Fission of super-heavy nuclei: Fragment mass distributions and their dependence on excitation energy

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The mass and total kinetic energy distributions of the fission fragments in the fission of even-even isotopes of superheavy elements from Hs (Z=108) to Og (Z=118) are estimated using a pre-scission point model. We restrict to nuclei for which spontaneous fission has been experimentally observed. The potential energy surfaces are calculated with Strutinsky’s shell correction procedure. The parametrization of the nuclear shapes is based on Cassini ovals. For the just before scission configuration we fix $\alpha=0.98$, what corresponds to $r_{neck} \approx 2$ fm, and take into account another four deformation parameters: $\alpha_1, \alpha_3, \alpha_4, \alpha_6$. The fragment-mass distributions are estimated supposing they are due to thermal fluctuations in the mass asymmetry degree of freedom just before scission. The influence of the excitation energy of the fissioning system on these distributions is studied. The distributions of the total kinetic energy (TKE) of the fragments are also calculated (in the point-charge approximation). In Hs, Ds and Cn isotopes a transition from symmetric to asymmetric fission is predicted with increasing neutron number N (at N≈168). Super-symmetric fission occurs at N≈160. When the excitation energy increases from 0 to 30 MeV, the peaks (one or two) of the mass distributions become only slightly wider. The first two moments of the TKE distributions are displayed as a function of the mass number A of the fissioning nucleus. A slow decrease of the average energy and a minimum of the width (at N≈162) is found.

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I. INTRODUCTION

Projects to measure fission fragment properties for the heaviest elements (Z > 106) are underway at several heavy-ion facilities around the world. The SHE Factory of JINR-Dubna [1], which will produce its first intense beam in 2019, the HIARY+CUBE of ANU-Canberra [2] and the tandem accelerator of the JAEA-Tokai [3] are just few examples. In this context, theoretical calculations are very useful. On one side they can improve the design of such experiments and on the other side they provide predictions to compare with data allowing to test various theoretical assumptions.

The SHE are synthesized in heavy-ion induced complete fusion reactions at Bass-barrier energies. Depending on the target used, they are usually divided in "cold" and "hot" fusion. In the first category enter closed shell $^{208}$Pb and $^{209}$Bi targets leading to compound nuclei (CN) with $\approx 20$ MeV excitation. After evaporation of maximum two neutrons the system finds itself below the fission barrier. In the second category enter neutron-rich actinides (U - Cf) targets bombarded with $^{48}$Ca leading to compound nuclei with 35 to 45 MeV excitation. Here the Coulomb repulsion of colliding nuclei, which is the main hindrance to fusion, is weaker and therefore the CN cross section larger. Of course higher excitation results in lower survival probability of CN but this is compensated by relatively high fission barriers close to N=184 shell. Nuclei with Z = 112 to 118 are produced with cross sections several orders higher in "hot" than in "cold" fussion.

The experimental method commonly used is to separate in flight the residues, formed after evaporation of neutrons and gamma rays, from the beam and implant them into a detector. The spontaneous fission occurs at the end of an $\alpha$ decay chain or competing with $\alpha$ decay during the chain. A promising alternative is to detect the primary fragments from the decay of the excited composite system before its cooling down by evaporation [4]. In this way one can study the fission of SHE at moderate excitation ($\approx 35$ MeV). It is of course necessary to separate the fusion-fission from the quasi-fission but the cross section is orders of magnitude higher than for the evaporation residue (microbarn vs. picobarn). From theoretical point of view it is therefore necessary to study both spontaneous fission of SHE and their fission at moderate excitation energies. This is the goal of the present work.

In the present study, an improved version of the scission point model [5] that has confirmed its ability to describe the mass and kinetic energy distributions of the fission fragments from the spontaneous fission of the heaviest actinides for which such distributions have been measured [6], is used. More recently, the same model was applied to long series of isotopes with atomic number Z from 110 to 126 in order to study general trends [2]. Thus, if one decreases the number of neutrons N and includes the octupole deformation $\alpha_3$ a transition from asymmetric to symmetric mass division takes place in Fl, Lv, Og and Z=126 isotopes. It is the mirror of the behaviour in Fl, No, Rf and Sg isotopes where the same
transition occurs with increasing \( N \). In this way the fragment mass systematics of the SHE and of the heavy actinides join smoothly together as shown in Fig. 1. It is a test of the reliability of the present approach. It is interesting to notice that each series of isotopes has its narrowest symmetric mass distribution with a full width at half maximum between 5 and 8 amu; hence extremely small. To distinguish it from the regular symmetric fission, we call this type of fission “super-symmetric”.

The influence of the double magic \(^{132}\text{Sn}\) is clearly seen by the almost constant mass (\( \approx 136 \)) of the heavy fragment in actinides and of the light fragment in superheavies. The masses of the complementary fragment lie on a straight line, simply reflecting the conservation of the total mass number \( A \).

However, the fission of many of these nuclei will never be observed and their study is only academic. Here we concentrate on nuclei with \( Z > 106 \) for which spontaneous fission has been already detected although the statistics was not enough to build distributions. As mentioned earlier, some of these nuclei will soon be remeasured in better conditions. Our goal is to anticipate such experiments through a detailed theoretical description of their fission fragments’ properties.

Although the total kinetic energy of the fragments is also calculated, the accent is put on their masses. Is the mass division expected to be symmetric or asymmetric? How does the excitation energy of the fissioning nucleus influences this mass division? To answer such questions, we use a pre-scission point model and therefore calculate the potential energy surface (PES) of deformation, with Strutinsky’s macroscopic-microscopic method \([9]\), for a fissioning nucleus just-before its separation into two fragments. These last mono-nuclear shapes are described by generalized Cassini ovals with up to five deformation parameters \( \{\alpha_i\} \). The corresponding collective degrees of freedom (normal to the fission direction) are supposed to be in statistical equilibrium. We therefore estimate the mass and TKE distributions using Boltzmann factors for the probability to populate the points \( \{\alpha_i\} \) on the PES.

As for the excitation energy dependence of PES, we a generalization of the Strutinsky shell correction to finite temperature, suggested in Appendix A. The generalization of the shell correction method is performed keeping the same entropy (not temperature) in the original and averaged quantities, so that the shell correction decreases monotonically, more or less exponentially with the excitation energy.

The computational approach is explained in Sec. II. In Sec. III the predicted fission fragment mass and TKE distributions are presented for the Hs, Ds, Cn, Fl, Lv and Og isotopes for which spontaneous fission has been detected. The effect of the excitation energy on the mass distributions is estimated. A summary and conclusions can be found in Sec. IV.

## II. COMPUTATIONAL DETAILS

Our model is a ”just-before scission” model \([7]\) that uses generalized Cassinian ovals

\[
R(x) = R_0[1 + \sum_n \alpha_n P_n(x)],
\]
to describe the ensemble of nuclear shapes involved. \( R_0 \)
the radius of the spherical nucleus, \( P_n(x) \) are Legendre
polynomials and \( \alpha_n \) are the shape (deformation) parameters.
These shapes have in common a parameter \( \alpha \) chosen such
that at \( \alpha = 1.0 \) the neck radius is equal to zero irre-
spective of the values of \( \alpha_n \). For the just before scission
configuration we fix \( \alpha = 0.98 \) [3], and take into account
another four deformation parameters \( \alpha_1, \alpha_3, \alpha_4, \alpha_6 \).

With the shape parametrization [11] we calculate the
potential energy of deformation using the microscopic -
macroscopic approach [3]:

\[
E_{def} = E_{def}^{LD} + \delta E,
\]

where \( E_{def}^{LD} \) is the macroscopic liquid-drop energy includ-
including surface and Coulomb energies and \( \delta E \) contains the
microscopic shell and pairing corrections.

Each point \((\alpha_1, \alpha_3, \alpha_4, \alpha_6)\) on the potential energy sur-
faces has a certain probability to be realized. Supposing
statistical equilibrium for the collective degrees of free-
dom normal to the fission direction [11], the distribution of these probabilities is

\[
P(\alpha_1, \alpha_3, \alpha_4, \alpha_6) \propto e^{-E_{def}(\alpha_1, \alpha_3, \alpha_4, \alpha_6)/T_{coll}}.
\]

Projecting \( P(\alpha_1, \alpha_3, \alpha_4, \alpha_6) \) on the fixed value of mass
asymmetry \( \eta = (A_H - A_L)/A \) one obtains the fission
fragment mass distribution \( Y(\eta) \),

\[
Y(\eta) = \frac{\sum_{ijk} P(\alpha_1(\eta), \alpha_3(\eta), \alpha_4(\eta), \alpha_6(\eta))}{\int d\eta \sum_{ijk} P(\alpha_1(\eta), \alpha_3(\eta), \alpha_4(\eta), \alpha_6(\eta))}.
\]

\( T_{coll} \) is an unknown parameter that controls the overall
width of the distribution. In a sense it takes partially into account the dynamics.

In a similar way one obtains the fission fragment total
kinetic energy (TKE) distribution. For each point
\((\alpha_1, \alpha_3, \alpha_4, \alpha_6)\) one calculates the Coulomb interaction of the fragments

\[
F_{col}^{int} = e^2 Z_l Z_H / D_{cm} = TKE
\]

Within a quasi-static approach one can not have access
to the pre-scission kinetic energy. Including only the
Coulomb repulsion energy into TKE, our estimates rep-
represent lower limits.

The TKE distribution

\[
Y(TKE) = \sum_{ijkl} P(\alpha_{i1}, \alpha_{3j}, \alpha_{4k}, \alpha_{6l}) e^{-\left(\frac{TKE_{ijkl} - TKE}{\sqrt{\pi \Delta E}}\right)^2}/\sqrt{\pi \Delta E},
\]

accounts for the finite energy resolution through the param-
parameter \( \Delta E \).

Here we will present only the first two moments calculated with the following equations:

\[
<TKE > = \frac{\int TKE Y(TKE) dTKE}{\int Y(TKE) dTKE},
\]

and

\[
\sigma_{TKE}^2 = \frac{\int (TKE - <TKE>)^2 Y(TKE) dTKE}{\int Y(TKE) dTKE}.
\]

III. PREDICTED FISSION FRAGMENT MASS
AND KINETIC ENERGY DISTRIBUTIONS

The model described in the previous section is now
applied to fragment properties for superheavy nuclei
for which spontaneous fission has been already detected
and could therefore be re-measured with better statistics
in the near future. These are \( ^{264-278}Hs, ^{268-280}Ds, ^{276-286}Ch, ^{285-287}Fl, ^{290-294}Lv \) and \( ^{294}Og \) [11, 12].

Fig. 2: (Color online) The calculated fragment mass distribu-
tributions for isotopes of Hs, Ds and Cn for which spontaneous
fission has been detected. Three values of the excitation en-
ergy \( E^* \) have been considered. \( T_{coll} = 2 \text{ MeV}, E_d = 40 \text{ MeV} \).

The calculated mass distributions corresponding to Hs,
Ds and Cn isotopes are presented in Fig. 2. In all these
three series, a transition from symmetric to asymmetric
fission is predicted with increasing neutron number \( N \).
The transition point is \( N \approx 168 \). The dependence on the exci-
tation energy (calculated under assumption of constant
entropy) is also shown. There is no noticeable differ-
ence between \( E^* = 0 \) and 15 MeV and very small differ-
ence between \( E^* = 15 \) and 30 MeV. It is good news
since the features discussed above are not expected to be
washed out if these nuclei are produced with moderate
excitation. So the mass distributions in the SHE region
is quite robust with respect to the excitation energy of the
fissioning nucleus. The case of the heaviest nuclei
ever produced, Fl, Lv and Og, is presented in Fig. 3.
All these nuclei are predicted to fission into fragments
with unequal masses. The dependence on the excitation
energy is slightly stronger this time. As expected, the
massymmetric yield increases with \( E^* \) but not enough
to overturn the mass-asymmetric character of the distrib-
 distributions. There are already experimental indications
that \( ^{282, 284, 286}Cn \) and \( ^{284, 292}Fl \), at an excitation energy
of about 30 MeV, may fission asymmetrically [12, 13].
These data are obtained by separating the fusion-fission
and the quasi-fission components. Since the error in-
volved in this procedure is difficult to assess, a measure-
ment of spontaneous fission of the same nuclei would be
very welcome.
FIG. 3: (Color online) The same as in Fig. 2 but for isotopes of Fl, Lv and Og.

Fig. 4 shows the calculated mass distributions for a long series of Ds isotopes. The heaviest three isotopes have not been detected but they are candidates to be found together with Platinum in cosmic rays or in ores (as eka-Pt) [17]. In agreement with the trends observed previously, they are predicted to fission asymmetrically.

FIG. 4: (Color online) The same as in Fig. 2 but for a larger series of Ds isotopes.

Let us now move to the total kinetic energy distributions. For all nuclei studied here the shapes of TKE-distributions are identical (quasi-gaussian) and therefore a similar presentation as for the mass distributions is not appropriate. Instead we show the first two moments of these distributions as a function of the mass A of the fissioning isotope in Figs 5 and 6 respectively.

The prescission contribution to the average total kinetic energy is neglected meaning that the values given are lower limits. For each series of isotopes there is a slight decrease with A due to the increase of the radius \((A^{1/3})\) and a decrease of the product \(Z_L \times Z_H\). Concerning the width of the TKE distributions, they exhibit a more complex variation with the mass of the isotope. For Hs and Ds isotopes there is a minimum at \(N \approx 162\) close to the neutron number where the mass distributions are also the narrowest. It is a sign of a strong nuclear structure effect in extremely deformed nuclei (pre-scission shapes).

FIG. 5: (Color online) The average total kinetic energy of the fragments for the fission of isotopes of Hs, Ds, Cn, Fl, Lv and Og for which spontaneous fission has been detected. \(T_{coll} = 2\) MeV, \(\Delta E = 10\) MeV.

FIG. 6: (Color online) Second moments of the total kinetic energy distributions for the fission of isotopes of Hs, Ds, Cn, Fl, Lv and Og for which spontaneous fission has been detected.

IV. SUMMARY AND CONCLUSIONS

The mass and TKE distributions of the fission fragments for the fission of selected even-even isotopes of Hs, Ds, Cn, Fl, Lv and Og are estimated using a pre-scission point model. The influence of the excitation energy of the fissioning system on these distributions is studied. The underlying potential energy surfaces are calculated with Strutinsky’s shell correction procedure in a four dimensional deformation space using Cassini ovals as basic shapes. The results at finite excitation energies are obtained with a generalization of this procedure keeping the same entropy in the original and averaged quantities.

With increasing neutron number \(N\), a transition from symmetric to asymmetric fission is predicted at \(N \approx 168\).
Very narrow symmetric fission occurs at $N \approx 160$. It is a signature of a shell effect in extremely deformed nuclei. There is no dramatic change in the mass distributions when the excitation energy increases from 0 to 30 MeV; they are therefore quite robust. With increasing the total mass $A$ of the fissioning nucleus, a slow decrease of the average total kinetic energy and a minimum (at $N \approx 162$) of the width of the TKE distribution is found.

We are therefore providing fission fragment distributions in a region that will be soon explored experimentally.

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Appendix A: The energy shell correction in excited nuclei

The macroscopic-microscopic approach originally was developed for the nuclei at zero excitation energy (temperature). In \[18, 19\] the shell correction $\delta E_{\text{shell}}$ to the liquid drop energy of nucleus was defined as the difference between the sum $E(0)$ of single-particle energies $\epsilon_k$ of occupied states and some averaged quantity $\tilde{E}$. In case of no pairing

$$\delta E = E(0) - \tilde{E}, \quad (A1)$$

where

$$E(0) = \sum_{\text{occ.}} \epsilon_k = \int_{-\infty}^{\mu} g_S(e)de, \quad g_S(e) \equiv 2 \sum_k \delta(e - \epsilon_k). \quad (A2)$$

The average part of energy is calculated by replacing in \[A2\] the exact density of states $g_S(e)$ by the averaged quantity $\bar{g}(e)$,

$$\tilde{E} = \int_{-\infty}^{\mu} \bar{g}(e)de. \quad (A3)$$

The generalization of Eqs. \[A1\]-\[A3\] to finite temperature is quite straightforward,

$$E(T) = 2 \sum_k \epsilon_k n_k^T, \quad \text{with} \quad n_k^T = \frac{1}{1 + e^{(\epsilon_k - \mu)/T}}. \quad (A4)$$

The averaged energy $\tilde{E}(T)$ is defined by replacing the sum over single-particle spectrum in \[A4\] by the integral with the smoothed density of states $\tilde{g}(e)$,

$$\tilde{E}(T) = \int_{-\infty}^{\mu} \tilde{g}(e)n_k^Tde \rightarrow \delta E(T) = E(T) - \tilde{E}(T), \quad (A5)$$

with $n_k^T \equiv 1/[1 + e^{(\epsilon_k - \mu)/T}]$. In the early works \[20-27\] on the shell effects in excited nuclei and more recent publications \[28, 29\] it was shown that the shell correction to the free energy, $\delta F = \delta E - T\delta S$ ($\delta S$ is the shell correction to the entropy) decays more or less exponentially with the excitation energy. The temperature dependence of shell correction to the energy $\delta E$ is more complicated. Up to temperature of approximately 0.5 MeV the shell correction increases (in absolute value) with growing temperature and decays for larger temperatures. Such dependence is somewhat strange. Up to now there is no experimental information that the shell effects in excited nuclei may become larger compared with cold nuclei.

Looking at Fig. 2 in \[29\] it is clear that the rise (in absolute value) with temperature of the shell correction $\delta E$ to the energy comes from the shell correction to the entropy $\delta S(T) \equiv S(T) - \tilde{S}(T)$.

The shell correction to the entropy appears because in \[29\] the excited nucleus was considered as a canonical ensemble at some fixed temperature. I.e., the entropy $S$ and the average component $\tilde{S}$ were calculated at the same temperature.

Meanwhile, it is clear that the nucleus is an isolated system. There is no thermostat which would keep the fixed temperature as the nucleus gets deformed. More meaningful would be to describe the excited nucleus by the microcanonical ensemble with energy and entropy as thermodynamical potentials. I.e., the shell correction to the energy of excited nucleus should be calculated at fixed entropy, not temperature. In other words, the energy $E$ and the energy $\tilde{E}$ of the system with smoothed density distribution $\tilde{g}(e)$ should be calculated at the same entropy, thus at different temperatures,

$$\delta E(S) = E(T(S)) - \tilde{E}(\tilde{T}(S)), \quad (A6)$$

where the smoothed temperature $\tilde{T}$ is defined by the requirement that the entropies calculated with exact $g_S(e)$ and averaged $\bar{g}(e)$ densities of states are the same,

$$\tilde{S}(\tilde{T}) = S(T). \quad (A7)$$

The comparison of $\delta E(T)$ and $\delta E(S)$ is shown in Fig.7a. Unlike the $\delta E(T)$ the shell correction $\delta E(S)$ decreases monotonically with the growing temperature, what is quite reasonable.

The total shell correction (the sum over neutrons and protons) $\delta E_{\text{shell}}(S)$ is shown in Fig.7b. In this calculation we include also the shell correction to the pairing energy,

$$\delta E(S) \equiv \delta E_{\text{shell}}(S) + \delta E_{\text{pair}}(S). \quad (A8)$$

The expressions for the pairing contributions to the averaged energy and entropy are given in \[29\]. As one could expect, the pairing correlations reduce the absolute value of the shell correction and up to the critical temperature the total shell correction $\delta E(S)$ does not depend much on the excitation energy. Above the critical temperature...
the total shell correction $\delta E(S)$ decreases approximately exponentially with the excitation energy.

Finally, in Fig. 7, we propose a simple approximation for the dependence of $\delta E(S)$ on the excitation energy $E^*$,

$$\delta E(E^*) = \begin{cases} 
\frac{\delta E(0)}{|\delta E(0)|} e^{-E^* / E_d}, & \text{if } |\delta E(0)| e^{-E^* / E_d} \geq |\delta E(0)|, \\
|\delta E(0)| e^{-E^* / E_d}, & \text{if } |\delta E(0)| e^{-E^* / E_d} \leq |\delta E(0)|.
\end{cases} \quad (A9)$$

In other words, up to the critical temperature, the shell correction is the same as at $E^* = 0$ and decays exponentially for higher excitation energies. In (A9) the excitation energy is defined as the sum of excitation energies of neutrons and protons.

The use of approximation (A9) makes it possible to avoid the very time consuming computations of the dependence of shell correction on the excitation energy.

In Fig. 8 we compare the calculated dependence of the shell correction (A8) on the excitation energy and the approximation (A9) for the “light” and “heavy” super-heavy elements $^{264}$Hs and $^{294}$Og at the ground state and at scission. One can see that the approximation (A9) works reasonably well. Note, that in order to approximate the $\delta E(E^*)$ by (A9) we had to use different values of the damping parameter, $E_d = 23$ MeV at the ground state and $E_d = 40$ MeV at scission.

The more accurate approximation for the dependence of $\delta E$ on $E^*$ can be achieved with the two-parametric damping factor suggested in (29). For the sake of simplicity we use in calculations reported in the main text the exponential damping factor $\exp(-E^*/E_d)$ with the damping parameter $E_d = 40$ MeV.

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