Two-Step Model of Fusion for Synthesis of Superheavy Elements

Caivan Shen\textsuperscript{1,2,4,6}, Grigori Kosenko\textsuperscript{3,4} and Yasuhisa Abe\textsuperscript{5}

\textsuperscript{1} China Institute of Atomic Energy, P.O.Box 275(18), Beijing 102415, China
\textsuperscript{2} INFN-LNS, 44 Via S. Sofia, I-95123 Catania, Italy
\textsuperscript{3} Department of Physics, University of Omsk, Omsk, Russia
\textsuperscript{4} RIKEN, 2-1 Hirosawa, Wako-shi, Saitama 351-0198, Japan
\textsuperscript{5} Yukawa Institute for Theoretical Physics, Kyoto University, Kyoto, Japan
\textsuperscript{6} Center of Theoretical Nuclear Physics, National Laboratory of Lanzhou, Lanzhou, China

A new model is proposed for fusion mechanisms of massive nuclear systems where so-called fusion hindrance exists. The model describes two-body collision processes in an approaching phase and shape evolutions of an amalgamated system into the compound nucleus formation. It is applied to \textsuperscript{48}Ca-induced reactions and is found to reproduce the experimental fusion cross sections extremely well, without any free parameter. Combined with the statistical decay theory, residue cross sections for the superheavy elements can be readily calculated. Examples are given.

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How many elements exist in the nature or what is the heaviest element has been an intriguing question since the periodic table was proposed for the chemical elements. The heaviest element that exists in the nature is now known to be Uranium with atomic number Z being 92. But the discovery of the magic numbers in atomic nuclei and their understanding by the shells of nucleonic motion\cite{1} suggest that much heavier atomic nuclei might exist, stabilized by extra-bindings due to possible shells next to the largests known, i.e., Z=82 and N=126. Actually, many theoretical calculations have been made, predicting the next double closed shell nucleus to be with Z=114, 120, or 126 and N=184\cite{2}. Naturally, enormous experimental efforts have been devoted to finding out traces of existence of the corresponding superheavy atomic nuclei and to synthesizing them with nuclear reactions, especially with heavy-ion fusion reactions \cite{3}. But what combination of ions is favorable as entrance channels and what incident energy is the optimum for residues are not predicted well, and thus, the experiments have been performed according to the results of systematic studies done so far. This is due to the lack of our knowledge of reaction mechanisms.

Based on the theory of compound nucleus reactions, the residue cross sections are given as follows,

\[ \sigma_{\text{res}} = \pi \lambda^2 \Sigma J (2 J + 1) \cdot P_{\text{fusion}}^J (E_{\text{c.m.}}) \cdot P_{\text{surv}}^J (E^*) , \quad (1) \]

where \( \lambda \) is the inverse of the wave number and \( J \) is the total angular momentum quantum number. \( P_{\text{fusion}} \) and \( P_{\text{surv}} \) denote the fusion and the survival probabilities, respectively. The latter is given by the statistical theory of decay, i.e., by competitions between neutron emission and fission decay. Essentially unknown is the fusion probability, i.e., fusion mechanism of massive systems, although there are ambiguities in the parameters in the properties of heavy and superheavy nuclei which give rise to uncertainties in calculating the survival probability.

In lighter systems, the fusion probability is well determined by the barrier defined with the Coulomb and the nuclear attraction between nuclei in the entrance channel, but in massive systems, the situation is not so simple. It has been well known experimentally that there is the fusion-hindrance \cite{4}, which is often described with so-called extra-push energy which is required for a system to fuse in addition to the barrier height \cite{5}. A physical origin or mechanism is not yet well clarified. There are two possible interpretations proposed. They both attribute it to energy dissipations; one is due to the dissipation of the initial kinetic energy during two-body collisions passing over the barrier \cite{6}, while the other is due to the dissipation of the energy of collective motions which would lead an amalgamated system to the spherical compound nucleus \cite{5}. It is natural to consider that both mechanisms exit, though we don’t know a priori which one dominates in which situation. We, thus, propose a new theoretical framework for fusion, i.e., a two-step model which incorporates both of them properly \cite{7}.

In the approaching phase of passing over the Coulomb barrier, we describe the system as collision processes under frictional forces, up to the contact point of two incident nuclear matters and then, we describe dynamical evolutions of the amalgamated mono-nuclear system toward the spherical shape under frictional forces acting in collective motions of excited nuclei. As is given below, both dynamical processes are described by Langevin equations which include random forces associated to the respective frictions. It would be worth to mention here that the fluctuations due to the random forces are crucially important in problems of small probability such as in synthesis of the superheavy elements (SHE), because we have to investigate cases where mean trajectories never reach the spherical shape. Another point to be mentioned is that since the two steps are connected successively, the results of the first step not only gives a probability for incident ions to stick to each other (sticking probability \( P_{\text{stick}} \)) but also gives initial conditions for the second step. Thus, the method of the connection from the first to the second steps is natural, which is neither related to the diabaticity nor the adiabaticity. It is
completely new and could be called as “statistical”, as will be seen below. In massive systems, there is a conditional saddle point, or a ridge line between the amalgamated configuration and the spherical shape on the potential energy surface calculated with the nuclear drop model (LDM), which could be considered to be another barrier inside and makes most trajectories to return back to re-separation (quasi-fission, etc.), i.e., gives rise to a small probability for forming the spherical shape (formation probability \( P_{\text{form}} \)). Thus, the fusion probability is given by the product of the two probabilities,

\[
P_{\text{fusion}}(E_{\text{c.m.}}) = P_{\text{stick}}^J(E_{\text{c.m.}}) \cdot P_{\text{form}}(E_{\text{c.m.}}).
\]  

(2)

In order to realize the model, we employ the surface friction model (SFM)[8] for the approaching phase and the one-body wall-and-window formula[9] of the dissipation for the shape evolutions, i.e., for the second step.

As for the approaching phase, the equation of motion is only for the radial degree of freedom and the orbital angular momentum, and is given below,

\[
\frac{dr}{dt} = \frac{1}{\mu} p,
\]

\[
\frac{dp}{dt} = -\frac{dV}{dr} - \frac{\partial}{\partial r} \frac{h^2 L(t)^2}{2\mu r^2} - \frac{C_r(r)}{\mu} p + \dot{R}_r(t),
\]

\[
\frac{dL(t)}{dt} = -\frac{C_l(r)}{\mu} \cdot (L(t) - L_{\text{st}}) + R_T(t),
\]  

(3)

where \( \mu \) is the reduced mass of the collision system, and \( V \) is the sum of the Coulomb potential \( V_\alpha \) and the nuclear potential \( V_n \). \( C_i(r) \) is the radial or the tangential friction coefficient which is assumed to have the following form factor,

\[
C_i(r) = K_i^0 \cdot \left( \frac{dV_i}{dr} \right)^2,
\]  

(4)

where \( K_i^0 = 0.035 \) and \( K_i^0 = 0.0001 \) in unit of \( 10^{-21} \) s/MeV. \( R_i \) denotes a random force associated with the friction for \( i = r \) (radial) or \( T \) (tangential), and assumed to be Gaussian, and to satisfy the following properties,

\[
\langle R_i(t) \rangle = 0,
\]

\[
\langle R_i(t) \cdot R_j(t') \rangle = 2\delta_{ij} \delta(t - t') \cdot C_i(r(t)) T^{J}(t),
\]  

(5)

where the last equation is the fluctuation-dissipation theorem with temperature \( T^{J}(t) \), \( J \) being equal to a total angular momentum of the system, i.e., an incident orbital angular momentum \( L \). \( L_{\text{st}} \) denotes the limiting orbital angular momentum under the friction, which is so-called the sliding limit in the SFM and is equal to \( 5/7 \cdot L \). We calculate many trajectories over relevant impact parameters and obtain probabilities for their reaching the contact point, respectively. Fig.1a shows the calculated sticking probability for \( L = 0 \) for the case of \(^{48}\text{Ca}-^{238}\text{U} \) system. Incident energy is given relative to the barrier height. It is readily seen that at energies just above the barrier there is almost no probability. This is due to the fact that the form factor assumed in SFM stretches over outside the barrier top position in massive systems. The results already appear to explain the fusion hindrance and at least partially the extra-push energy, while the second step is also expected to give rise to an additional contribution. In order to know the physical situation at the contact point, we analyze the radial momentum distribution as well as that of the orbital angular momentum. The radial momentum distribution is found to be almost purely Gaussian, as shown in Fig.1b. Its width is in consistence with the temperature of the heat bath of nucleons which is supposed to absorb the initial kinetic energy through the friction force. The example shown is for \( L = 0 \), but the other angular momentum cases behave in the same way. Therefore, the calculated distribution \( S_T^{J}(p_0, E_{\text{c.m.}}) \) can be expressed as follows for each angular momentum,

\[
S_T^{J}(p_0, E_{\text{c.m.}}) = P_{\text{stick}}^{J}(E_{\text{c.m.}}) \cdot g^{J}(p_0, \bar{p}_0^{J}, T_0^{J}),
\]  

(6)

where the normalized Gaussian distribution \( g^{J}(p_0, \bar{p}_0^{J}, T_0^{J}) \) is given generally so as to include an average mean momentum left \( \langle \bar{p}_0^{J} \rangle \) which is almost equal to zero in the present case. This distribution is used as the initial inputs to the dynamical evolutions in the second step, i.e., to Eq. (8) below. \( T_0^{J} \) denotes the temperature of the amalgamated system. The total energy available for the compound nucleus \( E^* \) is written by the energy conservation as follows,

\[
E^* = E_{\text{c.m.}} + Q = V_0 - E_{\text{shell}} + \varepsilon_0 + k_0,
\]  

(7)

where \( Q \) denotes the \( Q \)-value of the fusion reaction. \( E_{\text{shell}} \) the shell correction energy of the ground state, \( V_0 \) the LDM potential energy of the contact point, \( \varepsilon_0 \) the intrinsic excitation, and \( k_0 \) the radial kinetic energy left at the contact point. The latter two are averagely given as \( a_0 \cdot T_0^{J2} \) and \( (\bar{p}_0^{J})^2/2\mu + \frac{1}{2} T_0^{J} \), respectively with the
level density parameter $a_0$ which is calculated according to Töke and Swiatecki \[10\]. The orbital angular momentum is also analyzed. The average value is plotted as a function of the radial distance in Fig.1(c). It is seen that it approaches to the dissipation limit $L_0$ about the contact point, which indicates that the incident system reaches the sticking limit, if the rolling friction is properly taken into account. We, thus, can consider that the relative motion is completely damped and reaches the thermal equilibrium with the heat bath at the contact point, i.e., that the incident ions form an amalgamated mono-nuclear system, the probability of which depends on the incident energy and is extremely small just above the barrier. It should be noticed here that $\bar{p}_0^J = 0$ does not always hold, for example not in $^{100}$Mo-$^{100}$Mo system etc.

Subsequent shape evolutions of the pear-shaped mononucleus formed with the incident ions are described by the multi-dimensional Langevin equation which is the same as that used for dynamical studies of fission\[11\],

$$\frac{dq_{ij}}{dt} = (m^{-1})_{ij} \cdot p_j,$$
$$\frac{dp_i}{dt} = -\frac{\partial U^J}{\partial q_i} - \frac{1}{2} \frac{\partial}{\partial q_i} (m^{-1})_{jk} \cdot p_j \cdot p_k,$$
$$-\gamma_{ij} \cdot (m^{-1})_{jk} \cdot p_k + g_{ij} \cdot R_j(t),$$

$$g_{ik}g_{jk} = \gamma_{ij} \cdot T^J,$$  \(8\)

where summation is implicitly assumed over repeated suffixes. The collective mass tensor $m_{ij}$ is the hydrodynamical one and the potential $U^J$ is calculated by the finite range LDM with two-center parameterization of nuclear shapes\[12\], added with the rotational energy of the system calculated with the rigid body moment of inertia. The random force $R_j(t)$ is again gaussian with the normalization 2, and the tensor $g_{ij}$ is now related to the friction tensor $\gamma_{ij}$, as is given in the last equation, i.e., the generalized Fluctuation-Dissipation theorem in the multidimensional case. The friction tensor is calculated with the wall-and-window formula \[9\]. The temperature $T^J$ of the heat bath is better to be taken to be that at the conditional saddle point, but is approximated with that at the contact point, i.e. $T^J_0$. They are close to each other for the $^{48}$Ca-induced reactions. In the present calculations we only use the relative distance $R$ and the mass asymmetry coordinate $\alpha$ with the other degrees of freedom being frozen. For example, the neck parameter is taken to be 0.8, based on our experiences that it does not change so much during passing over the conditional saddle point in the three-dimensional calculations. Fig.2 shows examples of the trajectories on the LDM potential for $^{48}$Ca-$^{238}$U system for initial radial momenta and thus initial energies being equal to zero. Calculations of many trajectories, starting with various initial radial momenta give a distribution of formation probability $P^J(p_0, T^J)$. By making a convolution of it with the Gaussian distribution of the initial momentum $g^J(p_0, \bar{p}_0^J, T^J_0)$, we obtain the formation probability $P_{\text{form}}$

$$P_{\text{form}}(E_{\text{c.m.}}) = \int dp_0 F^J(p_0, T^J) \cdot g^J(p_0, \bar{p}_0^J, T^J_0).$$  \(9\)

Fig.3a shows the calculated formation probability for $^{48}$Ca-$^{238}$U system for L=0 and 30. In Fig.3b, the final fusion probability is plotted versus incident energy. At the first glance, the decreasing energy dependences seem to be peculiar, but the energy dependence of the passing-over probability under friction delicately depends on the strength of friction and the incident momentum. Actually, slightly weaker friction gives rise to an increasing energy dependence. A detailed analysis with the 1-dimensional model will be given elsewhere \[15\]. It should be also mentioned here that the present model is completely classical, and thus there is no quantum tunneling effect included, which limits the lowest energy to be reached.

Fusion cross sections are calculated with the fusion probability as usual, $\sigma_{\text{fusion}} = \pi a^2 \Sigma_f(2J + 1)$.
The factor 1/3 for Möller masses is rather arbitrarily chosen.

TABLE I: Calculated maximum residue cross sections of the available experimental data [13] for GSI and [14] for Dubna.

| 48Ca          | Prediction of $\Delta E_{\text{shell}}$ (MeV) | 3n: $\sigma_{\text{max,3n}}$ E | 4n: $\sigma_{\text{max,4n}}$ E |
|--------------|---------------------------------------------|-----------------------------|-----------------------------|
| $^{244}\text{Pu}$ | Liran $-0.23$ | 0.018 30.6 | 0.018 36.5 |
| Möller/3     | -2.96         | 7.39 30.1 | 6.00 35.3 |
| Experiment   | ≈ 1 $E_{\text{lab}}=236$ | ≈ 1 $E_{\text{lab}}=236$ | |
| $^{248}\text{Cm}$ | Liran $-1.37$ | 0.254 31.1 | 0.045 37.8 |
| Möller/3     | -2.86         | 4.56 30.4 | 2.98 35.6 |
| Experiment   |             | 0.6 35.8 | |
| $^{252}\text{Cf}$ | Liran $-3.24$ | 1.057 32.7 | 0.095 38.2 |
| Möller/3     | -2.41         | 0.216 28.8 | 0.086 33.5 |

$P_{\text{fusion}}^{48}$ (E$_{\text{c.m.}}$), and are shown in Fig.4 for the four systems with $^{48}\text{Ca}$ beam, together with some measured cross sections [14]. It is extremely surprising that the calculations well reproduce the experiments without any adjustment of the model parameters. Experimental measurements are highly desirable in other heavy systems for comparisons with the present calculations.

In order to show that we are ready for calculations of residue cross sections for SHE, we give examples for $Z=114, 116$ and 118, by the use of $P_{\text{surv}}$ calculated with HIVAP [16]. Actually, the shell correction energies are the most crucial quantities in residue calculations, because they effectively give the fission barriers for SHEs. And they are not yet firmly predicted, and thus we take those by Möller and Liran [17] as typical examples of mass predictions, and compare with the recent Dubna experiments [18], which are given in Table I.

In brief, the new two-step model has been found to be extremely successful in reproducing the available fusion data of $^{48}\text{Ca}$ induced reactions. By combining the present fusion probabilities with the standard statistical decay calculations, we have obtained residue cross sections for $Z=114, 116$, and 118, which are in a reasonable agreement with the recent Dubna experiments, but with rather small shell correction energies, much smaller than previously thought. A systematic study of residue cross sections are being made. Furthermore, the model is now being applied to other massive systems, such as $^{100}\text{Mo}$ etc.

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