which is marked in Fig. 3. In this calculation, we assume that both the shapes of the target and the projectile are spherical at

![Fig. 3. Potential energy surface of liquid drop mode with shell correction energy for $^{292}114$. The black arrow shows the injection point of the reaction $^{48}\text{Ca} + ^{244}\text{Pu}$. Symbols are given in the text.](image-url)

the touching point of the colliding system, even though $^{244}$Pu is a deformed nucleus. We define the fusion box as the inside of the fission saddle point, $\{ z < 0.6, \delta < 0.2, |\alpha| < 0.25 \}$. On the potential energy surface, due to the shell structure we can see the valley near the Pb and Sn nuclei, which correspond to $\alpha \sim 0.4$ and $\alpha \sim 0.12$, respectively, as shown in Fig. 3.

In the energy region corresponding to the excitation energy $E^* = 33-40$ MeV, the mean trajectory moves along the QF path, which is indicated by QF in Fig. 3. As a result, fission fragments such as Pb are produced. Actually, we can find Pb-like fragments in the experimental data at an excitation energy of 30–40 MeV [12].

Fig. 4(a) shows the mass distribution of fission fragments in the reaction $^{48}$Ca + $^{244}$Pu at $E^* = 37$ MeV, which corresponds to the nuclear temperature $T = 1.1$ MeV. The black and gray lines denote the calculation and experimental results, respectively. In this case, the mass asymmetric fission event is dominant. Our calculation reproduces the main tendency of the experimental data in the region of $\alpha < 0.4$.

Next we discuss the fusion cross section. The experiments on the fission of superheavy nuclei in the reaction $^{48}$Ca + $^{244}$Pu have been carried out by Dubna group and they have reported the fusion–fission cross section of compound nuclei, which are derived from mass symmetric fission fragments ($A/2 \pm 20$) [12]. In this superheavy mass region, the evaporation residue cross section is expected to be very small. The fusion cross section in our present calculation is almost equal to the fusion–fission cross section. Here, the important question to be considered is whether all of the mass symmetric fission fragments originate from the compound nuclei. In the experiments, the mass symmetric fission fragments were actually detected; however, two possibilities can exist as to the origin of the fission fragments. One possibility is that the mass symmetric fission fragments come from the

![Image](https://via.placeholder.com/150)

Fig. 4. (a) Mass distribution of fission fragments in the reaction $^{48}$Ca + $^{244}$Pu at $E^* = 37$ MeV. (b) Cross sections for calculation and experiment. Lines and symbols are given in the text.
compound nuclei and the other possibility is that they come from the quasi-fission, that is, they do not pass through the fusion box. In Fig. 3, these two paths are presented. We investigate these two possibilities using a three-dimensional trajectory calculation with the Langevin equation.

Fig. 5(a) shows three samples of the trajectories which are projected on the $z-\alpha$ plane ($\delta = 0$), for the excitation energy $E^* = 33$ MeV. Here, in the fusion–fission process, the range of the nuclear temperature is approximately from $T = 0.3$ MeV (near the touching point) to 1.0 MeV (near the compound nucleus). The touching point of the system

![Diagram](image)

Fig. 5. Three samples of trajectory which are projected on $z-\alpha$ plane ($\delta = 0$) (a) and $z-\delta$ plane ($\alpha = 0$) (b), for incident energy corresponding to $E^* = 33$ MeV in reaction $^{48}$Ca + $^{244}$Pu. The contact point is marked by +.
is \((z, \alpha) = (1.54, 0.67)\) marked by (+). The contour denotes the potential energy surface with a step of 2 MeV. Most of the trajectories enter the fission region. We can see that some trajectories enter the mass symmetric fission region around \(\alpha = 0\). These trajectories occupy 6.8% of all trajectories. It is significant that such trajectories do not always come from the fusion box.

Fig. 5(b) shows the same trajectories, which are projected on the \(z-\delta\) plane \((\alpha = 0)\). In Fig. 5(b), the touching point of the system is \((z, \delta) = (1.54, 0.0)\) marked by (+). In the heavy-mass system, due to the large Coulomb repulsive force, the potential energy surface has a very steep slope in the positive direction of \(z\) and \(\delta\). Therefore, even if trajectories overcome the fusion barrier, almost all the trajectories follow the \(+\delta\) direction and can hardly reach the spherical region. In Fig. 5(b), it is shown that the trajectories follow the \(+\delta\) direction, and they do not contribute to the formation of the compound nucleus. This means that the formation of the compound nucleus is extremely inhibited in the superheavy mass region. We can say that fusion is hindered due to the flow of the trajectory to the \(\delta\) degree of freedom.

Due to this hindrance, almost all the trajectories that enter the mass symmetric fission region do not pass through the fusion box. We call such trajectories the “deep quasi-fission process” (DQF) [38,39]. In fact, for \(E^* = 33\) MeV, only 0.08% of all trajectories can overcome the barrier and enter the spherical region or the fusion box. This very small rate forming the compound nucleus is approximately 1.2% of the trajectories that go to the mass symmetric fragments, which is estimated as 6.8% of all trajectories.

The excitation functions of the cross sections are shown in Fig. 4(b). The open and closed diamonds denote the experimental capture cross section \(\sigma_{\text{cap}}\) and the experimental cross section \(\sigma_{A/2 \pm 20}\) which are derived from the yield of the mass symmetric fission fragments whose mass number is greater than \(A/2 - 20\) and less than \(A/2 + 20\), respectively [12]. The calculated \(\sigma_{A/2 \pm 20}\) is denoted by the solid line. We can see good agreement between the experimental data and our calculations. The calculated fusion cross section \(\sigma_{\text{CN}}\) is denoted by the dashed line. This cross section \(\sigma_{\text{CN}}\) is derived from the trajectory crossing the three-dimensional fusion box. The fusion cross section \(\sigma_{\text{CN}}\) is one or two orders of magnitude smaller than the cross section \(\sigma_{A/2 \pm 20}\). We can see that the main part of the experimental cross section \(\sigma_{A/2 \pm 20}\) is the deep quasi-fission event which does not pass through the fusion box. Such classification is very important in the estimation of the evaporation residue cross section.

4. Summary

The fusion–fission process in the synthesis of superheavy elements was studied based on fluctuation–dissipation dynamics. We calculated the fusion cross section and compared with the experimental data. We took into account the competition between the fusion to form the compound nucleus and the quasi-fission followed by the contact stage.

Using the three-dimensional Langevin calculation, we found that the quasi-fission process contributes substantially to the yield of mass symmetric fission fragments. In this process, the trajectory on the three-dimensional potential energy surface does not pass through the fusion box in the deformation space. We call such trajectories the “deep quasi-
fission process”. When we calculate the trajectories for the fusion–fission process, we have to pay attention to the dynamical deformation of fragments. In the superheavy mass region, the degree of freedom of the fragment deformation is very important because many trajectories move in the direction of the large deformation of the fragment in the present two-center shell model shape parameterization. In our calculation, we would like to emphasize that one- or two-dimensional calculation is not sufficient to describe whether the trajectory passes the fusion box.

A three- or multi-dimensional model should be used to obtain a more reliable fusion–fission probability. The present calculation shows that the fusion cross section $\sigma_{\text{CN}}$ is one or two orders of magnitude smaller than the cross section of $\sigma_{A/2 \pm 20}$ which is derived from the experiments, that is, the main part of the cross section $\sigma_{A/2 \pm 20}$ is the deep quasi-fission process.

In the next step, we should consider a nuclear structure effect more precisely for fusion process. In the present calculation, while we take into account the shell correction energy for potential energy surface, we use transport coefficients which are calculated by the macroscopic model. In order to discuss the fusion–fission process more precisely and consistently, we should apply the transport coefficients of the microscopic model, for example, linear response theory [40–42]. The friction tensor calculated by the microscopic model is smaller than that by the macroscopic model. The one-dimensional calculation with the transport coefficients of microscopic model has been reported in the Ref. [43]. For the future study, we should investigate the influence of the transport coefficients of the microscopic model using the three-dimensional Langevin equation.

Acknowledgements

The authors are grateful to Prof. Yu.Ts. Oganessian, Prof. M.G. Itkis, Prof. V.I. Zagrebaev and Prof. F. Hanappe for their helpful suggestions and valuable discussion throughout the present work. The authors also wish to thank Prof. T. Wada, Dr. T. Tokuda and Mr. K. Okazaki who developed the calculation code for the three-dimensional Langevin equation. The authors would like to thank Dr. S. Yamaji and his collaborators who developed the calculation code for potential energy with a two-center parameterization.

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