REACTION MECHANISMS FOR SYNTHESIS OF SUPERHEAVY ELEMENTS*

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(Received November 18, 2002)

A theoretical model is presented for fusion mechanisms in massive systems, where the fusion process is divided into two steps; an approaching phase up to the contact of two incident ions and shape evolutions from the amalgamated configuration to the spherical shape. Results of the first step provide initial values for the second step. A qualitative explanation and a simple expression of the extra-push energy are obtained with an inverted parabola approximating the conditional saddle. Results obtained with more realistic two-dimensional model for shape evolutions are compared very well with the available data for ^{48}Ca + actinide target systems. Remarks are given for synthesis of the superheavy elements.

PACS numbers: 24.60.–k, 25.60.Pj, 25.70.–z, 25.70.Jj

* Presented at the XXXVII Zakopane School of Physics "Trends in Nuclear Physics", Zakopane, Poland, September 3–10, 2002.

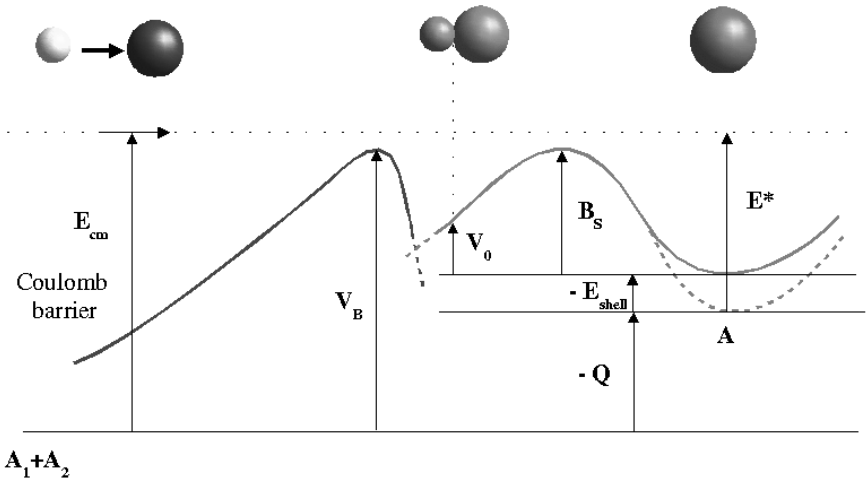
1. Introduction

According to the compound nucleus theory [1], residue cross sections are given by the product of fusion probability and survival probability. The latter can be calculated by the theory of statistical decays. They depend on detailed properties of relevant nuclei, especially their masses, or their shell correction energies which essentially determine fission barriers, but are unknown and thereby give rise to substantial ambiguities in practice. But the theory is well established. On the other hand, for the former, *i.e.*, for fusion probability, there is no reliable theoretical framework available for massive systems [2] where so-called fusion hindrance is known to exist experimentally [3]. Recently the present authors have proposed a two-step model [4] in order to describe the whole process from the encounter of incident ions up to the formation of the spherical compound nucleus. It consists of two-body collision process up to the contact of the incident ions after overcoming the Coulomb barrier under friction in action and of shape evolutions to the spherical configuration, starting from amalgamated configurations made by the sticking of the incident ions. The latter process is not necessary in usual heavy-ion fusion reactions, but is indispensable in massive systems, *i.e.*, for the synthesis of the superheavy elements (SHE), because the contact point is still outside of a conditional saddle or a ridgeline, as is shown in Fig. 1, while in usual systems it locates inside the conditional saddle point. Since the amalgamated system is expected to be excited due to heat-up processes during two-body collisions, subsequent collective shape motions of the system are under a strong dissipation stemming from frequent interactions with the nucleons at a finite temperature. Thus, both processes are described by dissipation-fluctuation dynamics, *i.e.*, by Langevin equations [5].

An important thing that remains is how to connect them. Naturally, two processes are in succession, so results of the first step give initial conditions of the second step. In this sense, analyses of results of the first step, *i.e.*, of the contact dynamics in the collisions of the incident ions are necessary for the purpose of obtaining initial conditions for subsequent shape evolutions as well as of obtaining a sticking probability $P_{\rm stick}$. The present connection method can be called to be "statistical", neither adiabatic, nor diabatic [6], because the first step results in distributions of physical quantities, for example, in a Gaussian distribution of the radial momentum as will be seen below. With them as initial values, shape evolutions of the amalgamated system are solved to give a formation probability of the spherical compound nucleus $P_{\rm form}$. Thus, the fusion probability $P_{\rm fusion}$ is given by a product of the two factors

$$P_{\rm fusion}(E_{\rm c.m.}) = P_{\rm stick}(E_{\rm c.m.})\, P_{\rm form}(E_{\rm c.m.})\,. \tag{1}$$

$$P^J_{stick}(E_{cm}) \times P^J_{form}(E_{cm}) = P^J_{fusion}(E_{cm})$$



$$\begin{aligned} E* &= E_{cm} + Q \\ &= V_0 - E_{Shell} + \varepsilon_0 + k_0 \end{aligned}$$

Fig. 1. Coulomb barrier, sticking configuration, and conditional saddle are shown schematically for massive systems, which illustrates a necessity of two-step treatment for fusion.

For a realization of the above theoretical framework, we employ the surface friction model (SFM) [7] for the first step of two-body collision processes and the one-body model (OBM) [8] of friction, *i.e.*, the one-body wall-and-window formula for the second step of shape evolutions.

In the next section, we describe the second step first, and then in Section 3 we describe the first step, *i.e.*, so-called SFM and discuss characteristic features of the results. Of course, SFM could be improved, or re-adjusted so as to be suitable in barrier energy region, because it is proved to be successful in Deep-Inelastic Collisions (DIC), *i.e.*, in much above the Coulomb barrier. But for the moment, we take precisely the original version. In Section 4, the method of connecting the first step to the second step is discussed briefly. At last, examples of calculations of fusion cross sections are given for ^{48}Ca + actinides target systems in Section 5.

2. Shape evolution from the contact to the spherical shape

Firstly, we discuss shape evolutions starting from the contact point toward the spherical shape by overcoming the conditional saddle point or toward re-separation under a conservative driving force which is approximated by an inverted parabola. The Langevin equation, thus, is written as follows:

$$\frac{d}{dt}\begin{pmatrix} q \\ p \end{pmatrix} = \begin{pmatrix} 0 & 1/m \\ m\omega^2 & -\beta \end{pmatrix}\begin{pmatrix} q \\ p \end{pmatrix} + \begin{pmatrix} 0 \\ R \end{pmatrix}, \tag{2}$$

where m denotes the inertia mass of the motion, ω the curvature of the potential energy surface at the saddle point, and β the so-called reduced friction, *i.e.*, the friction γ divided by the inertia mass. These parameters could be calculated by LDM and OBM with Two-Center Parameterization (TCP) in case of quantitative discussions and comparisons with experiments. R is an inhomogeneous term represents the random force associated with the friction γ, so is assumed to be Gaussian and to satisfy the dissipation-fluctuation theorem

$$\begin{aligned} \langle R(t) \rangle &= 0, \\ \langle R(t) R(t') \rangle &= 2\,\gamma\,T\,\delta(t-t'), \end{aligned} \tag{3}$$

where $\langle\ \rangle$ denotes an average over all the possible realizations of the random force and T specifies the temperature of the heat bath of the nucleons. Since Eq. (2) is an inhomogeneous linear equation, we can write down a general solution as a function of initial values q_0 and p_0. (The origin of the coordinate q is taken to be the top of the barrier, and q_0 and p_0 are taken to be negative and positive, respectively.) Then, a distribution function of the system in the phase space is calculated for any later time t in the following way with the solution

$$W(q,p,t,T;q_0,p_0) = \langle \delta\left(q - \langle q(t)\rangle\right)\ \delta\left(p - \langle p(t)\rangle\right)\rangle_{\{R\}}, \tag{4}$$

where an average of the functional over R, $\langle\ \rangle_{\{R\}}$ can be performed generally by the path integral method.

Then, we obtain an expression for a probability for the system to pass over the saddle point by integrating over the whole p-space and over the half q-space, *i.e.*, over the other side of the saddle point. The probability is now given by an error function

$$F(t,T;q_0,p_0) = \int_0^\infty \frac{dq}{\sqrt{2\pi}}\frac{1}{\sigma(t)}\exp\left[\frac{\left(q - \langle q(t)\rangle\right)^2}{2\,\sigma(t)^2}\right]$$

$$
\begin{aligned}
&= \frac{1}{2}\mathrm{erfc}\left[-\frac{\langle q(t)\rangle}{\sqrt{2}\sigma(t)}\right], \\
\langle q(t)\rangle &= A(t)q_0 + B(t)p_0, \\
\sigma(t)^2 &= \langle q^2\rangle - \langle q(t)\rangle^2, \qquad (5)
\end{aligned}
$$

where $A(t)$ and $B(t)$ are given by the function of t with the parameters of γ and ω, precise expressions of which are given in Ref. [9]. The formation probability is obtained by taking the limit $t \to \infty$. Thus, the final probability is given by the error function with the following argument

$$
-\frac{\langle q(t)\rangle}{\sqrt{2}\sigma(t)} \;\to\; \sqrt{\frac{\sqrt{x^2+1}+x}{2x}}\left[\sqrt{\frac{B}{T}} - \frac{1}{\sqrt{x^2+1}+x}\sqrt{\frac{K}{T}}\right], \qquad (6)
$$

where the parameter x denotes $\beta/2\omega$. And K and B denote the initial kinetic and potential energies, $p_0^2/2m$ and $m\omega^2 q_0^2/2$, respectively. Now it is apparent that in order for the probability to be $1/2$ like transmission coefficient in quantum mechanics, the argument of the error function should be equal to zero, then $K^{1/2} = \left(\sqrt{x^2+1}+x\right)^2 B = B_{\mathrm{eff}}$, where B_{eff} can be interpreted as an effective barrier height required by the dissipative dynamics. The formula indicates that even in the cases with a small saddle point height B of LDM, the effective barrier height is much higher, for example, about 10 times of it if we take OBM for the friction. Therefore, this provides us with a simple dynamical explanation of the fusion hindrance by an analytic expression, assuming the schematic parabolic potential which is not so far from the reality. Now, we need to find the initial values q_0 and p_0, which is obtained in the next section.

3. Contact dynamics of massive systems

As is usual in heavy-ion collisions, we have to take into account the Coulomb barrier of the entrance channel. A special aspect in massive systems is that a friction between the incident ions is expected to be active around or even outside the barrier top. This is conceivable from measured excitation functions for so-called capture cross section, where limiting orbital angular momenta for the process are known to be smaller than grazing angular momenta [10]. Actually, SFM shows that the form factor of the friction stretches outside the barrier top position in massive systems. In Fig. 2, an example of the potential and the friction form factor is given as functions of the relative distance, calculated with SFM, exactly with their original parameter values which were fixed by the analyses of DIC. Since it

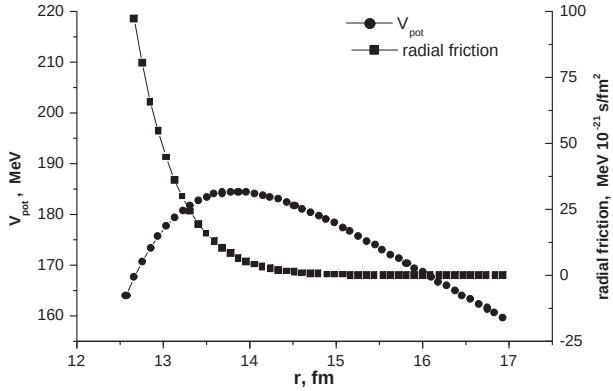


Fig. 2. The radial potential for s-wave and the radial form factor of the friction are shown for ^{48}Ca+^{244}Pu system, calculated with SFM.

is well known that dissipation is always associated with fluctuation, an associated fluctuation to the dissipation is to be introduced, so SFM equation is extended [11] as follows:

$$\begin{aligned} \frac{dp}{dt} &= -\frac{dV}{dt} - \frac{K_r}{\mu}\, p + \theta_1\, \omega_1\,, \\ \frac{dL}{dt} &= -\frac{K_\phi}{\mu}\left(L - \frac{5}{7}L_0\right) + \theta_2\, \omega_2\,, \\ \langle w_i\, w_j \rangle &= 2\,\delta_{ij}\,\delta(t-t'), \quad i = r \ \text{ or } \ \phi\,, \end{aligned} \tag{7}$$

where μ denotes the reduced mass of the entrance channel and L_0 does the incident orbital angular momentum, *i.e.*, the total angular momentum of the system. The potential V is the sum of nuclear V_N and the Coulomb V_C potentials. The limiting angular momentum $5/7\,L_0$ is a so-called rolling limit according to Bass [12]. If we include a rolling friction acting on the relative angular velocity of the two ions, it becomes to be the sticking limit. The dissipation-fluctuation theorem is as follows: $\theta_1^2 = K_r\,T(t)$ and $\theta_2^2 = r^2\,K_\phi\,T(t)$. And from the expression of temperature above which has time t as an argument, the equation describes a heat-up process during the approaching phase. $K_i(r)$ with $i = r$, or ϕ are the radial and the tangential friction, respectively, and are given as $K_i(r) = K_i^0\ (dV_N/dr)^2$ with the strength parameters, where $K_i^0 = 4$ and 0.01 for $i = r$ and ϕ, in unit of 10^{-23} sec/MeV.

As stated in Section 1, our purpose of employing SFM is to know how the system reaches the contact point q_0, so we solve Eq. (7) up to that point to analyze the results which are to be used as initial values for the

subsequent evolutions. Naturally, we can take q_0 to be the sum of R_1 and R_2 where R_i's denote the half density radii of the projectile and the target of the entrance channel, respectively. Of course, it could be smaller or larger than that, or even it could have a distribution in general. The model is applied to ^{100}Mo+^{100}Mo system ($Z_1\,Z_2$ =1764, Z_i = 42 being atomic numbers of the projectile and targets) where the fusion hindrance starts to be observed experimentally. Fig. 3 shows calculated sticking probability $P_{\rm stick}^{J=0}$ as a function $E_{\rm lab}$. As expected, $P_{\rm stick}^{J=0}$ increases gradually as $E_{\rm lab}$ increases above the Coulomb barrier, which indicates an possible existence of the extra-push energy of only a few MeV or less. Interesting are distributions of the radial momentum at the contact point which are shown in Fig. 4 for four different incident energies. The distributions all appear to be Gaussian, though the center or the mean value of the momentum decreases as the energy increases. (Note that the incoming radial momentum has a negative sign.) At the same time, the width or the variance increases in consistence with the temperature defined with the internal excitation energy, *i.e.*, with the loss of the kinetic energy. This means that the initial value p_0 in Section 2 is not a single value, but has a distribution.

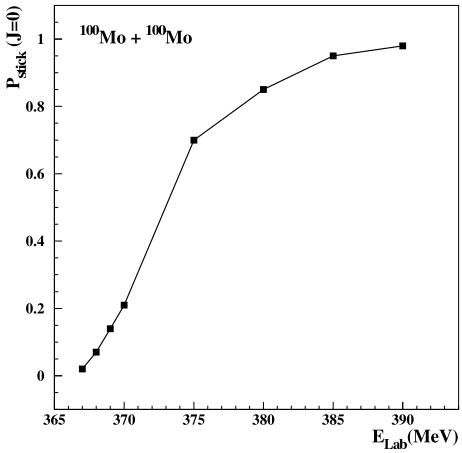


Fig. 3. Energy dependence of the sticking probability obtained by SFM is shown for ^{100}Mo–^{100}Mo system.

Thus, the calculated results are casted approximately into the following form

$$S\left(p_0, E_{\rm c.m.}\right) = P_{\rm stick}\left(E_{\rm c.m.}\right)\, g\left(p_0, \bar{p}_0, T_0\right), \tag{8}$$

where S denotes a probability for the system to have a radial momentum p_0 at the contact point for a given incident energy $E_{\rm c.m.}$ And the Gaussian

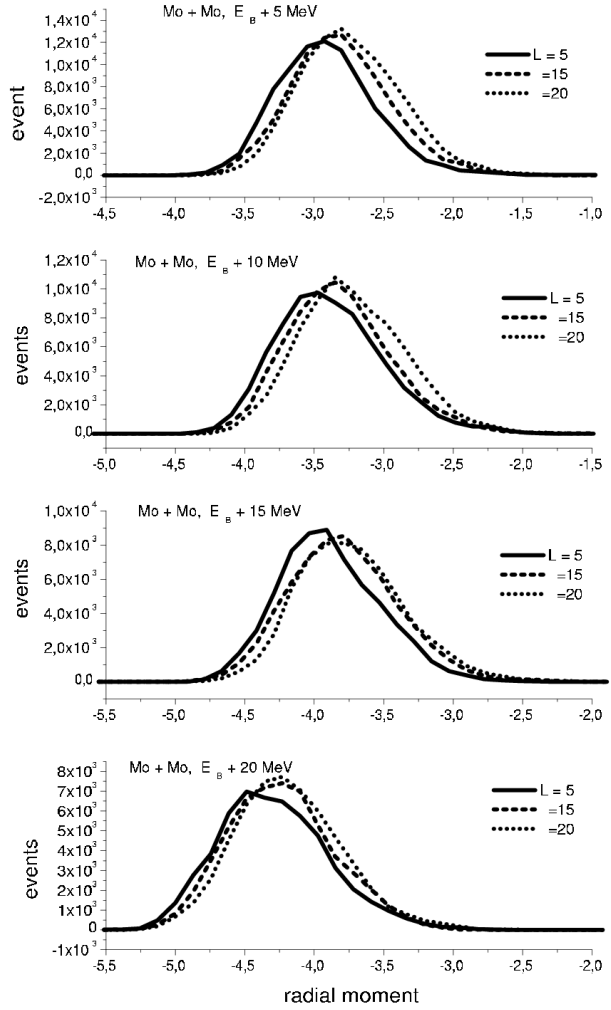


Fig. 4. Calculated radial momentum distributions are shown for ^{100}Mo-^{100}Mo system at four different incident energies. Note that incident radial momenta are negative by definition.

distribution function is defined as usual

$$g\left(p_0, \bar{p}_0, T_0\right) = \frac{1}{\sqrt{2\pi\mu T_0}} \exp\left[-\frac{\left(p_0 - \bar{p}_0\right)^2}{2\mu T_0}\right], \tag{9}$$

where the mean value of the radial momentum is denoted by $\bar{p}_0$ and the temperature of the heated-up amalgamated system is denoted by T_0. The

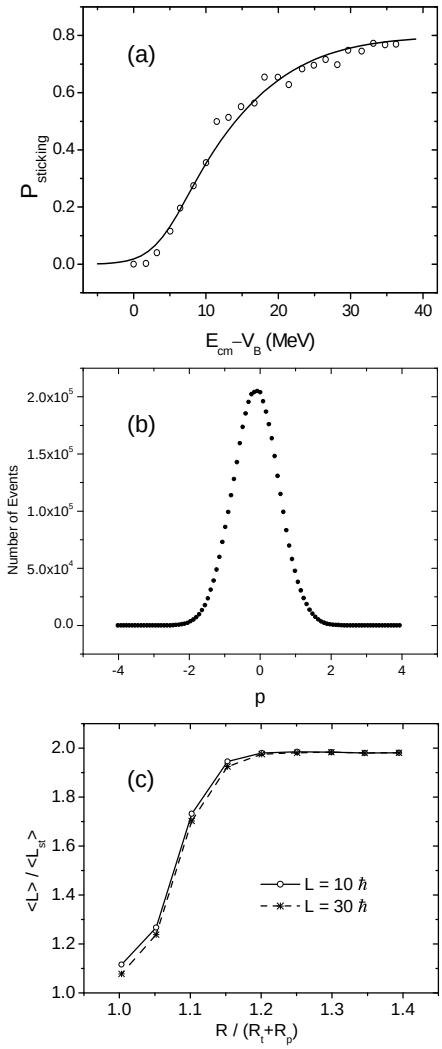


Fig. 5. Results obtained by SFM are shown for ^{48}Ca+^{244}Pu; (a) the sticking probability, (b) the radial momentum distribution in unit of 10^{21} MeV sec/fm and (c) the orbital angular momentum divided by the dissipation limit. (b) and (c) are an example with incident energy of 5 MeV above the Coulomb barrier.

energy conservation in average is expressed as follows:

$$E_{\text{c.m.}} = U_0 - E_{\text{shell}} + \varepsilon_0 + K_0 + T_0/2 - Q\,, \tag{10}$$

where Q, U_0 and E_{shell} are the Q-value of the fusion reaction, the LDM potential energy at the contact point and the shell correction energy of

the compound nucleus, neglecting that at the contact point. ϵ_0 and K_0 denote the internal excitation energy of the system and the average kinetic energy left, $\varepsilon_0 = a_0 T_0^2$ and $K_0 = \bar{p}_0^2/2\mu$ respectively, where a_0 denotes the level density parameter to be calculated with the formula by Töke and Swiatecki [13] for the corresponding nuclear shape.

Next, the model is applied to synthesis of SHE. Fig. 5 shows the results on ^{48}Ca + ^{244}Pu system, as an example. The top panel shows probability for the system to reach the contact point as a function of center-of-mass incident energy relative to the Coulomb barrier top. It is readily seen that more than 10 MeV is necessary for the probability to reach 1/2, which could be a part of the extra-push energy. The middle panel shows a Gaussian distribution of the radial momentum at the contact point in the same as in ^{100}Mo–^{100}Mo system. The center of the distribution, *i.e.*, the average value is almost equal to zero, which is very different from ^{100}Mo–^{100}Mo system and indicates a complete damping of the incident energy. Its width is consistent with a temperature of the system. The bottom panel shows radial distance-dependences of average orbital angular momenta for the cases of incident angular momenta 10 $\hbar$ and 30 $\hbar$, respectively. It is remarkable that they approach almost the dissipation limit at the contact point, which, together with the damping of the radial motion shown above, indicates a formation of sticking configuration with the thermal fluctuation.

4. Connection of the two steps

The connection is a delicate problem about amalgamation dynamics, but the present treatment provides a promising approach, which could be called "statistical method", and is different from diabatic or adiabatic viewpoint [6]. According to the results obtained in the previous section, the formation probability is given by a convolution of the probability F and the distribution g

$$P_{\rm form}\left(E_{\rm c.m.}\right) = \lim_{t\to\infty} \int_{-\infty}^{\infty} dp_0 F(t, T; q_0, p_0)\, g\left(p_0, \bar{p}_0, T_0\right) , \qquad (11)$$

where the temperature T should be determined by a similar relation to Eq. (5) around the saddle point, but for ^{48}Ca induced reactions, the contact point and the saddle point are close to each other, so they could be taken to be the same. Then, the formation probability takes an extremely simple form

$$P_{\rm form}\left(E_{\rm c.m.}\right) \cong \frac{1}{2}{\rm erfc}\left[\sqrt{\frac{B}{T}} - \frac{1}{\sqrt{x^2+1}+x}\sqrt{\frac{K_0}{T}}\right] . \qquad (12)$$

Here, we can once more make the same argument as in Section 2. That is, in order for the formation probability to be equal to 1/2, an average kinetic energy to be left at the contact point is given as follows:

$$K_0^{1/2} = \left(\sqrt{x^2+1}+x\right)^2 B\,. \tag{13}$$

It should be noticed here that the extra-push energy should be discussed not only by the formation probability, nor only by the sticking probability, but by the fusion probability which is given by Eq. (1), *i.e.*, by their product.

In massive systems where K_0 is almost equal to zero, we can use an asymptotic expansion of the error function for the case of $B \gg T$ and then

$$P_{\rm form}\left(E_{\rm c.m.}\right) \cong \frac{1}{2}\frac{1}{\sqrt{\pi}}\sqrt{\frac{T}{B}}\mathrm{e}^{-B/T}\,, \tag{14}$$

which appears to be similar to Kramers formula for fission, having an Arrhenius factor and thereby could be called as "inverse Kramers formula" [14]. But it should be noticed that Kramers formula [15] is on the transition rate, while the present formula is on the transition probability.

5. Examples of fusion cross sections and comparisons with the experiments

In order to make more realistic calculations, we employ two-dimensional model for shape evolutions in TCP, where not only the center-of mass distance but also the mass asymmetry are used as collective coordinates for q_i's. (The other parameters are freezed. The neck parameter ε is taken to be 0.8, and the fragment deformations are to be zero.) Then, multi-dimensional Langevin equation is written [16] as follows:

$$\begin{aligned}
\frac{dq_i}{dt} &= \left(m^{-1}\right)_{ij} p_j\,,\\
\frac{dp_i}{dt} &= -\frac{\partial U}{\partial q_i} - \frac{1}{2}\frac{\partial}{\partial q_i}\left(m^{-1}\right)_{jk} p_j\, p_k - \gamma_{ij}\left(m^{-1}\right)_{jk} p_k + g_{ij}R_j(t)\,,\\
g_{ik}g_{jk} &= \gamma_{ij}\,T\,,
\end{aligned} \tag{15}$$

where p_i's denote the conjugate momenta, and summations over repeated indices are implicitly assumed. The inertia tensor m_{ij} is calculated by Werner-Wheeler approximation [17] for each shape, *i.e.*, as a function of the coordinates, and thereby the inertia term appears in the r.h.s. of the second equation of Eq. (15). The potential U is the macroscopic LDM one with TCP. For finite total spins, rotational energies should be added, calculated

with the rigid moment of inertia [18]. The microscopic shell correction energy is neglected by considering that the composite system formed is rather excited already in the approaching phase, *i.e.*, in the first step. The friction tensor γ_{ij} is calculated by OBM, and the random force $\{R_i\}$ is assumed to be Gaussion, but is chosen in consistence with the dissipation-fluctuation theorem which is expressed in the last equation of Eq. (15).

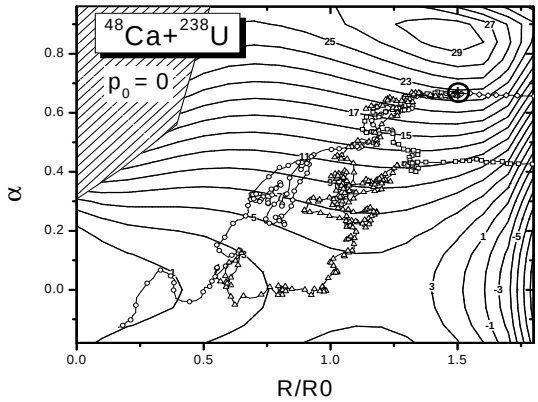


Fig. 6. Examples of Langevin trajectories of shape evolutions in two-dimensional space of the distance R and the mass-asymmetry α. (R_0 denotes a radius of the spherical compound nucleus.) The system is ^{48}Ca+^{238}U with zero initial radial momentum and the temperature corresponding to 70 MeV of intrinsic excitation.

In order to obtain the formation probability $F(t, T, q_0, p_0)$ at $t \to \infty$ for a given initial momentum p_0, we have to calculate many trajectories due to the existence of the stochastic force. Fig. 6 shows examples of trajectories for ^{48}Ca+^{238}U on the LDM energy surface, where the initial momentum p_0 is taken to be zero at the contact point and the temperature is calculated with the excitation energy of 70 MeV taken as an example. It is readily seen that some trajectories go toward the spherical shape, while some others go back to re-separation. Repeating the same type of calculations with various initial momentum p_0, and putting them into Eq.(11), we obtain the formation probability $P_{\rm form}(E_{\rm c.m.})$ for the two-dimensional model, then consequently the fusion probability $P_{\rm fusion}(E_{\rm c.m.})$. Fig. 7 shows examples of ^{48}Ca+^{244}Pu system.

Then, fusion excitation functions are calculated as usual

$$\sigma_{\rm fusion} = \pi \lambda\!\!\!^{-2} \Sigma \, (2J+1) \; P^J_{\rm fusion} \,, \tag{16}$$

where $\lambda\!\!\!^{-}$ is wave length divided by $2\,\pi$. In Fig. 8, several examples of excitation functions are shown for ^{48}Ca induced reactions, together with the available experimental cross sections. Firstly, the calculations reproduce the

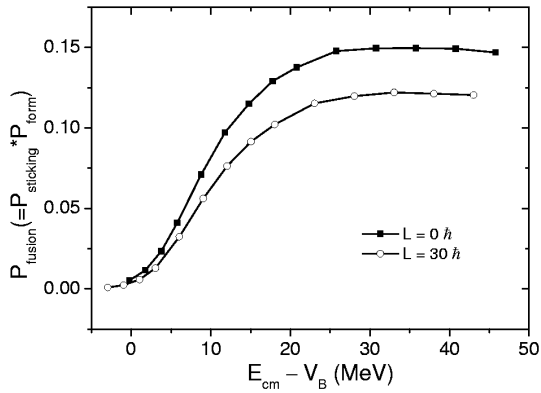


Fig. 7. Fusion probability calculated by products of sticking probabilities and of formation probabilities is shown for ^{48}Ca+^{238}U system.

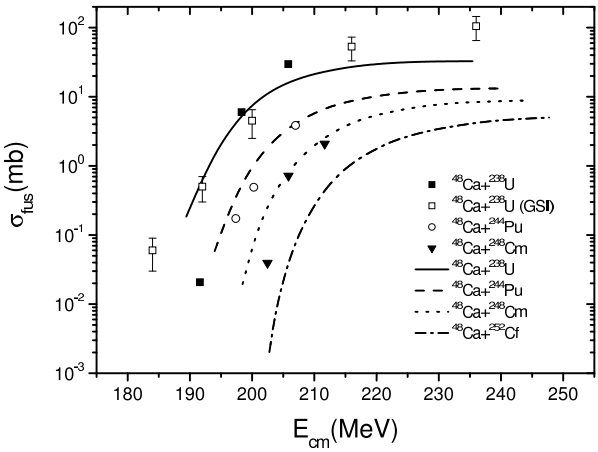


Fig. 8. Calculated fusion excitation functions for four systems of ^{48}Ca + actinide targets are shown, together with the available experimental data; GSI [19] and Dubna [20].

characteristic feature of saturation of the cross section in higher energies which is typically seen in ^{238}U target case. Secondly, they reproduce the measured excitation functions [19,20] for three systems systematically without any adjustable parameters. Experiments on ^{252}Cf target are strongly desired in order to verify the present prediction also given in the figure.

Combined with the statistical theory of decay for calculations of $P_{\rm surv}$, we can calculate residue cross sections. Preliminary results are compared with Dubna experiments [21] and have turned out to be promising. A systematic study [22] is now being made for predictions for residue cross sections for

SHE's, where precise values of the shell correction energy are crucial. Unfortunately, several theoretical predictions [23] available from the structure studies are different from each others, which gives rise to a rather large dispersion of the predicted cross sections. More precise predictions of the shell correction energy are eagerly waited for.

The authors like to thank B. Giraud, T. Wada, and B. Bouriquet for helpful discussions and encouragements. D. Boilley acknowledges supports by JSPS for visiting Yukawa Institute for Theoretical Physics, and kind hospitalities extended to him by the staffs and the secretaries of the Institute. Y. Abe appreciates supports given by GANIL and encouragements by the experimentalist of GANIL. The work is partially supported by the Grant-in-Aid of JSPS (no.13640278).

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