Mass Yields of Fission Fragment of Pt to Ra Isotopes

Krzysztof Pomorski\textsuperscript{1}, Artur Dobrowolski\textsuperscript{1}, Rui Han\textsuperscript{1}, Bożena Nerlo-Pomorska\textsuperscript{1}, Michał Warda\textsuperscript{1}, Zhigang Xiao\textsuperscript{2}, Yongjing Chen\textsuperscript{3}, Lilie Liu\textsuperscript{3}, Jun-Long Tian\textsuperscript{4}

\textsuperscript{1} Institute of Physics, Maria Curie Skłodowska University, 20-031 Lublin, Poland \textsuperscript{2} Physics Department, Tsinghua University, Beijing 100084, China \textsuperscript{3} China Institute of Atomic Energy, Beijing 102413, China \textsuperscript{4} School of Physics and Electrical Engineering, Anyang Normal University, Anyang 455000, China

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An effective Fourier nuclear shape parametrization which describes well the most relevant degrees of freedom on the way to fission is used to construct a 3D collective model. The potential energy surface is evaluated within the macroscopic-microscopic approach based on the Lublin-Strasbourg Drop (LSD) macroscopic energy and Yukawa-folded single particle potential. A phenomenological inertia parameter is used to describe the kinetic properties of the fissioning system. The fission fragment mass yields are obtained by using an approximate solution of the underlying Hamiltonian. The predicted mass fragmentations for even-even Pt to Ra isotopes are compared with available experimental data. Their main characteristics are well reproduced when the neck rupture probability dependent on the neck radius is introduced.

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I. INTRODUCTION

A proper reproduction of the fission fragments mass distribution (FMD) is one of the most important tests of any theoretical model describing the nuclear fission process. A very nice review of the existing fission models can be found in Ref. \textsuperscript{[1]}, which is dedicated to the memory of Arnie J. Sierk, one of the leaders in this field of physics. Readers who are interested in the theory of nuclear fission can find more details in the textbook \textsuperscript{[2]}. So, we are not going to recall similar information here.

In the present paper we obtain such a distribution by an approximate solution of the eigenproblem of the three-dimensional collective Hamiltonian, which corresponds to the fission, neck, and mass-asymmetry modes, respectively. The nonadiabatic and dissipative effects in low energy fission were taken into account in a similar way as in Refs. \textsuperscript{[3–6]}. The potential energy surface (PES) is obtained by the macroscopic-microscopic (mac-mic) method, where the Lublin-Strasbourg Drop (LSD) model \textsuperscript{[4]} has been used for the macroscopic part of the energy, while the microscopic shell and pairing corrections have been evaluated through the Yukawa-folded (YF) single-particle levels \textsuperscript{[8, 9]}. The shape of the fissioning nucleus was described by the three-dimensional Fourier parametrization \textsuperscript{[10, 11]}. It was shown in Refs. \textsuperscript{[10, 12]} that this parametrization described very well the shapes of the nuclei even those close to the scission configuration.

The paper is organized in the following way. First we present shortly the details of the theoretical model, then we show the collective potential energy surface evaluated by the mac-mic model for the selected Pt to Ra isotopes. The calculated fission FMD’s are compared with the existing experimental data in the next section. Conclusions and plans of further calculations are presented in Summary.

II. MODEL OF THE FISSION DYNAMICS

The evolution of a nucleus from the equilibrium state towards fission is simulated by a simple dynamical approach based on the PES, which depends on three relevant collective degrees of freedom describing the nuclear shape in this process: elongation of the nucleus ($q_2$), asymmetry of left and right mass fragments ($q_3$), and the neck size ($q_4$). As demonstrated in Refs. \textsuperscript{[10, 11]}, the shape parametrization of the deformed nucleus, which gives an expansion of the nuclear surface in the form of the Fourier series of dimensionless coordinate $(z-z_{sh})/z_0$:

$$
\frac{\rho_s(z)^2}{R_0^2} = \sum_{n=1}^{\infty} \left[ a_{2n} \cos \left( \frac{(2n-1)\pi}{2} \frac{z-z_{sh}}{z_0} \right) + \theta_{2n+1} \sin \left( \frac{2n\pi}{2} \frac{z-z_{sh}}{z_0} \right) \right]
$$

is rapidly converging. As in the famous “Funny-Hills” parametrization \textsuperscript{[13]}, $\rho_s(z)$ defines the distance of the surface point from the $Oz$ symmetry axis, and $z_0$ is the half elongation of a nucleus between extreme points located at $z_{\min} = z_{sh} - z_0$ and $z_{\max} = z_{sh} + z_0$. The quantity $z_{sh}$ is responsible for shifting the center of mass of axially symmetric nuclear drop to be located at the origin of coordinate system. $R_0$ represents the radius of the corresponding spherical nucleus of the same volume. In Eq. (1), the parameters $a_2$, $a_3$, $a_4$ are related to the $q_2$, $q_3$, $q_4$ deformation parameters through the following
formulas, respectively,
\[ q_2 = q_2^0/a_2 - a_2/a_2^0, \quad q_3 = a_3, \]
\[ q_4 = q_4^0 + \sqrt{(q_2^0/9)^2 + (q_4^0)^2}, \quad q_5 = a_5 - (q_2 - 2) a_3^2, \]
\[ q_6 = a_6 - \sqrt{(q_2/100)^2 + (a_6^0)^2}. \]

These relations proposed in Ref. \[14\] transform the original deformation parameters \( a_i \) to the more natural parameters \( q_i \), which ensure that only minor variations of the liquid drop fission paths occur around \( q_4 = 0 \). In addition, more and more elongated prolate shapes correspond to decreasing values of \( a_2 \), while oblate ones are described by \( a_2 > 1 \), which is in contradiction to the traditional definition of quadrupole deformation. Here \( \alpha_n^{(0)} \) stands for the value of the \( \alpha_n \) coefficient for a sphere:

\[ \alpha_n^{(0)} = (-1)^{n-1} \frac{32}{15} \pi (2n-1)^3. \]

Having defined the shape parametrization of nuclear surface, one can now calculate the PES in 3D collective space. The nuclear deformation energies are determined in the mac-mic approach, where the smooth energy part is given by the Lublin-Strasbourg Drop (LSD) model \[17\] and the microscopic effects have been evaluated through a Yukawa-folded (YF) single-particle potential \[8, 9\]. The Strutinsky shell-correction method \[13-15\] with a 6th order correctional polynomial and a smearing width \( \gamma_S = 1.2 \hbar \omega_0 \) is used, where \( \hbar \omega_0 = 41/A^{1/3} \text{MeV} \) is the distance between the spherical harmonic-oscillator major shells. The BCS theory \[10\] with the approximate GCM+GOA-like particle number projection method \[17\] is used for the pairing correlations.

The pairing strengths \( G \Lambda^2/3 = 0.28 \hbar \omega_0 \), with \( \Lambda = Z, N \) for protons or neutrons, was adjusted to the experimentally measured mass differences of nuclei in this region with a “pairing window” containing \( \sqrt{15} \text{MeV} \) mean-field time-dependent degenerated levels lying around the Fermi level \[18\]. The mean-field used to generate the single-particle energy levels, entering the Strutinsky and Fermi level \[18\]. The mean-field used to generate the single-particle energy levels, entering the Strutinsky and Fermi level \[18\]. The mean-field used to generate the single-particle energy levels, entering the Strutinsky and Fermi level \[18\].

The present research is a continuation and extension of our previous works \[3-6\]. The fundamental idea of the fission dynamics discussed in this work is that the relatively slow motion towards fission, mainly in \( q_2 \) direction, is accompanied by the fast vibrations in the “perpendicular” \( q_3 \) and \( q_4 \) collective variables. This allows us to treat both of these two types of motion as decoupled which, in consequence, gives the wave function corresponding to the total eigenenergy \( E \) of fissioning nucleus approximately as

\[ \Psi_{nE}(q_2, q_3, q_4) = u_{nE}(q_2) \phi_n(q_3, q_4; q_2). \]

The function \( u_{nE}(q_2) \) is the eigenfunction corresponding to the motion towards fission which depends mainly on a single variable \( q_2 \), while the \( \phi_n(q_3, q_4; q_2) \) simulates the \( n \)-phonon “fast” collective vibrations on the perpendicular 2D \( \{q_3, q_4\} \) plane for a given elongation \( q_2 \).

There is a proper way to determine the \( u_{nE}(q_2) \) and \( \phi_n(q_3, q_4; q_2) \) wave function components in the discussed 3D collective space, respectively. For \( u_{nE}(q_2) \) one can use the WKB approximation for a single \( q_2 \) mode as it has been done in Ref. \[5\], in which a 2D collective space has been considered, only. For \( \phi_n(q_3, q_4; q_2) \), one can solve the eigenproblem of the underlying Hamiltonian in the perpendicular directions numerically. However, when limiting only to the low energy fission, the density of probability \( W(q_3, q_4; q_2) \) of finding the system for a given elongation \( q_2 \), within the area of \( (q_3 \pm dq_3, q_4 \pm dq_4) \), is given as

\[ W(q_3, q_4; q_2) = |\Psi(q_3, q_4; q_2)|^2 = |\phi_0(q_3, q_4; q_2)|^2. \]

A further simplification of the model is to approximate the modulus square of the total wave function in Eq. (5) by the Wigner function in the form of

\[ W(q_3, q_4; q_2) \propto \frac{V(q_3, q_4; q_2) - V_{\text{min}}(q_2)}{E_0}, \]

where \( V_{\text{min}}(q_2) \) is the minimum of the potential for a given elongation \( q_2 \), and \( E_0 \) is the zero-point energy which is treated as an adjustable parameter.

To obtain the fragment mass yield for a given elongation \( q_2 \) one has to integrate the probabilities (6) coming from different neck shapes, simulated basically by the \( q_4 \) parameter

\[ w(q_3; q_2) = \int W(q_3, q_4; q_2) dq_4. \]

It is clear that the fission probability may strongly depend on the neck thickness, strictly speaking, its radius \( R_{\text{neck}} \). Following the idea from Ref. \[5\] one assumes the neck rupture probability \( P \) to be equal to

\[ P(q_2, q_3, q_4) = \frac{k_0}{K} P_{\text{neck}}(R_{\text{neck}}), \]

where \( P_{\text{neck}} \) is a geometrical factor indicating the neck breaking probability proportional to the neck thickness, while \( k_0/K \) describes the fact that the larger collective velocity towards fission, \( v(q_2) = q_2 \), gives the less probable neck rupture. The constant parameter \( k_0 \) plays the role of scaling parameter which is finally eliminated when calculating the resulting FMD. The expression for the geometrical probability factor \( P_{\text{neck}}(R_{\text{neck}}) \) can be chosen in an arbitrary way to some extent, however after a number of trials we have used the Gaussian form \[6\]

\[ P_{\text{neck}}(R_{\text{neck}}) = \exp \left[ - \log \left( 2(R_{\text{neck}}/d)^2 \right) \right], \]

where \( d \) is the “half-width” of the probability and is treated here as another adjustable parameter. The momentum \( k \) in Eq. (8) simulates the dynamics of the fission.
yield

\[ \frac{\hbar^2 k^2}{2M(q_2)} = E_{\text{kin}} = E - Q - V(q_2) , \]

with \( \bar{M}(q_2) \) standing for the averaged inertia parameter over \( q_3 \) and \( q_4 \) degrees of freedom at a given elongation \( q_2 \), and \( V(q_2) \) is the averaged potential. In the further calculations we assume that the part of the total energy converted into heat \( Q \) is negligibly small due to the very low friction force when the collective velocity \( v(q_2) \) is small in low energy fission. A good approximation of the inertia \( \bar{M}(q_2) \) is to use the irrotational flow mass parameter \( B_{\text{irr}} \), which is derived initially as a function of the single collective parameter \( R_{12} \), the distance between fragments, and the reduced mass \( \mu \) of both fragments

\[ \bar{M}(q_2) = \mu[1 + 11.5(B_{\text{irr}}/\mu - 1)] \left( \frac{\partial R_{12}}{\partial q_2} \right)^2 . \]

In order to make use of the neck rupture probability \( P(q_3, q_4; q_2) \) in Eq. (8), one has to rewrite the integral over \( q_4 \) probability distribution \( \tilde{w}(q_3; q_2) \) in the form of

\[ w(q_3; q_2) = \int W(q_3, q_4; q_2)P(q_2, q_3, q_4)dq_4 , \]

in which now the neck rupture probability is, in addition, taken into account. The above approximation implies a very important fact that, for a fixed \( q_3 \) value, the fission may occur within a certain range of \( q_2 \) deformations with different probabilities. Therefore, to obtain the true fission probability distribution \( w'(q_3; q_2) \) at a strictly given \( q_2 \), one has to exclude the fission events occurred in the “previous” \( q_2' < q_2 \) configurations, i.e.,

\[ w'(q_3; q_2) = w(q_3; q_2) \frac{1 - \int w(q_3; q_2')dq_2'}{\int w(q_3; q_2')dq_2'} . \]

The normalized mass yield is then obtained as the sum of partial yields at different given \( q_2 \):

\[ Y(q_3) = \frac{\int w'(q_3; q_2)dq_2}{\int w'(q_3; q_2)dq_3dq_2} . \]

Since there is an one-to-one correspondence between \( q_3 \) deformation and the masses of the left \( (A_L) \) and right \( (A_R = A - A_L) \) fission fragments, the yield function of Eq. (14) can be directly compared with the experimental FMD’s now. One should notice that the scaling parameter \( k_0 \) introduced in Eq. (8) does not longer appear in the definition of mass yield. Therefore, the only free parameters of the above model are: zero-point energy parameter \( E_0 \) in Eq. (3) and the half-width parameter \( d \) appearing in the probability of neck rupture (9).

In Ref. [6], the parameter \( d = 0.15R_0 \) was adjusted to reproduce the experimental fragment mass yields measured in the low energy fission of \( ^{236-244}\text{Pu} \) isotopes. On the other hand, zero-point energy parameter \( E_0 = 1 \) MeV related to the \( q_3 \) and \( q_4 \) degrees of freedom is kept constant. The comparison of the estimates obtained by the above model with the data taken from Ref. [20] is shown in Fig. 1. It can be seen that the agreement of this model predictions with the experimental yields is pretty good.

III. RESULTS

The results obtained in Ref. [6] for Pu isotopes have encouraged us to investigate the possibilities of extend-
FIG. 3: Potential energy surface cross-sections of $^{184}$Hg on the plane $(q_3, q_4)$. The panels from top to bottom correspond to elongations $q_2 = 2.0, 2.1, 2.2,$ and 2.3, respectively. The solid red lines drawn in each panel correspond to the neck radius equaling to the nucleon radius.

A typical potential energy difference between the energies of the saddle and the most probable scission point is of the order of a few MeV, which is about only one order of magnitude smaller than that in actinides. It is then clear that in these two nuclear regions, the role of the fission dynamics should be significantly different.

The PES of all the considered nuclei are evaluated on the following 4D grid in the deformation parameter space (2):

$$
q_2 = -0.60 \ (0.05) \ 2.35, \quad q_3 = 0.00 \ (0.03) \ 0.21, \quad q_4 = -0.21 \ (0.03) \ 0.21, \quad \eta = 0.00 \ (0.03) \ 0.21, \quad (15)
$$

where the parameter $\eta$ describes the non-axial shapes of nuclei as defined in Ref. [11]. It turns out that for all the considered isotopes $\eta \neq 0$ may only appear in rather less elongated nuclei. Its influence practically ends around the deformations corresponding to the first saddle point in the fission barrier, i.e., at $q_2 \approx 1.3$. Since in the following we will only consider the fission fragment mass distribution of non-rotating nuclei, this non-axial mode will be neglected.

FIG. 4: Fission fragment distribution probability of $^{184}$Hg integrated over the deformation parameter $q_4$ (see Eq. 7) at the $(A_f, q_2)$ plane.

A typical example of PES on the $(q_2, q_3)$ plane for $^{184}$Hg, where the macroscopic-microscopic energy of $^{184}$Hg is minimized with respect to the neck degree of freedom $q_4$, is shown in Fig. [2]. The labels at the layers correspond to the energy (in MeV) with respect to the LSD macroscopic energy of spherical nucleus. The first saddle is noticed at $q_2 = 1.28$ and $q_3 = 0$, while the second one is at $q_2 = 1.69$ and $q_3 = 0.03$. Let us notice that this 2D energy map is only a projection of the full 3D PES onto $(q_2, q_3)$ plane. A more complete PES structure of $^{184}$Hg can be observed in Fig. [3] where the $(q_3, q_4)$ cross-sections corresponding to different elongations $q_2=2.0, 2.1, 2.2,$ and 2.3 are shown, respectively.
FIG. 5: Fission fragment mass yields of Pt (top part) and Hg (bottom part) isotopes. Experimental data (points with bars) are taken from Refs. [23–25].
FIG. 6: Fission fragment mass yields of Pb (top part) and Po (bottom part) isotopes. Experimental data (points with bars) are taken from Ref. [21].
FIG. 7: Fission fragment mass yields of Rn (top part) and Ra (bottom part) isotopes. Experimental data (points with bars) are taken from Refs. [21, 22, 28].
It can be seen that in Fig. 3 there are two competing minima, or better to say, fission valleys: one corresponding to the asymmetric fission around $q_3 = 0.06$ and $q_4 = 0$, and the second one towards symmetric fission around $q_3 = 0$ and $q_4 = -0.15$. One can also see that with increasing $q_2$ the symmetric valley becomes deeper. The solid red lines marked in maps of Fig. 3 correspond to the liquid drop neck radius equaling to the nucleon radius, which roughly approximate the scission lines.

The probability distribution (7) for different mass numbers ($A_f$) as a function of the elongation $q_2$ is shown in Fig. 4. It is seen that at deformations $q_2 \leq 2.15$, the asymmetric fission with the heavier fragment mass number $A_f \approx 106$ is the most probable mode, while at larger elongations, the symmetric fission channel begins to dominate. The interplay between these different fission modes depends on the fission dynamics and the neck break probability as described in the previous section. Similarly as in Ref. [6], the $d$ parameter in Eq. (9) is adjusted to the known experimental FMD’s [21, 22], while $E_0 = 1$ MeV remains unchanged. The fitted half-width $d = 1.5$ fm according to the data of Hg-Ra isotopes slightly differs from those adjusted to Pu nuclei in Ref. [6] ($d \approx 1.1$ fm). This difference may be related to different geometrical features of fission barriers in these two regions. One can say that in the considered Pt-Ra isotopes, the parameter $d$ tunes the interplay between the asymmetric and symmetric peaks in the FMD’s.

In Fig. 5 we can see the FMD’s of Pt-Hg isotopes. In the examined isotopic chains of even-even Pt nuclei, the FMD’s gradually evolve from two-peak asymmetric division with a non-zero admixture of symmetric fission towards the division with dominating symmetric channel in the neutron deficient isotopes. Moreover, the asymmetric channel strongly competes with the symmetric one in $^{178-188}$Pt, while the latter one is gradually suppressed in $^{190-198}$Pt.

Similar behaviors exhibit FMD in Hg (Fig. 5), Pb and Po (Fig. 6) isotopes. For the three experimentally measured isotopes $^{180-184}$Hg [23–25], the theoretical FMD’s asymmetric peaks are underestimated distinctly, however, their overall shapes are reproduced. Note that in contrary to the estimates made in Ref. [26], our model predicts the asymmetric fission to be the most probable mode in the lightest Hg isotopes. Our prediction for $^{190}$Hg is in line with the experimental data [23] and in addition is confirmed by Ref. [27], where the asymmetric fission mass distribution was deduced from total kinetic energy yield of the fragments. The asymmetric peaks in the FMD for $^{182}$Hg is less visible, because the measurement reported in Ref. [24] was performed at excitation energy $E_x = 33.5$ MeV, which slightly suppressed the shell effect. A substantially better reproduction of measured mass divisions is observed in Po isotopes. For $^{194}$Po and $^{196}$Po, theoretical curves fit within the error-bars of the experimental distributions taken from Ref. [21].

In Fig. 7 the predicted evolution of FMD’s in Rn isotopic chains is completely opposite to the previously discussed Pt-Po even-even chains. Neutron deficient $^{196-204}$Rn preferentially fission into two symmetric fragments, while in $^{206-212}$Rn an asymmetric component is getting more and more pronounced. The asymmetric peak becomes comparably high in moderately neutron excessed $^{214-218}$Rn, while the symmetric peak in strongly neutron rich $^{222-226}$Rn is suppressed significantly. In the experimentally measured $^{202-208}$Rn isotopes, the dominating symmetric divisions are reproduced with a tendency to slightly underestimate the asymmetric fission. Similar situations appear in Ra isotopes. The rather broad symmetric FMD’s, which are found experimentally in $^{210-218}$Ra, is confronted with the rather narrow symmetric peaks and the smaller asymmetric bump around $A_f = 126$, which are predicted by our model. The dominating asymmetric fission with heavier mass fragment around $A_f = 138$ is predicted to be heaviest for the investigated Ra isotopes. In the cases when the experimental FMD’s are available as functions of the fragment charge ($Z_f$), we have simply assumed that $Z/A = Z_f/A_f$, where $Z$ and $A$ are the charge and mass numbers of the mother nucleus, respectively.

It must be pointed out that the experimental FMD’s in the above Rn and Ra isotopes correspond to their fission at $E^* \approx 18$ MeV, i.e., about 10 MeV above the saddle point, which corresponds to the initial temperature of fissioning nucleus around $T = 0.7$ MeV. At such temperature the pairing correlations in nuclei become weaker or even disappear, which could influence the width of symmetric and asymmetric valleys in the PES’s. In addition, one has to remind that there is no dissipation in our model and it is known that this effect enlarges the width of the fission fragment yields.

IV. SUMMARY AND CONCLUSIONS

Summarizing our investigations we can write:

- a three-dimensional set of the Fourier deformation parameters is sufficient to describe the properties of the fission process,
- potential energy surfaces of nuclei are evaluated in the macroscopic-microscopic model, where the LSD energy has been used for the macroscopic smooth part, while the shell and pairing corrections are estimated on the basis of the Yukawa-folded single particle potential,
- a collective 3D model based on the elongation, mass asymmetry and neck modes is introduced,
- a Wigner function is used to approximate the probability distribution related to the neck and mass asymmetry degrees of freedom,
- a neck-breaking probability depending on the neck-size is introduced in order to reproduce the measured fission fragment mass yields.
It is shown that our collective 3D model which couples fission, neck and mass asymmetry collective modes is able to describe the main features of the fragment mass yields in Pt-Ra and Pu isotopes. Due to its simplicity, this model may serve for the rapid and pilot-type calculations of fission properties. To obtain more precise results one has to use more advanced models in which the energy dissipation and particle evaporation are taken into account in the fission dynamics, e.g. the Langevin dynamics (confer Ref. [2]) or the improved quantum molecular dynamics model (ImQMD), which has been successfully applied to describe the fission process in the heavy ion induced fission reactions, where the excitation energy increases leading possibly to shorter fission time scale and even to the occurrence of a ternary fission [29–31]. Such calculations, based on the Fourier shape parametrization, as well as on the self-consistent method, are planned to be carried out by our group in the close future.

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