PHYSICAL REVIEW C **80**, 011302(R) (2009)

Systematic study of fission barriers of excited superheavy nuclei

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(Received 22 April 2009; published 6 July 2009)

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 the nuclei ^{240}Pu and ^{256}Fm, which exhibit asymmetric spontaneous fission, our calculations predict a transition to symmetric fission at high excitation energies owing to the thermal quenching of static reflection asymmetric deformations.

DOI: 10.1103/PhysRevC.80.011302 PACS number(s): 24.75.+i, 21.60.Jz, 27.90.+b, 24.10.Pa

Introduction. The mere existence of the heaviest and superheavy nuclei with $Z > 104$ is primarily determined by shell effects [1–7]. The ground-state (g.s.) shell corrections also determine fission barriers of those systems [8–12] as their liquid-drop fission barriers are negligible. The discoveries of new elements using cold- and hot-fusion reactions [13,14] over the past 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 small, the major experimental challenge is to find optimal conditions that would lead to the synthesis of the species of interest [13–15]. 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 ∼10–12 MeV. Recently, by 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, with $E^* \sim 36$–40 MeV.

The crucial quantity that determines the synthesis of superheavy elements is the CN survival probability [10,15–17], which strongly depends on the fission barrier characteristics. Since shell effects are quenched at high temperatures (see, e.g., Refs. [18–23]), the stability of the heaviest and superheavy elements with respect to particle emission and fission is expected to strongly depend on excitation energy.

In a 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 of 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. First, we briefly summarize the finite-temperature Hartree-Fock–Bogoliubov (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 finite-temperature Hartree-Fock + BCS (FT-HF + BCS) model applied in our work is then presented. Excitation-energy dependence of fission pathways for two representative nuclei, ^{240}Pu and ^{256}Fm, is discussed next 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, we present a summary of our work.

Finite-temperature HFB approach. Within the mean-field approach, heated nuclei can be self-consistently treated by the finite-temperature DFT, either within the Hartree-Fock (HF) method [26–29] or, if pairing is considered, in the FT-HFB method [23,30–33]. The equilibrium state of a nucleus at a fixed temperature T and chemical potential μ is obtained

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from the minimization of the grand canonical potential Ω [29,34]. The variation of Ω, with respect to density, leads to the temperature-dependent HFB equations [35] with finite-temperature particle and pairing density HFB matrices [30] depending on the Fermi occupations.

The isothermal scenario, sometimes assumed in the context of the fission process [26,27], cannot be correct as the compound nucleus is not in contact with a heat bath. By considering fission as an adiabatic process, the isentropic picture seems to be more appropriate [34,36]. As discussed in Refs. [24,34,36], the two descriptions of fission can be operationally related through the thermodynamical identity $(\frac{\partial E}{\partial Q_{20}})_S = (\frac{\partial F}{\partial Q_{20}})_T$, 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 Ref. [24] (i.e., it corresponds to the free energy minimum at temperature $T_{\text{g.s.}} = T$). 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.

The model. Barrier heights obtained within the HFB and HF + BCS approaches are quite similar at low temperatures [37,38]. Moreover, beyond $kT \sim 0.7$ MeV, the two approaches are identical because 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 [39] 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]:

$$V(\mathbf{r} - \mathbf{r}') = V_0[1 - \rho(\mathbf{r})/2\rho_0]\delta(\mathbf{r} - \mathbf{r}'), \quad (1)$$

where $\rho_0 = 0.16$ 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 are -438 and -372 MeV fm^3 for protons and neutrons, respectively. They were adjusted to reproduce the experimental odd-even mass differences in ^{252}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 that is capable of breaking axial and mirror symmetries simultaneously. For that reason, we employed a symmetry-unrestricted DFT solver HFODD [42] 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 described earlier.

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 be broken (i.e., the multipole moments Q_{22} and/or Q_{30} may be nonzero). Figure 1 shows the fission pathways for ^{240}Pu and ^{256}Fm. The former nucleus is known to fission asymmetrically whereas the later one is on the edge of the transition from asymmetric to symmetric fission [43,44]. It is, therefore, expected that the fission pathways of these two nuclei would evolve somewhat differently with increasing excitation energy.

For ^{240}Pu, the optimal fission pathway at zero temperature exhibits the familiar two-humped structure. At $kT_{\text{g.s.}} = 1.0$ MeV ($E^* = 13.82$ 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 $kT_{\text{g.s.}} = 2$ MeV ($E^* = 70.88$ MeV) owing to the thermal melting of shell effects.

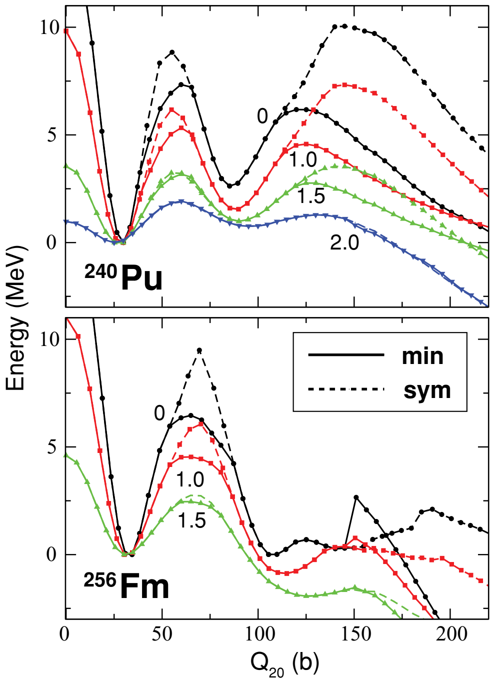

FIG. 1. (Color online) Fission pathways of ^{240}Pu (top) and ^{256}Fm (bottom) as functions of the mass quadrupole moment Q_{20} at different values of the ground-state temperature $kT_{\text{g.s.}}$ (marked by numbers, in MeV). Along the minimum-energy pathways ("min"; solid lines), all self-consistent 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 $kT_{\text{g.s.}} = 1$, 1.5, and 2 MeV correspond to excitation energies of 13.82, 36.79, and 70.88 MeV for ^{240}Pu and 14.93, 39.20, and 75.16 MeV (not shown) for ^{256}Fm.

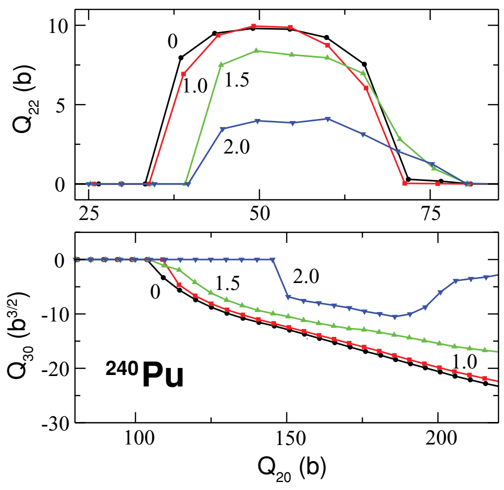

FIG. 2. (Color online) Variation of nonaxial (Q_{22}, top) and reflection asymmetric (Q_{30}, bottom) mass moments as a function of Q_{20} and temperature (indicated in MeV) for ^{240}Pu. It is seen that triaxiality and reflection asymmetry persist to $kT_{\rm g.s.} = 2$ MeV. However, as indicated in Fig. 1, their impact on the total energy is negligible at the largest temperatures considered.

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}Pu (marked as "sym" in Fig. 1). The nonaxial (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 resulting from triaxiality, which is quite appreciable at $T = 0$, becomes practically negligible at $kT_{\rm g.s.} = 1.5$ MeV, whereas the corresponding quadrupole moment Q_{22} is nonzero even at $kT_{\rm g.s.} = 2$ MeV. This indicates that at large excitation energies the energy surface of ^{240}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 $kT_{\rm g.s.} = 2$ 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}Fm. In the heavy Fm isotopes, a sharp transition has been observed [43] from an asymmetric mass division of spontaneous fission products in ^{256}Fm to a symmetric mass split in ^{258}Fm. As seen in Fig. 1, and discussed in detail in Ref. [45], at $T_{\rm g.s.} = 0$ 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 $kT_{\rm g.s} = 1.5$ 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}Fm at lower excitation energies than in ^{240}Pu.

A word of caution needs to be provided here. The one-dimensional fission pathways such as these in Fig. 1 with one multipole moment as a driving operator are clearly insufficient to describe the competition between multiple fission valleys, in particular, the identification of scission points [46] and, in some cases, saddle points [47–50]. An example of such competition can be seen in the $T_{\rm g.s.} = 0$ results for ^{256}Fm as a sudden jump between two different energy sheets. It is only through the analysis of two- or many-dimensional potential energy surfaces in the collective space that one can overcome this problem [46,50]. A more detailed analysis along such lines will be discussed in a forthcoming paper [51] in the context of multiple fission pathway investigations.

We would now like to address the 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^*. To obtain a better understanding of how the shell effects impact the E^* dependence of the first saddle point of superheavy nuclei, we performed systematic FT-HFB calculations for 48 even-even nuclei with $110 \leqslant Z \leqslant 124$ and $166 \leqslant N \leqslant 188$. A sample result illustrating our methodology is displayed in Fig. 3 for $Z = 112$, 118, and 124.

The dependence of a fission barrier (E_B) on E^* is usually approximated by a phenomenological expression [10,16]

$$E_B \propto e^{-\gamma_D E^*}, \tag{2}$$

where the barrier damping parameter γ_D characterizes the rate of the barrier quenching with excitation energy. It is clearly seen from Fig. 3 that the ansatz (2) well describes the FT-HF + BCS results and the parameter γ_D can be meaningfully extracted for every nucleus. This is in spite of the fact that many physical effects impact the E_B-versus-E^* dependence.

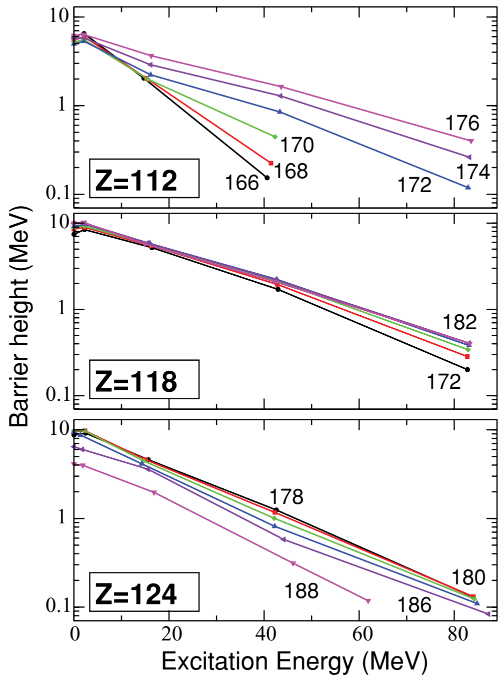

FIG. 3. (Color online) Predicted excitation energy dependence of barrier heights of even-even superheavy elements with $Z = 112$, 118, and 124.

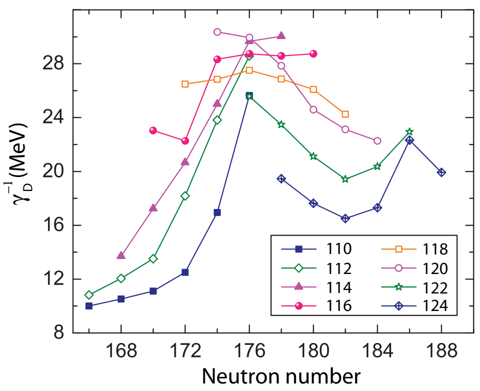

FIG. 4. (Color online) Inverse barrier damping parameter γ_D^{-1} extracted from our FT-HF + BCS calculations for 48 even-even superheavy nuclei with $110 \leqslant Z \leqslant 124$ and $166 \leqslant N \leqslant 188$.

(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 γ_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 the saddle point.

The survey of γ_D^{-1} obtained in this work, shown in Fig. 4, nicely illustrates the appreciable particle-number dependence of barrier damping. The maximum of γ_D^{-1} is predicted for $N = 176$ and 178, whereas for $N = 166$ and 168 γ_D^{-1} is fairly small, indicating a rapid decrease of barrier heights with E^* around $^{280}112$ (i.e., in the region of deformed superheavy nuclei stabilized by the deformed subshell closure $N = 162$ [4,7]). 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 [5–7]). 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.

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 γ_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. However, for the transitional nuclei around $N = 176$, the barrier damping is relatively weak. The particle-number dependence of γ_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 γ_D.

Useful discussions with Arthur Kerman, Yuri Oganessian, and Andrzej Staszczak are gratefully acknowledged. Computational resources were provided by the National Center for Computational Sciences at Oak Ridge National Laboratory. This work was supported in part by the National Nuclear Security Administration under the Stewardship Science Academic Alliances program through Grant DE-FG03-03NA00083 and by the US Department of Energy under Contract Nos. DE-FG02-96ER40963 (University of Tennessee), DE-AC05-00OR22725 with UT-Battelle, LLC (Oak Ridge National Laboratory), and DE-FC02-07ER41457 (UNEDF SciDAC Collaboration).

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