The character and prevalence of third minima in actinide fission barriers

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The double-humped structure of many actinide fission barriers is well established both experimentally and theoretically. There is also evidence, both experimental and theoretical, that some actinide nuclei have barriers with a third minimum, outside the second, fission-isomeric minimum. We perform a large-scale, systematic calculation of actinide fission barriers to identify which actinide nuclei exhibit third minima. We find that only a relatively few nuclei accessible to experiment exhibit third minima in their barriers, approximately nuclei with proton number \( Z \) in the range 88 \( \leq Z \leq 94 \) and nucleon number \( A \) in the range 230 \( \leq A \leq 236 \). We find that the third minimum is less than 1 MeV deep for light Th and U isotopes. This is consistent with some previous experimental and theoretical results, but differs from some others. We discuss possible origins of these incompatible results and what are the most realistic predictions of where third minima are observable.

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Third minima were first obtained in calculated fission potential-energy surfaces in 1972 [1], in macroscopic-microscopic calculations based on the modified-oscillator (Nilsson) single-particle potential. A year later similar results were found in macroscopic-microscopic calculations based on a substantially different single-particle potential, namely the folded-Yukawa potential [2,3], for two different shape parametrizations. Subsequently, other studies found similar results [4]. Such studies have usually found only a few actinide nuclei with third minima, mainly elements in the range 88 \( \leq Z \leq 94 \) for isotopes in the range 228 \( \leq A \leq 236 \). Normally, these minima are about 5 MeV above the ground states and are surrounded by saddles rising to about one MeV above the minimum. In our discussion, we adopt the common notation of \( E_A \), \( E_B \), and \( E_C \) for the energies of the ground state minimum, the fission isomeric (second) minimum and the third minimum, respectively, and \( E_A \), \( E_B \), and \( E_C \) for the energies of the three barrier peaks, starting from the inner one.

Since third minima appeared relatively consistently in macroscopic-microscopic calculations with different single-particle potentials and their results therefore appeared quite robust, it was suggested by Ray Nix in 1973 [2] that experimental signatures of these third minima might be observed. He also suggested that the discrepancy between the calculated height of the inner peak in the barrier, about 4 MeV, of light actinides and the consistent experimental result of 6 MeV might be resolved if the experimental barrier peak listed as 6 MeV high was actually \( E_B \) and not \( E_A \).

Inspired by these suggestions, during the next decade Blons and collaborators carried out a series of increasingly detailed experiments aimed at elucidating the properties of the third minimum [5,6]. These revealed rotational-like level structures at 5–6 MeV excitation energy, with spacings corresponding to moments of inertia consistent with the large deformations of the calculated third minima. More recent experimental results were obtained for nearby nuclei by Csige and collaborators [5,6]. For \(^{232}\)Pa the authors obtained results similar to those obtained by Blons et al. for nearby nuclei, namely a shallow, less than 1-MeV deep, third minimum at about 5 MeV above the ground state. However, their results for \(^{232}\)U deviated from this pattern. They concluded the minimum was surrounded by considerably higher peaks. According to their analysis of the experimental data the third barrier peak lies 2.8 MeV above the third minimum, which in turn is only 3.2 MeV above the ground state.

There are several motivations to carry out yet another study of third minima, despite the many previous studies. First, the results obtained in different models are sometimes mutually inconsistent [1–4,10–13] and we comment on possible reasons for these differences. Second, we have a more well-benchmarked model than those used earlier, both in terms of the number of nuclides studied (globality) and the number of properties calculated (universality). It is therefore of interest to discuss results obtained within this approach.

Constrained Hartree-Fock (and HFB) calculations, as applied so far, do not unambiguously find the correct fission saddle points due to the inherent limitations of constrained-minimization methods when applied to the problem of locating saddle points, as is discussed in some detail in Ref. [13], and is also pointed out by other authors [14,15]. All of the earlier studies based on macroscopic-microscopic models, for example [1–4], investigated a very limited number of shapes, less than 1000. Moreover, the multipole expansions used in many of these studies are of questionable suitability for the elongated shapes occurring in the later stages of fission. Furthermore, the modified-oscillator potential is very different from the folded-Yukawa potential we use here.

Therefore, when we performed our first calculations in very large deformation spaces [16,18] we anticipated a possible result could be that the third minima we had seen in more limited calculations would no longer be present. However, in these more detailed calculations we found, somewhat to our surprise, similar third minima as found earlier. In subsequent more extensive large-scale calculations of fission potential-energy surfaces we obtained results for 5254 heavy nuclei [13]. For each nucleus the energy was calculated for more than 5 million shapes as a function of five deformation parameters: elongation or quadrupole moment, neck radius, left nascent fragment spheroidal \( (\epsilon_1) \) deformation, right nascent fragment spheroidal \( (\epsilon_2) \) deformation, and nascent fragment mass asymmetry. We now analyze the calculated surfaces specifically for third minima by immersion techniques (which
FIG. 1. Calculated fission barrier for $^{236}$Ra. This is one of the deepest third minima we find, for which the surrounding saddles are of about equal height. The saddles rise about 1.5 MeV above the third minimum.

...were first used in nuclear physics in Ref. [19] and all minima deeper than 0.2 MeV and saddles between all possible pairwise combinations of minima are determined and tabulated. Full details of the calculations are given in Ref. [13]. However, in that paper third minima were discussed only briefly.

In Fig. 1 we show an example of a three-humped barrier, namely the barrier of $^{236}$Ra. This nucleus is currently not observed, but it is interesting because it has one of the deeper third minima we have found. Also interesting is that at the second saddle peak, the nascent fragments have not yet emerged. In contrast they are already very prominent at the third peak. This is also well expressed in the increasingly negative value of the shell-plus-pairing correction. The shell-plus-pairing energy is -3.39 MeV, -1.724 MeV, -5.63 MeV and -7.39 MeV at the fission-isomeric minimum, at the second saddle, at the third minimum, and at the third (outermost) saddle, respectively. Some additional barriers of currently known nuclei, for which we find third minima, are plotted in Refs. [13, 18]. The reason the nascent fragments have not yet emerged at the second peak is that this shape is too compact to (geometrically) allow the formation of two distinct fragments with a well-expressed narrow neck region in between.

In Fig. 2 we summarize our results for even-even systems. Although we have available the results for odd-even and odd-odd systems, we do not include them here because they can be estimated by interpolation, since barrier parameters change in a smooth manner with neutron and proton number. Fig. 2 shows the height of the second and third barrier peaks relative to the third minimum. We only show results for nuclei where the third minimum is deeper than 0.2 MeV relative to the lower of the two surrounding saddles. We only extend the plot to the vicinity of the last known neutron-rich nuclei. Furthermore we exclude many nuclei where we feel a third minimum would not be observable. For example for $^{242}$U we find that the second peak is about 4 MeV above the third minimum, whereas the third peak is only about 0.5 MeV above. Therefore, because of the substantial second barrier peak, resonances in the shallow third minimum would likely not be observable. A general result is that for the very lightest isotopes we consider, the outer third peak is higher than the second peak. As the neutron number increases the second peak becomes higher and the third peak drops to lower energies and is also very low with respect to the third minimum. Therefore we find that the most likely nuclei to exhibit experimental evidence of third minima in the barrier are those shown in Fig. 2 and nearby odd nuclei. Possibly the best candidates for observable third minima are those nuclei where the second and third barrier peaks are of nearly equal height. Thus our calculations predict that $^{228}$Th–$^{236}$Th and $^{228}$U–$^{236}$U would exhibit the most clear experimental signatures of third minima. The height of the third minimum relative to the ground state is shown in Fig. 3.

Our results are very similar to the macroscopic-microscopic results shown in Fig. 19 of Ref. [4], although a different po-
FIG. 2. Depth of the third minimum relative to surrounding barrier saddle points for four heavy elements. In uranium, $^{234}$U and $^{236}$U also exhibit third minima with $E_B$ about 0.6 MeV and $E_C$ about 2 MeV above the minimum for both these isotopes. U and Pu isotopes do not have third minima for systems heavier than those plotted. Th and Ra do have third minima for systems heavier than those shown, but since we mainly focus on relatively accessible nuclei here, we do not discuss them.

The results of Čwiok et al. [10] and Ter-Akopian et al. [11] show, in contrast to our results, 4-MeV deep third minima for both light Th and U isotopes [10] and the heavy actinide $^{252}$Cf [11]. In their calculations 2D potential-energy surfaces were calculated as functions of quadrupole and octupole moments $\beta_2$ and $\beta_3$, with the energy minimized with respect to several additional, higher multipoles. This approach suffers from the same mathematical limitations as the “constrained” self-consistent calculations discussed above. This has now been confirmed in an independent study [15]. This calculation, based on exactly the same macroscopic-microscopic Woods-Saxon single-particle potential model that was used in the Čwiok et al. studies, employed instead immersion methods [13, 18, 19], known to be required to locate optimal saddle points on multi-dimensional potential-energy surfaces. It was found, in close agreement with our results using a different macroscopic-microscopic model, that the third minima for Th are very shallow and only surrounded by saddles less than 1 MeV high. Therefore, we feel it may be unrealistic to expect such deep hyper-deformed minima to occur in actinide nuclei.

Csige and collaborators [8, 9] have analyzed experiments assuming a “three-humped” barrier. Somewhat in agreement with the theoretical suggestion by Čwiok et al. [10] they find for $^{232}$U a third minimum stabilized with respect to fission.
by a 2.8 MeV high third peak relative to this third minimum. This minimum is deduced to lie 3.2 MeV above the ground state. However for the nearby nucleus C$^{232}$Pa they find that the third minimum is less than 1 MeV deep and lies at an excitation energy of 5 MeV above the ground state. It is previously unheard of, both in experiments and theoretical calculations, that fission-barrier parameters change by such large amounts from commonly used values. For example for the third barrier peak in C$^{232}$U we calculate a shell-plus-pairing correction of \(-4.85\) MeV, which translates to a very low level density here. At the second peak the calculated shell-plus-pairing correction is \(-2.59\) MeV, which implies a significantly different level density.

In conclusion, we have determined the region of occurrence of triple-humped fission barriers. Only a relatively few nuclei in the light actinide region exhibit this structure. The isotope sequences C$^{228}$Th, C$^{230}$Th and C$^{228}$U, C$^{230}$U (along with neighboring odd nuclei) appear to be the best candidates for observable third minima. Our results, employing a folded-Yukawa single-particle potential in a macroscopic-microscopic model, are quite consistent both in terms of which nuclei exhibit this feature and the heights of the saddle points and minima with: 1) the experimental studies by Blons and collaborators [5][7], 2) with early computer-limited theoretical studies in both the oscillator and folded-Yukawa macroscopic-microscopic models, and 3) with recent state-of-the-art Woods-Saxon macroscopic-microscopic calculations [15] exploring a deformation space with about 1 000 000 grid points.

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1. P. M"oller, Nucl. Phys. A192 (1972) 529
2. P. M"oller and J. R. Nix, Proc. Third IAEA Symp. on the physics and chemistry of fission, Rochester, 1973, vol. I (IAEA, Vienna, 1974) p. 103
3. P. M"oller and J. R. Nix, Nucl. Phys. A229 (1974) 269
4. R. Bengtsson and I. Ragnarsson, S. Åberg, A Gyurkovich, A Sobiczewski, and K. Pomorski Nucl. Phys. A473 (1987) 77
5. J. Blons, C. Mazur, D. Paya, Phys. Rev. Lett. 35 (1975) 1749
6. J. Blons, C. Mazur, D. Paya, M. Ribrag, H. Weigmann, Phys. Rev. Lett. 41 (1978) 1282
7. J. Blons, R Fabbro, C. Mazur, D. Paya, M. Ribrag, Y. Patin, Nucl. Phys. A477 (1988) 231
8. L. Csige, M. Csatlós, T. Faestermann, Z. Gácsi, J. Gulyás, D. Habs, R. Hertenberger, A. Krzanahorkay, R. Lutter, H. J. Maier, P. G. Thirolf, H.-F. Wirth, Phys. Rev. C 80 (2009) 054306
9. L. Csige, M. Csatlós, T. Faestermann, J. Gulyás, D. Habs, R. Hertenberger, M. Hunyadi, A. Krzanahorkay, H. J. Maier, P. G. Thirolf, H.-F. Wirth, Phys. Rev. C 85 (2012) 054306
10. S. Ćwiok, W. Nazarewicz, J. X. Saladin, W. Plościennik, A. Johnson, Phys. Lett. B322, 304 (1994)
11. G. M. Ter-Akopian, J. H. Hamilton, Y. T. Oganessian, A. V. Daniel, J. Kormicki, A. V Ramayya, G. S. Popeko, R. R. Babu, Q. H. Lu, K. Butlerrmore, W. C. Ma, S. Ćwiok, W. Nazarewicz, J. K. Deng, D. Shi, J. Kliman, M. Morhac, J. D. Cole, R. Aryaeinejad, N. R. Johnson, I. Y. Lee, F. K. McGowan, J. X. Saladin, Phys. Rev. Lett. 77, 32 (1996)
12. M. Samyn, S. Goriely, and J.M. Pearson, Phys Rev. C 72 (2005) 044316
13. P. Möller, A. J. Sierk, T. Ichikawa, A. Iwamoto, R. Bengtsson, H. Uhrenholt, and S. Åberg, Phys. Rev. C 79 (2009) 064304
14. N. Dubray, D. Regnier, Comp. Phys. Comm. 183 (2012) 2035
15. M. Kowal and J. Skalski, Phys. Rev. C 85 (2012) 061302
16. P. Möller and A. Iwamoto, Proc. Conf. on Nuclear Shapes and Motion. Symposium in Honor of Ray Nix, 25–27 Oct. 1998, Sante Fe, NM, USA Acta Physica Hungarica, New Series, 10 (1999) 241
17. P. Möller and A. Iwamoto, Phys. Rev. C 61 (2000) 047602
18. P. Möller, D. G. Madland, A. J. Sierk, and A. Iwamoto, Nature 409 (2001) 785
19. A. Mamdouh, J. M. Pearson, M. Rayet, and F. Tondeur, Nucl. Phys. A644 (1998) 389