Systematic Study of Fission Barriers of Excited Superheavy Nuclei

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A systematic study of fission-barrier dependence on excitation energy has been performed using the self-consistent finite-temperature Hartree-Fock+BCS (FT-HF+BCS) formalism with the SkM* Skyrme energy density functional. The calculations have been carried out for even-even superheavy nuclei with $Z$ ranging between 110 and 124. For an accurate description of fission pathways, the effects of triaxial and reflection-asymmetric degrees of freedom have been fully incorporated. Our survey demonstrates that the dependence of isentropic fission barriers on excitation energy changes rapidly with particle number, pointing to the importance of shell effects even at large excitation energies characteristic of compound nuclei. The fastest decrease of fission barriers with excitation energy is predicted for deformed nuclei around $N=164$ and spherical nuclei around $N=184$ that are strongly stabilized by ground-state shell effects. For nuclei $^{240}$Pu and $^{256}$Fm, which exhibit asymmetric spontaneous fission, our calculations predict a transition to symmetric fission at high excitation energies due to the thermal quenching of static reflection asymmetric deformations.

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I. INTRODUCTION

The mere existence of the heaviest and superheavy nuclei with $Z>104$ is primarily determined by shell effects [1, 2, 3, 4, 5, 6, 7]. The ground-state (g.s.) shell corrections also determine fission barriers of those systems [8, 9, 10, 11, 12] as their liquid-drop fission barriers are negligible. The discoveries of new elements using the cold- and hot-fusion reactions [13, 14] over the last decade provide us with fundamental information about the structure of the nucleus and the possible existence of the “island of stability” at the limit of the nuclear mass and charge.

Since the cross sections for production of superheavy nuclei using combinations of available stable projectiles and targets are exceedingly low, the major experimental challenge is to find optimal conditions that would lead to the synthesis of the species of interest [13, 14, 15]. Isotopes of elements with $Z$ up to 113 have been produced in cold-fusion reactions using lead or bismuth targets. In these experiments, the compound nucleus (CN) is formed at relatively low excitation energies $E^*$ of $\sim$10-12 MeV. Recently, using the beams of $^{48}$Ca and actinide targets, superheavy elements with $Z=112-116$ and 118 have been synthesized [14]. The compound nuclei formed in such hot-fusion reactions are more neutron-rich than those produced in cold-fusion experiments, and they are significantly more excited, $E^*$~36-40 MeV.

The crucial quantity that determines the synthesis of superheavy elements is the CN survival probability [10, 13, 16, 17], which strongly depends on the fission barrier characteristics. Since shell effects are quenched at high temperatures (see, e.g., Refs. [18, 19, 20, 21, 22, 23]), the stability of the heaviest and superheavy elements with respect to particle emission and fission is expected to strongly depend on excitation energy.

In the previous paper [24], it was demonstrated that fission barriers of excited superheavy nuclei vary rapidly with particle number. The main objective of the present study is to address this question globally by performing systematic calculations of fission barriers of superheavy nuclei as a function of excitation energy. Our survey has been carried out within the nuclear density functional theory (DFT) generalized to finite temperatures. Guided by results of Ref. [24], we assume that the fission process is isentropic in character. The effects due to the $E^*$ dependence of triaxial and reflection asymmetric deformations are quantified and the resulting barrier damping parameters are extracted.

We also investigate the transition from asymmetric to symmetric fission with increasing excitation energy. Experimental studies [25] indicate that there is a systematic increase in the symmetric mass yield relative to the asymmetric one with excitation energy. By calculating the reflection-asymmetric deformations along static fission pathways, we show that such a transition indeed takes place in selected nuclei.

The manuscript is organized as follows. Section II briefly summarizes the FT-HFB formalism. In particular, the need for an isentropic, rather than an isothermal, description of the fission process at finite excitation energy is emphasized. The particular realization of the FT-HF+BCS model applied in our work is presented in Sec. III. Excitation-energy dependence of fission pathways for two representative nuclei, $^{240}$Pu and $^{256}$Fm, is discussed in Sec. IV together with the results of our systematic calculations of the excitation-energy dependence of the inner fission barrier of superheavy elements. Our survey clearly demonstrates that the damping of the first barrier with $E^*$ exhibits an appreciable dependence on
shell effects. Finally, the summary of our work is contained in Sec. \( \Delta \)

**II. FINITE-TEMPERATURE HFB APPROACH**

Within the mean-field approach, heated nuclei can be self-consistently treated by the finite-temperature DFT, either within Hartree-Fock (HF) \([26, 27, 28, 29]\) or, if pairing is considered, in the Finite-Temperature Hartree-Fock-Bogoliubov (HFB) method \([24, 30, 31, 32, 33]\). The equilibrium state of a nucleus at a fixed temperature \( T \) and chemical potential \( \mu \) is obtained from the minimization of the grand canonical potential \([29, 34]\):

\[
\Omega = E - TS - \mu N,
\]

where \( E = \text{Tr}(\hat{D} \hat{H}) \) is the average energy, \( S = -k \text{Tr}(\hat{D} \ln \hat{D}) \) is the entropy, \( N = \text{Tr}(\hat{D} \hat{N}) \) is the particle-number, and the density operator \( \hat{D} \) given by

\[
\hat{D} = e^{-\beta(H-\mu N)/T} / \text{Tr}(e^{-\beta(H-\mu N)}) ,
\]

with \( \beta = 1/kT \). In the mean-field approximation, the two-body density operator defined in Eq. (2) is replaced by a one-body counterpart. The variation of \( \Omega \), with respect to density, leads to the temperature-dependent HFB equations \([35]\):

\[
\mathcal{H} \begin{pmatrix} U_i \\ V_i \end{pmatrix} = E_i \begin{pmatrix} U_i \\ V_i \end{pmatrix} ,
\]

where \( \mathcal{H} \) is the temperature-dependent HFB Hamiltonian. Finite-temperature particle and pairing density matrices \([30]\) in the FT-HFB formalism are given by

\[
\rho(\beta) = U f U^\dagger + V^*(1-f)\tilde{V} ,
\]

\[
\kappa(\beta) = U f V^\dagger + V^*(1-f)\tilde{U} ,
\]

and depend on the Fermi occupations \( f_i = (1 + e^{\beta E_i})^{-1} \).

The isothermal scenario, sometimes assumed in the context of fission process \([26, 27]\), cannot be correct as the compound nucleus is not in contact with a heat bath. Considering the fission as an adiabatic process, the isentropic picture seems to be more appropriate \([31, 30]\). As discussed in Refs. \([24, 31, 36]\), the two descriptions of fission can be operationally related through the thermodynamical identity \( \frac{\partial E}{\partial \Delta_20} \bigg|_S = \frac{\partial E}{\partial \Delta_20} \bigg|_T \) which simply states that the generalized driving force associated with the deformation \( Q_{20} \) depends only on the state of the system. This identity, useful in practical calculations, has recently been verified numerically in Ref. \([24]\) wherein the importance of self-consistency has been pointed out.

In this work, we shall follow the isentropic picture. The entropy \( S = S(T) \) has been defined as in \([24]\), i.e., it corresponds to the free energy minimum at temperature \( T_{g.s.} = T \). This value of \( S \) is then kept fixed along the fission path. In this way, the temperature changes with deformation. In particular, the temperature of the lowest minimum is always greater than that of the first barrier, and this difference is crucial for the fission barrier damping.

**III. THE MODEL**

Barrier heights obtained within the HFB and HF+BCS approaches are quite similar at low temperatures \([37, 38]\). Moreover, beyond \( kT \sim 0.7 \text{ MeV} \), the two approaches are identical as the static pairing vanishes \([23, 30, 32]\). For that reason, in this study we shall present the FT-HF+BCS results only.

Our FT-HF+BCS calculations were carried out with the Skyrme SkM* functional \([32]\) in the particle-hole channel. This functional has been optimized at large deformations; hence, it is often used for fission barrier predictions. In the pairing channel, we employed the density-dependent delta interaction in the mixed variant \([40]\):

\[
V(r-r') = V_0 (1 - \rho(r)/2\rho_0) \delta(r-r') ,
\]

where \( \rho_0 = 0.16 \text{fm}^{-1} \). The pairing-active space in BCS was assumed to consist of the lowest \( Z/N \) proton/neutron HF levels. The pairing interaction strengths \( V_0 = -438 \) and \(-372 \) (in \( \text{MeV fm}^3 \)) for protons and neutrons, respectively. They were adjusted to reproduce the experimental odd-even mass differences in \( ^{252}\text{Fm} \).

It is known from numerous studies \([9, 11, 12, 41]\) that the first saddle point is lowered by several MeV by triaxial degrees of freedom and that beyond the first barrier reflection-asymmetric deformations may become important. Therefore, when studying saddle points and fission pathways, it is imperative to employ a model which is capable of breaking axial and mirror symmetries simultaneously. For that reason, we employed a symmetry-unrestricted DFT solver HFODD \([42, 43]\) capable of treating simultaneously all possible collective degrees of freedom that might appear on the way to fission. In the present work, we adopted the HFODD solver to the FT-HFB and FT-HF+BCS frameworks along the lines of Sec. \( \Delta \).

**IV. RESULTS AND ANALYSIS**

The main objective of this study is to provide a microscopic description of fission of excited nuclei, based on the nuclear DFT. To this end, we solve the constrained FT-HF+BCS problem along a collective path defined by a mass quadrupole moment \( Q_{20} \). At each value of \( Q_{20} \), self-consistent equations are solved, whereupon the total energy of the system is always minimized with respect to all remaining shape parameters. Along the optimum path found in this way, axial and mirror symmetries can
symmetric energy curve for second saddle point, we computed the axial reflection of triaxiality on the first, and mirror asymmetry on the melting of shell effects. In order to assess the impact $kT$ at $Q$ (Fig. 1). The non-axial ("min", solid lines), all self-consistent mean-field symmetries can be broken. To illustrate the corresponding energy gain, the axial, reflection-symmetric energy curves are also shown ("sym", dashed lines). The energy curves have been normalized to zero at the ground-state minimum. The values of $kT_{g.s.} = 1, 1.5, \text{and} 2 \text{ MeV}$ correspond to excitation energies of 13.82, 36.79, and 70.88 MeV for $^{240}\text{Pu}$, and 14.93, 39.20, and 75.16 MeV (not shown) for $^{256}\text{Fm}$.

As seen in Fig. 1, and discussed in detail in Ref. [46], the triaxial direction. It is, therefore, expected that the fission pathways of these two nuclei would evolve somewhat differently with increasing excitation energy.

For $^{240}\text{Pu}$, the optimal fission pathway at zero temperature exhibits the familiar two-humped structure. At $kT_{g.s.}=1.0\text{ MeV} (E^*=13.82\text{ MeV})$, both saddle points are reduced by 2-2.5 MeV. The isentropic barriers are rapidly quenched with $E^*$, and they become very small at $kT_{g.s.}=2\text{ MeV} (E^*=70.88\text{ MeV})$ due to the thermal melting of shell effects. In order to assess the impact of triaxiality on the first, and mirror asymmetry on the second saddle point, we computed the axial reflection-symmetric energy curve for $^{240}\text{Pu}$ (marked as "sym" in Fig. 1). The non-axial ($Q_{22}$) and reflection asymmetric ($Q_{30}$) moments along the optimal fission pathway are shown in Fig. 2. The energy gain on the first barrier due to triaxiality, quite appreciable at $T=0$, becomes practically negligible at $kT_{g.s.}=1.5\text{ MeV}$ while the corresponding quadrupole moment $Q_{22}$ is nonzero even at $kT_{g.s.}=2\text{ MeV}$. This indicates that at large excitation energies the energy surface of $^{240}\text{Pu}$ becomes very soft in the triaxial direction.

A similar conclusion can be drawn for the reflection asymmetric degree of freedom $Q_{30}$ and its impact on the outer barrier. Experimentally, there is clear evidence for a transition from asymmetric to symmetric fission with excitation energy [25]. The results displayed in Fig. 1 are consistent with the observed change in the pattern of fission yields. Indeed, at $kT_{g.s.}=2\text{ MeV}$ the calculated optimal fission pathway shows a very weak octupole effect.

To further explore the transition from asymmetric to symmetric fission, we now consider $^{256}\text{Fm}$. In the heavy Fm isotopes, a sharp transition has been observed [44] from an asymmetric mass division of spontaneous fission products in $^{256}\text{Fm}$ to a symmetric mass split in $^{258}\text{Fm}$. As seen in Fig. 1 and discussed in detail in Ref. [46], at $T_{g.s.}=0$ the second barrier along the symmetric fission pathway is very broad as compared to the asymmetric case, and this explains the asymmetric distribution of fission products observed experimentally. However, at $kT_{g.s.}=1.5\text{ MeV}$, the symmetric pathway becomes close in energy to the asymmetric one. This indicates that competition between asymmetric and symmetric fission is expected to occur in $^{256}\text{Fm}$ at lower excitation energies than in $^{240}\text{Pu}$.

We would now like to address the important question of the synthesis of superheavy elements in heavy-ion fusion reactions. It has already been mentioned that the crucial quantity in the synthesis is the survival probability, which depends on the quenching of the fission barrier height with $E^*$. In order to obtain a better understand-
The dependence of a fission barrier \( E_B \) on \( E^* \) is usually approximated by a phenomenological expression

\[ E_B \propto e^{-\gamma_D E^*}, \tag{7} \]

where the barrier damping parameter \( \gamma_D \) characterizes the rate of the barrier quenching with excitation energy. It is clearly seen from Fig. 3 that the ansatz (7) well describes the FT-HF+BCS results and the parameter \( \gamma_D \) can be meaningfully extracted for every nucleus. This is in spite of the fact that many physical effects impact \( E_B \)-vs-\( E^* \) dependence. In addition to a direct dependence of \( E_B \) on entropy, significant contributions come from self-consistent variations of nuclear mean fields with \( S \), most notably the gradual decrease of triaxiality. The quenching of the pairing energy does not impact the extracted values of \( \gamma_D \) as the low-\( E^* \) part of \( E_B \) was not considered when extracting the slope of \( \ln E_B \). When inspecting Fig. 3 one can notice rather dramatic isotonic variations of the damping rate for \( Z=112 \). As discussed in Ref. 24, in the isentropic picture, the observed pattern can be attributed to the higher temperature of the lowest minimum as compared to that of saddle point.

The survey of \( \gamma_D^{-1} \) obtained in this work, shown in Fig. 4, nicely illustrates the appreciable particle number dependence of barrier damping. The maximum of \( \gamma_D^{-1} \) is predicted for \( N=176 \) and 178, while for \( N=166 \) and 168 \( \gamma_D^{-1} \) is fairly small, indicating a rapid decrease of barrier heights with \( E^* \) around 280, i.e., in the region of deformed superheavy nuclei stabilized by the deformed subshell closure \( N=162 \). For heavier systems with \( Z=122 \) and 124, the largest barrier damping effect is expected around \( N=182 \) and 184, i.e., in the region of the enhanced shell stability around the expected spherical \( N=184 \) magic gap. The strong dependence of the barrier damping parameter on \( N \) and \( Z \) indicates the importance of shell effects when modeling the formation of superheavy elements.

**V. SUMMARY**

In conclusion, we performed systematic self-consistent calculations of thermal fission barriers of superheavy nuclei based on the FT-HF+BCS extension of the solver HFODD that is capable of describing arbitrary shapes free from self-consistent symmetry constraints. Our survey of the fission barrier damping parameter demonstrates the existence of strong shell effects on \( \gamma_D \). In particular, the fastest decrease of fission barriers with excitation energy is predicted for deformed nuclei around \( N=164 \) and spherical nuclei around \( N=184 \) that are strongly stabilized by g.s. shell effects. On the other hand, for the transitional nuclei around \( N=176 \), the barrier damping is relatively weak. The particle-number dependence of \( \gamma_D \) shown in Fig. 4 is expected to impact the survival probability of the superheavy compound nuclei produced in heavy-ion fusion experiments; we hope that the values of the damping parameter obtained here can be useful in guiding future theoretical work on the production of superheavy nuclei.
We also studied the quenching of triaxial and reflection asymmetric deformations with excitation energy. For nuclei $^{240}$Pu and $^{256}$Fm, which exhibit asymmetric spontaneous fission, the FT-HF+BCS theory predicts a transition to symmetric fission at higher excitation energies. Finally, the thermal quenching of triaxiality at the first saddle point provides a significant contribution to $\gamma_D$.

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