Test of the adequacy of using smoothly joined parabolic segments to parametrize the multihumped fission barriers in actinides

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The adequacy of using smoothly joined parabolic segments to parametrize the multihumped fission barriers has been tested by examining its simultaneous consistency with the three relevant fission observables, namely, the near-barrier fission cross sections, isomeric half-lives, and the ground-state spontaneous fission half-lives of a wide variety of a total of 25 actinide nuclides. The penetrabilities through such multihumped fission barriers have been calculated in the Wentzel-Kramers-Brillouin approximation, and the various fission half-lives have been determined using the formalism given earlier by Nix and Walker. The results of our systematic analysis of these actinide nuclides suggest that such a parametrization is quite adequate at least for the even-even nuclei, as it reproduces satisfactorily their various observed fission characteristics. Major difficulties remain, however, for the odd mass and for the doubly odd nuclei where the calculated ground-state spontaneous fission half-lives are found to be several orders of magnitude larger than those measured. Possible reasons for such discrepancies are discussed. Fission branching ratios of the decay of the shape isomers in various actinide nuclides have also been calculated and are compared with their measured values.

I. INTRODUCTION

Most of the actinide nuclei are now known to exhibit double- or triple-humped fission barriers. Information concerning the various parameters of such fission barriers is therefore of importance not only in understanding the physics of fission phenomenon but also for practical applications as some of these nuclei are used as the fuel in the nuclear reactors while some others are produced during the operation of such reactors. Measurements of the fission half-lives, the fission cross sections, and of the angular distributions of the fission fragments of such nuclei provide us a means to estimate the heights and curvature parameters of the corresponding fission barriers. In view of the extremely small cross sections in the sub-barrier excitation region, most of such experiments have so far been performed in excitation regions near or above the top of the fission barriers in such nuclei. These measurements therefore explore only the upper part of the fission barrier shapes and determine the heights and the curvature parameters of the potential barrier only near its top. The heights of the fission barriers thus estimated are reasonably reliable. However, extrapolating the same values of the curvature parameters from those near the top of the barriers down to much lower excitations and to a wider range of the deformation regions can be questioned in view of the absence of any compelling physical reasons suggesting that the ground-state spontaneous fission penetrability parameters should be identical to those governing the fission reaction rates at higher excitation energies.

Except for the above difficulty concerning the extrapolation of the curvature parameters to the lower excitations and wider deformation regions, the knowledge of the height and of the curvature parameter is sufficient to sketch the actual shape of the single-humped fission barrier in terms of an inverted parabola. Such a potential parametrization has been used commonly in fission literature in analyzing the observed fission cross sections in order to extract information concerning the heights and curvature parameters of such potential shapes in the corresponding nuclei. Extending such a procedure to the more complicated multihumped fission barriers, the most commonly used potential parametrization is in terms of smoothly joined parabolic segments, each of which represents either a potential barrier or a potential well as the case may be. The potentials as well as their first derivatives are matched at the joining points between the successive parabolic segments. such smoothly joined parabolic segments then represent reasonably realistic double- and triple-humped fission barriers in various actinide nuclei and have been used extensively in recent fission literature over the past two decades.

The use of such smoothly joined parabolic segments in parametrizing the multihumped fission barriers is, of course, only an approximation. There are no strong physical reasons to suggest that the shapes of the multihumped fission barriers must be exactly quadratic in the entire range of the deformations involved during the fission process. Thus, an extrapolation of the curvature parameters of the individual parabolic segments in the deformation regions far from their corresponding extrema (maxima or minima) is open to question. The inertial parameters may also vary significantly with deformation through the barrier. There might also be some other effects of interaction between the deformation mode and other degrees of freedom. As a result, the potential shapes in reality may well be quite different from those obtained using the smoothly joined parabolic segments.

The knowledge of the exact shapes of such multihumped fission barriers is essential to the detailed understanding of the various fission phenomena such as the shape isomeric fission and gamma decay, the spectroscopy of the excited states of the shape isomer, the so-called "isomeric-shelf" observed in the low-energy photofission cross sections, and the observed structure in the near-barrier fission cross sections of the actinide nuclei. The shape isomers are commonly interpreted as the lowest state of the intermediate minimum, or the so-called second well, between the two barriers of the double-humped fission potential shapes in such actinide nuclides. The barrier as a whole also determines the ground-state spontaneous fission half-life of such nuclei. As smoothly joined parabolic segments are a rather convenient and thus commonly used means of parametrizing the multihumped fission barriers, it is therefore of interest, and thus the purpose of this manuscript, to test the adequacy of such a potential parametrization by examining its simultaneous consistency with the various relevant observed fission characteristics of a wide range of the actinide nuclei. The three fission observables that we have chosen for the present investigation are the near-barrier fission cross sections, isomeric half-lives, and the ground-state spontaneous fission half-lives. These three characteristics, taken together, cover the entire range of the excitations as well as all the relevant deformations involved in the subbarrier fission of the actinide nuclei and should thus provide a reasonably reliable means of testing the consistency of parametrizing the multihumped fission barriers in terms of the smoothly joined parabolic segments.

II. METHOD

Bjørnholm and Lynn[1] have listed the detailed sets of the fission barrier parameters for most of the actinide nuclei in Tables XXXI and XXXII of their review paper on the double-humped fission barrier. These parameters have been obtained from analyzing the near-barrier fission cross-section data on the corresponding actinide nuclides. Table XXXII of their review paper[1] summarizes the "best" values of the inner- and outer-barrier heights (E_A and E_B) in a double-humped fission barrier and of the isomeric energies (E_i). They have also suggested three pairs of values of the barrier penetrability parameters ($\hbar\omega_A$ and $\hbar\omega_B$), each pair being common to the even-even, the odd-mass, and the odd-odd actinide nuclides, respectively. These recommended values of the barrier penetrability parameters also reflect the various odd-even effects, as discussed in detail by these authors. As a starting point, we have taken these listed values of the barrier parameters as representing the set of values consistent with the near-barrier fission cross-section data on the corresponding actinides. Using these values of the various parameters, we have then parametrized the corresponding double-humped fission barrier shapes in terms of the three smoothly joined parabolic segments as discussed in detail by Cramer and Nix.[2] Penetrability through such potential shapes has been calculated in Wentzel-Kramers-Brillouin (WKB) approximation. The details of such tunneling calculations have already been reported by us in some of our previous publications.[3-9] To study the simultaneous consistency of the above sets of barrier parameters with the observed fission half-lives, we have calculated the isomeric half-lives and the ground-state spontaneous fission half-lives for our static potential shapes with a constant mass parameter (with respect to the deformation coordinate) using the formalism given earlier by Nix and Walker.[10] Values of the minima of the second well and of its characteristic frequency parameter (E_2 and $\hbar\omega_2$) have been suitably chosen so as to be consistent with the observed isomeric energies, $E_i=(E_2+\frac{1}{2}\hbar\omega_2)$, in our parabolic potential parametrization. Suitable minor variations on the sets of values recommended by Bjørnholm and Lynn for the parameters of the double-humped fission barriers in various actinide nuclei have then been made so as to reproduce the observed isomeric half-lives, and the corresponding ground-state spontaneous fission half-lives have been calculated assuming a constant value of (1 MeV)/$\hbar$ for the frequency (ω_0) of assaults on the fission barrier corresponding to each of the fissioning compound nuclei.

It is useful to recall here that the parameter $\hbar\omega$ in a given parabolic segment, assumed to be harmonic about its extremum, is given by the square root of the ratio between the respective curvature parameter and the effective mass or the inertia parameter at the deformation corresponding to the extremum. As most of the analyses of the observed data on the fission excitation functions have so far been carried out assuming a mass parameter that is taken constant with respect to the deformation coordinate, the parameters $\hbar\omega$ have frequently been loosely termed as the curvature parameters in the fission literature. It should, however, be realized that any large variation in the value of the mass parameter as a function of the deformation coordinate could significantly influence the value of the parameter $\hbar\omega$ in the corresponding deformation region. Following the terminology of Bjørnholm and Lynn,[1] these parameters in the inner- and the outer-barrier regions ($\hbar\omega_A$ and $\hbar\omega_B$) have been called the corresponding "penetrability parameters" in the present work.

A. Analytical expressions for the half-lives

The expressions used in the present work to compute the various fission half-lives have been taken from the earlier work of Nix and collaborators,[2,10] and are briefly summarized here. The spontaneous fission (s.f.) decay half-life from the ground-state (g.s.) level E_0 can be written as

$$\tau_{\text{g.s.}}^{\text{s.f.}}=(\ln 2)(2\pi/\omega_0)[P(E_0)]^{-1} , \tag{1}$$

where ω_0 is the frequency of assaults on the fission barrier, taken as 1 MeV/$\hbar$ for each of the nuclei, and $P(E_0)$ is the penetrability through their corresponding double-humped fission barriers at E_0.

The total isomeric half-life from the isomeric state E_i in the second well can be written in terms of its partial decay half-lives as

$$(\tau_i)^{-1}=(\tau_i^{\gamma})^{-1}+(\tau_i^{\text{s.f.}})^{-1} . \quad (2)$$

In this expression, τ_i^{γ} represents the half-life for the gamma decay from the isomeric state to the ground state of the fissioning compound nucleus. This is a two-step process: first the tunneling through the inner barrier at energy E_i and subsequently the gamma deexcitation to the ground state of the fissioning nucleus. An empirical expression for this decay half-life has been obtained earlier by Nix and Walker,[10] and is given as

$$\tau_i^{\gamma}\cong 10^{-14}[P_A(E_i)]^{-1} \text{ sec} , \quad (3)$$

where $P_A(E_i)$ is the penetrability through the inner barrier at energy E_i.

The spontaneous fission decay half-life from the isomeric state E_i, denoted as $\tau_i^{\text{s.f.}}$ in Eq. (2), depends only upon the tunneling through the outer barrier, and can be written as

$$\tau_i^{\text{s.f.}}=(\ln 2)(2\pi/\omega_2)[P_B(E_i)]^{-1} , \quad (4)$$

where $\hbar\omega_2$ is the characteristic frequency parameter or the vibrational energy in the second-well region, and $P_B(E_i)$ is the penetrability through the outer barrier at energy E_i. The various penetrabilities (P, P_A, and P_B) have been calculated in the present work in the WKB approximation and the details can be seen in some of our earlier publications.[3–9]

III. RESULTS AND DISCUSSION

A. Fission half-lives and isomeric energies

In Table I, we have shown a comparison of the double-humped fission barrier parameters used in the present work with those recommended by Bjørnholm and Lynn[1] for 25 actinide nuclides known to exhibit the shape isomeric phenomenon and for which the measured values of the half-lives and/or isomeric energies are known. The energies E_A, E_2, and E_B are all relative to that of the ground state of the fissioning compound nucleus. Most of the parameter values used in the present work are seen to be within the listed uncertainties in their recommended values by these authors and are thus in excellent agreement. The resulting values of the fission half-lives and of the isomeric energies calculated in the present work using the double-humped fission barriers parametrized by smoothly joined parabolic segments are compared with their corresponding experimentally measured values in Table II. The measured values of the isomeric energies and of the isomeric half-lives have been taken from their compilation in the review paper by Bjørnholm and Lynn.[1] On the other hand, the measured values of the ground-state spontaneous fission half-lives have been taken from their compilation by Vandenbosch and Huizenga in their text[11] on nuclear fission. These values have been updated, where necessary, to be compatible with those listed in the most recent review papers[12–15] on these topics. Our results on the comparison of the calculated fission half-lives and isomeric energies with those measured can be summarized individually for the even-even, odd-mass, and the odd-odd actinide nuclides, respectively, as in the following.

1. Even-even actinides

The agreement between the calculated and the measured values of the fission half-lives and of the isomeric energies for the eight even-even (*e-e*) actinides listed in Table II is indeed remarkable. Not only are the isomeric energies and the isomeric half-lives reasonably reproduced, there is also an excellent agreement on the ground-state spontaneous fission half-lives. Such a striking agreement for all the eight even-even actinides cannot be just a pure coincidence and thus suggests the reasonable accuracy of our knowledge of the fission barrier parameters of such nuclei. This agreement also demonstrates clearly that our parametrization of the corresponding double-humped fission barriers in terms of the smoothly joined parabolic segments is quite adequate as it reproduces the various observed fission characteristics in a wide range of excitations and deformations for such even-even actinides.

2. Odd-mass actinides

Among the 14 odd-mass (*e-o* or *o-e*) actinides listed in Table II, the measured ground-state spontaneous fission half-lives are apparently known for only six such nuclei. This makes it difficult to draw any reasonable conclusions regarding the consistency of our calculated values with those measured. For the isomer energies and for the isomeric half-lives of such nuclei, however, the agreement between the calculated and the corresponding measured values is excellent. For the six odd-mass actinides where the ground-state spontaneous fission half-lives have been measured, we note that the calculated values of this half-life differ from those measured by 5–6 orders of magnitude. The only exception is that of ^{237}Np, where the calculated value of the ground-state spontaneous fission half-life is in reasonable agreement with that measured. While such discrepancies are not totally unexpected in view of the strong exponential dependence of the barrier penetrability and thus of the spontaneous fission half-lives on barrier heights and curvatures, it certainly also reflects on our lack of accurate knowledge of the parameters of the corresponding double-humped fission barriers for such nuclei.

Hindrance factors have long been known to be associated with the spontaneous fission decay of nuclides with an odd number of protons or neutrons relative to the half-lives of their even-even neighbors. The experimentally observed hindrance factors in the region of the actinide nuclei considered in the present work are approximately of the order of 10^5, as summarized recently in a review paper by Hoffman and Somerville.[13] These hindrance factors are usually explained in terms of the so-called specialization energy[16,17] arising from the conservation of spin and parity of the odd particle during the fission process. It is not always possible for the odd nucleon to transfer to another level at a level crossing and still conserve the total spin and parity. When such

TABLE I. Comparison of the double-humped fission barrier parameters for the 25 actinide nuclides used in the present work with those recommended by Bjørnholm and Lynn (Ref. 1).

Compound nucleus even-odd (Z-N)	Source of the barrier parameters	E_A (MeV)	E_2 (MeV)	E_B (MeV)	$\hbar\omega_A$ (MeV)	$\hbar\omega_2$ (MeV)	$\hbar\omega_B$ (MeV)
^{236}U	present work	5.63	2.27	5.53	1.04	0.70	0.60
(e-e)	recommended	5.6±0.2		5.5±0.2	1.04		0.60
^{238}U	present work	5.90	2.06	5.60	1.04	1.00	0.60
(e-e)	recommended	5.70±0.2		5.7±0.2	1.04		0.60
^{237}Np	present work	5.70	2.40	5.40	0.80	1.00	0.52
(o-e)	recommended	5.7±0.2		5.4±0.2	0.80		0.52
^{235}Pu	present work	5.80	2.10	5.10	0.80	1.00	0.52
(e-o)	recommended			5.1±0.4	0.80		0.52
^{237}Pu	present work	5.90	2.10	5.20	0.80	1.00	0.52
(e-o)	recommended	5.90		5.20	0.80		0.52
^{238}Pu	present work	5.60	2.03	5.00	1.04	1.00	0.60
(e-e)	recommended	5.5±0.2		5.0±0.2	1.04		0.60
^{239}Pu	present work	6.00	2.20	5.65	0.80	1.00	0.52
(e-o)	recommended	6.2±0.2		5.5±0.2	0.80		0.52
^{240}Pu	present work	5.57	1.90	5.07	1.04	1.00	0.60
(e-e)	recommended	5.6±0.2		5.1±0.2	1.04		0.60
^{241}Pu	present work	6.10	1.70	5.25	0.80	1.00	0.52
(e-o)	recommended	6.1±0.2		5.4±0.2	0.80		0.52
^{242}Pu	present work	5.50	1.95	5.10	1.04	1.00	0.60
(e-e)	recommended	5.6±0.2		5.1±0.2	1.04		0.60
^{243}Pu	present work	5.70	1.95	5.00	0.80	1.00	0.52
(e-o)	recommended	5.9±0.2		5.2±0.2	0.80		0.52
^{244}Pu	present work	5.55	2.05	5.00	1.04	1.00	0.60
(e-e)	recommended	5.4±0.2		5.0±0.2	1.04		0.60
^{245}Pu	present work	5.40	1.93	5.00	0.80	1.00	0.52
(e-o)	recommended	5.6±0.2		5.0±0.2	0.80		0.52

transfers to the lowest levels cannot take place, the fission barrier for a nucleus with an unpaired nucleon will be higher and wider than for an even-even nucleus.

In addition to increasing the height and the width of the fission barrier, an unpaired odd nucleon also influences the dynamical inertia or the effective mass parameter of the fissioning system. Pair correlations seem to facilitate the inertial response to the changes in shapes of the nuclei undergoing fission. Thus, the systems with unpaired nucleons may be expected to have increased inertia. Urin and Zaretsky[18] and Sobiczewski[19] have made theoretical estimates of the increase due to one unpaired particle and found it to be of the order of 25% in the inertia. This leads to a decrease in the values of the parameters $\hbar\omega_A$ and $\hbar\omega_B$ for the odd-mass nuclides relative to those for their even-even neighbors. Such changes have the effect of increasing the spontaneous fission half-lives of the odd-mass nuclei relative to their even-even neighbors.

The odd-even effects discussed above are already reflected in the three pairs of values of the penetrability parameters $\hbar\omega_A$ and $\hbar\omega_B$ suggested by Bjørnholm and Lynn[1] for the even-even, the odd-mass, and the odd-odd actinide nuclides, respectively. As can be seen in Table I, the recommended values of such parameters are smaller for the odd-mass, and still smaller for the doubly odd, actinides relative to those for their even-even neighbors. The fact that we obtain much longer half-lives in our present calculation for the odd-mass nuclei when using such recommended values in the fission literature seems to suggest that the effect of the odd nucleon on the values of these parameters may have been somewhat overestimated. For example, the relatively smaller value of the outer-barrier penetrability parameter $\hbar\omega_B$ seems to be one of the most significant factors in obtaining rather large values of the ground-state spontaneous fission half-lives for the odd-mass nuclei in our calculations.

3. *Odd-odd actinides*

Among the three odd-odd (*o-o*) actinides listed in Table II, the measured ground-state spontaneous fission

TABLE I. (*Continued*).

Compound nucleus even-odd (Z-N)	Source of the barrier parameters	Double-humped fission barrier parameters E_A (MeV)	E_2 (MeV)	E_B (MeV)	$\hbar\omega_A$ (MeV)	$\hbar\omega_2$ (MeV)	$\hbar\omega_B$ (MeV)
^{239}Am	present work	6.40	2.10	5.22	0.80	1.00	0.52
(o-e)	recommended	6.2±0.3		5.60	0.80		0.52
^{240}Am	present work	6.50	2.30	5.70	0.65	1.00	0.45
(o-o)	recommended	6.5±0.2		5.2±0.3	0.65		0.45
^{241}Am	present work	6.00	1.70	5.05	0.80	1.00	0.52
(o-e)	recommended	6.0±0.2		5.1±0.3	0.80		0.52
^{242}Am	present work	6.78	2.20	5.78	0.65	1.00	0.45
(o-o)	recommended	6.5±0.2		5.4±0.3	0.65		0.45
^{243}Am	present work	5.80	1.80	5.20	0.80	1.00	0.52
(o-e)	recommended	5.9±0.2		5.4±0.3	0.80		0.52
^{244}Am	present work	6.50	2.30	5.70	0.65	1.00	0.45
(o-o)	recommended	6.3±0.2		5.4±0.3	0.65		0.45
^{245}Am	present work	5.90	2.10	5.335	0.80	1.00	0.52
(o-e)	recommended	5.9±0.2		5.2±0.3	0.80		0.52
^{241}Cm	present work	6.60	1.60	4.52	0.80	1.00	0.52
(e-o)	recommended	6.3±0.3		4.3±0.5	0.80		0.52
^{242}Cm	present work	5.70	1.27	4.00	1.04	1.00	0.60
(e-e)	recommended	5.8±0.4		4.0±0.5	1.04		0.60
^{243}Cm	present work	6.70	1.40	4.40	0.80	1.00	0.52
(e-o)	recommended	6.4±0.3		4.30	0.80		0.52
^{244}Cm	present work	5.60	1.67	4.20	1.04	1.00	0.60
(e-e)	recommended	5.8±0.2		4.3±0.3	1.04		0.60
^{245}Cm	present work	6.00	1.90	4.80	0.80	1.00	0.52
(e-o)	recommended	6.2±0.2		5.00	0.80		0.52

half-life is apparently known for only one such nuclide. While the measured isomeric energies and the isomeric half-lives for such nuclides are reasonably reproduced by our calculations, the calculated values of the ground-state spontaneous fission half-lives are extremely large. Such discrepancies in the ground-state spontaneous fission half-lives of doubly odd actinides have already been found in the past in fission literature, as noted in detail, for example, by Nix and Walker[10] for ^{242}Am. There have been suggestions in fission literature that the hindrance factors for the doubly odd nuclei may be the product of the odd-proton and odd-neutron hindrance factors. Howver, as only a few spontaneous fission half-lives for the odd-mass and for the doubly odd actinide nuclei have been measured, few such hindrance factors can be determined. The experimentally observed evidence on such a multiplicative nature of the individual odd-particle hindrance factors for the doubly odd actinides is thus not yet conclusive, as summarized recently by Hoffman and Somerville.[13]

The large discrepancy between our calculated values of the ground-state spontaneous fission half-lives for the doubly odd nuclei and those measured, for example, for ^{242}Am, may again by related to the relatively much smaller values of the penetrability parameters ($\hbar\omega_A$ and $\hbar\omega_B$) as recommended by Bjørnholm and Lynn,[1] and used in our work. An alternative possible explanation of the relatively long ground-state spontaneous fission half-lives obtained in the present work for the odd-mass and for the doubly odd actinide nuclei could be in terms of the different spins of the isomeric and the ground states of the fissioning nucleus. The two such states may therefore "see" or "feel" rather different fission barriers. An increase in the ground-state energy by approximately 1 MeV or lowering the corresponding fission barriers by approximately the same amount may be sufficient to resolve the discrepancies observed on the odd-mass nuclei in the present work. It will, however, not suffice to resolve the considerably large discrepancy on the ground-state spontaneous fission half-life of the doubly

TABLE II. Comparison of the calculated fission half-lives and the isomeric energies with those measured for the 25 actinide nuclides listed in Table I. The energies are relative to those of the ground states of the fissioning compound nuclei. The sources of the experimental results are mentioned in the text.

Compound nucleus	Even-odd character of Z-N	Calculated results τ_i (sec)	E_i (MeV)	$\tau_{g.s.}^{s.f.}$ (yr)	Experimental results τ_i (sec)	E_i (MeV)	$\tau_{g.s.}^{s.f.}$ (yr)
^{236}U	e-e	119×10^{-9}	2.62	9.2×10^{15}	$(125\pm15)\times10^{-9}$	2.3 ± 0.2 2.77[a]	2×10^{16}
^{238}U	e-e	200×10^{-9}	2.56	2.4×10^{14}	$(195\pm30)\times10^{-9}$	2.56	6×10^{15}
^{237}Np	o-e	37.9×10^{-9}	2.90	1.9×10^{19}	$(40\pm12)\times10^{-9}$	2.8 ± 0.3	$>10^{18}$
^{235}Pu	e-o	37.9×10^{-9}	2.60	4.1×10^{18}	$(30\pm5)\times10^{-9}$	2.6 ± 0.4	
^{237}Pu	e-o	127×10^{-9}	2.60	3.7×10^{19}	$(110\pm12)\times10^{-9}$	2.8 ± 0.2	
^{238}Pu	e-e	0.5×10^{-9}	2.53	3.5×10^{10}	$(0.5\pm0.2)\times10^{-9}$	2.7 ± 0.2	$(5\pm0.6)\times10^{10}$
^{239}Pu	e-o	8.8×10^{-6}	2.70	1.9×10^{22}	$(8.1\pm0.8)\times10^{-6}$	2.6 ± 0.2	5.5×10^{15}
^{240}Pu	e-e	4.1×10^{-9}	2.40	1.0×10^{11}	$(3.8\pm0.3)\times10^{-9}$	2.4 ± 0.3	1.2×10^{11}
^{241}Pu	e-o	29.6×10^{-6}	2.20	1.9×10^{21}	$(24\pm1)\times10^{-6}$	1.9 ± 0.3	$\sim6\times10^{16}$
^{242}Pu	e-e	3.3×10^{-9}	2.45	7.3×10^{10}	$(3.6\pm0.6)\times10^{-9}$	2.2 ± 0.3	7.5×10^{10}
^{243}Pu	e-o	69.4×10^{-9}	2.45	9.3×10^{17}	$(60\pm15)\times10^{-9}$	1.7 ± 0.3	
^{244}Pu	e-e	0.4×10^{-9}	2.55	2.3×10^{10}	$(0.4\pm0.1)\times10^{-9}$		$(6.5\pm0.3)\times10^{10}$
^{245}Pu	e-o	88.4×10^{-9}	2.43	7.5×10^{16}	$(90\pm30)\times10^{-9}$		
^{239}Am	o-e	162×10^{-9}	2.60	3.7×10^{21}	$(163\pm12)\times10^{-9}$	2.4 ± 0.2	
^{240}Am	o-o	11.1×10^{-4}	2.80	9.9×10^{32}	$(9.1\pm0.7)\times10^{-4}$	3.0 ± 0.2	
^{241}Am	o-e	2.6×10^{-6}	2.20	6.4×10^{19}	$(1.5\pm0.6)\times10^{-6}$	2.2 ± 0.2	$(2.3\pm0.8)\times10^{14}$
^{242}Am	o-o	13.7×10^{-3}	2.70	1.0×10^{35}	$(14\pm0.7)\times10^{-3}$	2.9 ± 0.2	$>3\times10^{12}$
^{243}Am	o-e	4.8×10^{-6}	2.30	5.4×10^{19}	$(5.5\pm0.5)\times10^{-6}$	2.3 ± 0.2	$(2.0\pm0.5)\times10^{14}$
^{244}AM	o-o	1.1×10^{-3}	2.80	9.9×10^{32}	$(1.0\pm0.15)\times10^{-3}$	2.8 ± 0.4	
^{245}Am	o-e	652×10^{-9}	2.60	2.1×10^{20}	$(640\pm60)\times10^{-9}$		
^{241}Cm	e-o	14.4×10^{-9}	2.10	1.6×10^{19}	$(15\pm1)\times10^{-9}$	2.1 ± 0.3	
^{242}Cm	e-e	40.4×10^{-12}	1.77	8.7×10^{6}	$(40\pm15)\times10^{-12}$	1.9 ± 0.2	7.2×10^{6}
^{243}Cm	e-o	37.9×10^{-9}	1.90	1.5×10^{19}	$(42\pm6)\times10^{-9}$	1.9 ± 0.3	
^{244}Cm	e-e	4.9×10^{-12}	2.17	1.4×10^{7}	5×10^{-12}		1.4×10^{7}
^{245}Cm	e-o	11.3×10^{-9}	2.40	1.1×10^{18}	$(13\pm2)\times10^{-9}$	2.1 ± 0.3	$(1.4\pm0.2)\times10^{12}$

[a]Reference 14.

odd nucleus ^{242}Am. It may be appropriate to recall here that Nix and Walker[10] also could not simultaneously reproduce the isomeric- and ground-state fission characteristics of this doubly odd nucleus in terms of a single fission path in the deformation space and thus proposed the possibility of a two-dimensional potential energy surface in ^{242}Am such that it may contain both a high thin barrier through which the ground-state spontaneous fission proceeds and a thicker two-peaked barrier in which the isomer is located and through which the induced (isomeric) fission proceeds.

B. Fission branching ratios of the shape isomers

Except for the first three actinides listed in Table II, almost all the other known shape isomers decay[20] predominantly by spontaneous fission; the measured half-lives thus relate directly to the penetration of the outer barrier. With a negligible gamma-decay branch, their total lifetime is thus the same as the partial spontaneous fission lifetime. However, for nuclei with a charge number lower than that of plutonium, it is believed that the isomer may have a significant branching ratio for decay by gamma emission. For the first three actinides listed in Table II, namely, ^{236}U, ^{238}U, and ^{237}Np, the decay probability thus has to be corrected for the gamma-decay mode, which dominates in these cases, as revealed by abnormally low partial reaction cross sections for observation of the delayed fission mode. The appreciable competition from the gamma-decay branch in these cases leads to the much reduced values of the relative branching fraction in fission representing the average ratio of the probability of the spontaneous fission and that of the total decay including the radiative deexcitation to the ground state of the fissioning nucleus from the isomeric state. Using our double-humped potential parametrization in terms of the smoothly joined parabolic segments, we have also calculated the fission branching ratio (R) of the shape-isomeric decay, defined in this work as the ratio of the gamma-decay half-life (τ_i^{γ}) to the total half-life (τ_i) of the shape isomer, as given in Eqs. (2)–(4).

Using our values of the double-humped fission barrier parameters listed in Table I, which are reasonably consistent with the values recommended by Bjørnholm and Lynn, we find that the fission branching ratios of the shape-isomeric decay for all the nuclei listed in Tables I and II are almost equal to unity. Such values are indeed expected for most of the above actinide nuclei where the shape isomers predominantly decay by the spontaneous fission and are thus in agreement with the observed characteristics, except, as noted earlier, for the first three actinides listed in Tables I and II. For example, the calculated values of the fission branching ratio (R) of the decay of the shape isomer in these three nuclei, namely, ^{236}U, ^{238}U, and ^{237}Np, have been found to be 0.975, 0.982, and 0.999, respectively. Such values are totally incompatible with the known predominant gamma-decay modes of the corresponding shape isomers in these nuclei and are thus much higher than those observed. The experimentally measured values of such isomeric-decay branching ratios (R) in fission have been found to be approximately of the order of 10^{-1}, 10^{-2}, and 10^{-3}, respectively, for ^{236}U,[21,22] ^{238}U,[23–25] and ^{237}Np.[26]

The first direct observation of the gamma decay of shape isomers was reported by Russo *et al.*[23] for the ^{238}U-shape isomer. They found two gamma transitions of 2.514 and 1.879 MeV from the $J^{\pi}=0^{+}$ ground state in the second well, to the 45 keV, 2^{+} level and 680 keV, 1^{-} level in the first well, respectively. This led to the first precise determination of the corresponding isomeric excitation energy, equal to 2.559 MeV. Kantele *et al.*[27] also later succeeded in observing an $E0$ transition between the isomeric and the ground states of ^{238}U by conversion electron spectroscopy. A new two-detector gamma-ray spectrometry technique was used recently by Kantele *et al.*[28] to reinvestigate the gamma branch of the ^{238}U-shape isomer. In this study, only one gamma transition of 2512.7 keV to the first excited 44.9 keV, 2^{+} state was confirmed. Other lines previously determined by Russo *et al.*[23] were either not visible, or were found to belong to the background. The cross section for the production of the 2.51-MeV gamma ray was also found to be about one-half of that reported by Russo *et al.*[23] The isomeric decay was found to be about 5% by fission, and 95% by gamma rays and conversion electrons. However, almost half of the gamma branch still remains unobserved.[28]

After a number of unsuccessful attempts, a gamma-decay branch of the ^{236}U-shape isomer has also been found recently.[14] The isomer excitation energy has been determined to be 2.77 MeV, and the branching ratio for the gamma decay compared to the delayed fission decay was found to be (8±3). It is also interesting to note that the decay pattern for the shape isomer in ^{236}U is significantly different from that in ^{238}U, with the decay to the 1^{-} state being stronger than to the 2^{+} ground-state rotational level in ^{236}U. This may be due to the different structure of the 0^{+} states in the first well for the two nuclei at the isomeric excitation energy. Gamma-decay branches have so far been uniquely identified only for the above two uranium-shape isomers in the actinide region of the nuclei.

In order to satisfactorily reproduce the observed values of the fission branching ratios for the first three actinide nuclei listed in Tables I and II, we find it necessary to make the inner barriers much more penetrable than those suggested by the $\hbar\omega_A$ values recommended by Bjørnholm and Lynn.[1] For example, in Table III, we have shown a comparison of an alternate set of barrier parameter values obtained in our present work for each of these three actinide nuclei with those recommended by Bjørnholm and Lynn. These alternate sets of barrier parameter values satisfactorily reproduce the fission half-lives, the isomeric energies, and also the fission branching ratios (R) of the decay of the shape-isomeric states in these nuclei, as seen in Table IV, where we have compared our calculated values for these fission characteristics with those expeimentally measured. The agreement for these fission observables is seen to be quite good. The comparison of the barrier parameters in Table III reveals, however, that the values of the inner-barrier penetrability parameter $\hbar\omega_A$ need to be increased by almost 50–70 % over those recommended by Bjørnholm and Lynn for these nuclei to obtain an agreement with the measured fission branching ratios of their shape-isomeric states. Evidences for such higher values of the inner-barrier penetrability parameter $\hbar\omega_A$, meaning, thereby, much more penetrable inner barriers, have indeed been found[29] in the past in fission literature, at least for the even-even uranium isotope, ^{238}U.

We also note that the values of the characteristic frequency parameter or the vibrational energy in the second well ($\hbar\omega_2$) are found (Table III) to be of the order of approximately 600 keV for the two even-even uranium isotopes, as compared to 1 MeV for the odd-mass nucleus ^{237}Np. As these values correspond to the spacings of the vibrational states in the second well of the corresponding double-humped fission barriers in such nuclei, it is of in-

TABLE III. Comparison of an alternative set each of the barrier parameters consistent with the fission branching ratios of the shape-isomeric decays for the three actinides, namely, ^{236}U, ^{238}U, and ^{237}Np, with those recommended by Bjørnholm and Lynn (Ref. 1).

Compound nucleus even-odd (Z-N)	Source of the barrier parameters	Double-humped fission barrier parameters E_A (MeV)	E_2 (MeV)	E_B (MeV)	$\hbar\omega_A$ (MeV)	$\hbar\omega_2$ (MeV)	$\hbar\omega_B$ (MeV)
^{236}U	present work	5.63	2.34	5.53	1.60	0.56	0.58
(e-e)	recommended	5.6±0.2		5.5±0.2	1.04		0.60
^{238}U	present work	5.90	2.25	5.60	1.73	0.62	0.56
(e-e)	recommended	5.7±0.2		5.7±0.2	1.04		0.60
^{237}Np	present work	5.70	2.40	5.40	1.20	1.00	0.43
(o-e)	recommended	5.7±0.2		5.4±0.2	0.80		0.52

TABLE IV. Comparison of the calculated fission half-lives, isomeric energies, and of the fission branching ratios (R) of the shape isomers with those measured for the three actinide nuclei using their corresponding barrier parameter values as listed in Table III. The energies are relative to those of the ground states of the fissioning compound nuclei. The sources of the experimental results are mentioned in the text.

Compound nucleus	Even-odd character of Z-N	Calculated results τ_i (sec)	E_i (MeV)	R	$\tau_{g.s.}^{s.f.}$ (yr)	Experimental results τ_i (sec)	E_i (MeV)	R	$\tau_{g.s.}^{s.f.}$ (yr)
^{236}U	e-e	133×10^{-9}	2.62	0.151	9.0×10^{15}	$(125\pm15)\times10^{-9}$	2.77[a]	0.125[a]	2×10^{16}
							2.3 ± 0.2	0.166[b]	
								0.143[c]	
^{238}U	e-e	195×10^{-9}	2.56	2.8×10^{-2}	9.7×10^{15}	$(195\pm30)\times10^{-9}$	2.56[d]	2×10^{-2}[d]	6×10^{15}
								1×10^{-2}[e]	
								7×10^{-2}[f]	
								5×10^{-2}[g]	
^{237}Np	o-e	44.3×10^{-9}	2.90	2.1×10^{-3}	1.6×10^{19}	$(40\pm12)\times10^{-9}$	2.8 ± 0.3	1.9×10^{-3}[h]	$>10^{18}$

[a]Reference 14.
[b]Reference 21.
[c]Reference 22.
[d]Reference 23.
[e]Reference 24.
[f]Reference 25.
[g]Reference 28.
[h]Reference 26.

terest to inquire if such differences in their values are supported by the relevant cross-section measurements. To obtain any estimates of the spacing of the vibrational states in the second well, it is necessary to resolve at least two successive vibrational resonances in the sub-barrier fission cross sections of such nuclei. There are some evidences of such structures in the even-even nuclei, ^{238}U (Refs. 30–32) and ^{240}Pu,[33–35] indicating values of such spacings as approximately of the order of 500–600 keV. These are in reasonable agreement with their values obtained above in the present work for the two even-even uranium isotopes. No such estimates of the spacings of the vibrational states in the second well of the corresponding double-humped fission barriers in the odd-mass nuclei are readily available in fission literature.

IV. SCOPE AND LIMITATIONS OF PRESENT WORK

The aim of the present work was to test the adequacy of the use of smoothly joined parabolic segments to parametrize the multihumped fission barriers in actinides. We have attempted to test this adequacy by examining the consistency of such a parametrization in simultaneously reproducing the observed values of the three fission characteristics, namely, the near-barrier fission cross sections, isomeric half-lives, and the ground-state spontaneous fission half-lives. These three fission observables were chosen for such a systematic study, because, taken together, they cover the entire range of the excitations as well as all the relevant deformations involved in the sub-barrier fission of the actinide nuclei. Suitable double-humped fission barriers have been constructed using three smoothly joined parabolic segments to calculate the values of the above fission observables for a wide variety of a total of 25 actinide nuclides and the various results and the systematics have been discussed. No triple-humped fission barriers have been considered in the present work as there is not sufficiently conclusive information available as yet in the fission literature on the existence of any fission isomers in the thorium nuclei where such three-humped potential barriers are widely predicted. One interesting new observation[14] in this connection is that of the 17 nsec isomer, found recently in ^{233}Th. There are hints that this shape isomer decays from the second minimum only back to the first minimum.

Before summarizing the main conclusions of this systematic investigation, it is useful here to include a brief discussion of the various assumptions as well as of the consequent limitations of our approach. We have parametrized the relevant double-humped fission barriers corresponding to the various actinide nuclei considered in this work using three smoothly joined parabolic segments along a single-, one-dimensional static fission path in the deformation space with constant mass parameters, and have calculated the corresponding values of the various fission observables for a wide variety of the actinide nuclides. While such assumptions have also almost routinely been made in most of the other analyses of various fission cross-section measurements in the recent fission literature, it is important to realize that some of these assumptions may eventually be shown to be incorrect and that some of the observed structures in certain measurements may, in fact, be the manifestations of either another equally probable fission path in the deformation space and/or due to any significant variations of the inertial parameters along the multidimensional fission potential-energy surfaces. The results of our systematic analysis of a wide variety of the actinide nuclei in terms of single, one-dimensional static fission paths can therefore be useful only in a qualitative sense. The same limitations will also hold for most of the other analyses reported in fission literature over the past five decades where such assumptions have been commonly made. It is about time that at least some of the observed data on fission cross sections as well as on fission half-lives are attempted to be analyzed in terms of multidimensional fission paths (at least, say, a two-dimensional fission barrier) with variable inertia. Such an analysis may help reveal the different qualitative features, if any, obtained using such more complicated and perhaps more realistic fission barrier shapes.

It also seems appropriate here to distinguish between the relative empirical nature of our present work and some of the more detailed calculations of the spontaneous fission half-lives of the actinide nuclei reported in fission literature earlier by Randrup *et al.*,[36,37] and more recently by Ĉwiok *et al.*,[38] Möller *et al.*,[39,40] and by Baran *et al.*[41,42] These authors have calculated the potential-energy surfaces (fission barriers) using the macroscopic-microscopic method,[43] and have either evaluated the inertia tensor using the cranking model or have used a phenomenological expression for the inertia. Spontaneous fission is then treated as a one-dimensional barrier penetration problem in multidimensional deformation space. The action integral is minimized using the variational procedures and the spontaneous fission half-lives are calculated in the WKB approximation. Such half-lives can then be used to evaluate the odd-particle hindrance factors. The scope of the present work is relatively much more limited as outlined earlier in Secs. I and II.

V. CONCLUSIONS

Having outlined the various assumptions and the consequent limitations of our approach in the present work, we can now briefly summarize the main conclusions of our systematic investigation on a wide variety of a total of 25 actinide nuclides as in the following.

(i) No unique sets of double-humped fission barrier parameters can be obtained for any of the actinide nuclei in view of a large number of independent parameters required to parametrize such shapes in terms of the three smoothly joined parabolic segments. For example, six such independent parameters, two for each parabolic segment, are required to parametrize a double-humped barrier. Ten such parameters would be needed to parametrize a triple-humped fission barrier in terms of five such smoothly joined parabolic segments. All these parameters do influence, in some way, the values of the penetrability through such potential barriers, and thus the fission probability as well as the fission half-lives. However, there are some useful constraints which guide us in assigning certain values to the various parameters of the multihumped fission barriers parametrized in terms of the smoothly joined parabolic segments. For example, the observed isomeric energies and any observed vibrational resonances in the sub-barrier fission cross sections help us choose suitable values for the two parameters characterizing the second well (E_2 and $\hbar\omega_2$). For the predominantly spontaneously fissioning shape isomers, the isomeric half-lives are approximately the same as their partial spontaneous fission half-lives determined mainly by the parameters of the outer barrier (E_B and $\hbar\omega_B$). On the other hand, for the lighter actinides with a nuclear charge lower than that of plutonium, the inner-barrier parameters (E_A and $\hbar\omega_A$) significantly affect the values of the fission branching ratios of the decay of the shape isomers. The heights of the two barriers and the overall width of the entire double-humped barrier determine the ground-state spontaneous fission half-lives. Thus, while there are certain definite criteria for varying the values of the various parameters so as to reproduce the observed values of certain fission characteristics, no unique values (or sets) of such parameters can still be given for any of the actinide nuclides. The values of the various parameters of the double-humped fission barriers as obtained in the present work for a variety of the actinide nuclei should therefore be considered purely empirical in nature and no claim is thus made of any possible physical meaning that these may carry.

(ii) The results of our analysis indicate that our parametrization of the double-humped fission barrier in terms of three smoothly joined parabolic segments is quite adequate at least for the even-even nuclei as it reproduces quite satisfactorily the values of the various fission observables for all the eight such actinides considered in the present work. Not only are the isomeric energies and the isomeric half-lives reasonably reproduced, there is also an excellent agreement on the ground-state spontaneous fission half-lives. The values of the various parameters used in the present work for these eight even-even actinides are also in excellent agreement with those obtained from analyzing the near-barrier fission cross-section data on such nuclei as recommended by Bjørnholm and Lynn. Thus, our potential parametrization is also reasonably consistent with the near-barrier fission cross-section data on such even-even actinide nuclides.

(iii) Major difficulties remain, however, for the odd-mass and also for the doubly odd actinide nuclides. Using the values of the various parameters as recommended by Bjørnholm and Lynn, we find that although the isomeric energies and the isomeric half-lives of such nuclei are reasonably reproduced, the calculated ground-state spontaneous fission half-lives turn out to be several orders of magnitude larger than those measured. The discrepancy is especially large for the doubly odd nuclides as noted earlier in Table II for the nucleus ^{242}Am. The difficulty is further compounded by the fact that the measured values of the ground-state spontaneous fission half-lives are known only for 6 of the 14 odd-mass actinides, and, similarly, for only one of the three doubly odd nuclei considered in the present work. Some of the possible explanations for such discrepancies have been discussed earlier in the text. It seems that the unpaired odd nucleons influence rather significantly the characteristics of the fission barrier and also modify the dynamical inertia of the corresponding fissioning system. The simple model of a double-humped fission barrier parametrized in terms of three smoothly joined parabolic segments and with a constant inertia as a function of the deformation coordinate as used in the present work may thus not be adequate enough to fully describe the observed fission characteristics of such nuclides. It is also possible that the ground-state spontaneous fission may proceed through a path entirely different from that for the isomeric (induced) fission in the deformation space and, in that case, our assumption of a single, one-dimensional static fission path with a constant mass parameter as used in the present work may not be fully justified. It seems that the so-called odd-even effects and their possible implications on the fission half-lives and also on the various fission barrier parameters are not reasonably understood at present and that more work is clearly needed to understand why such odd-mass and doubly odd nuclei behave so differently from their even-even neighbors in the Periodic Table. The relative values of the inertial parameters and their variations with deformations may be one of the major causes of such a different behavior.

(iv) The observed data on the fission branching ratios of the decay of the shape isomers in various actinide nuclides are reasonably reproduced by our calculations using the double-humped fission barrier shapes parametrized in terms of three smoothly joined parabolic segments. For the first three actinides listed in Tables I and II where the gamma-decay mode dominates for their corresponding shape isomers, we find that relatively narrower, and thus more penetrable, inner barriers than those recommended by Bjørnholm and Lynn are needed to satisfactorily reproduce the observed data on the fission branching ratios. Whether this is a manifestation of the relatively smaller value of the mass parameter (inertia) in the corresponding deformation region is not quite clear however.

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