Three-hump fission barrier in ^{232}Th

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Subbarrier photofission cross-sections and the fission half-lives have been calculated for ^{232}Th in terms of a suitable three-hump fission barrier model consistent with the suggestion of Möller and Nix. The competition due to the gamma deexcitation to the shape-isomeric state in the second well (and its consequent fission) has been included by introducing an absorptive part in the potential in the second well region. The calculated cross sections reproduce satisfactorily the recently observed "shelf" in the deep-subbarrier energy region and provide a further quantitative evidence in favor of resolving the "thorium anomaly" along the lines suggested by Möller and Nix.

[NUCLEAR REACTIONS Fission, calculated subbarrier photofission cross-sections and fission half-lives using a three-hump fission barrier in ^{232}Th. Results reproduce the observed "shelf" in the cross-sections in deep-subbarrier energy region.]

I. INTRODUCTION

For thorium isotopes the calculated[1] first saddle and second minima of the two-hump fission barriers are approximately 3 MeV lower than the experimental values[2,3] commonly attributed to them. This discrepancy constitutes the well-known "thorium anomaly" in the fission literature. However, Möller and Nix[4] have suggested a possible resolution of this anomaly in terms of a third mass-asymmetric minimum in the fission barriers for thorium isotopes. Since then several experimental evidences[5–8] have also accumulated in favor of a third minimum at deformation much larger than that of the second well. A number of fission penetrability calculations through such three-humped potential barriers have also been reported,[9–12] and it has been shown more recently[12] that the known subbarrier fission characteristics of ^{234}Th are consistent with the hypothesis of a third minimum. The purpose of this manuscript is to consider the subbarrier fission characteristics of the even-even isotope ^{232}Th, where relatively much more information has become available recently and thus could provide a more stringent test of the above hypothesis to resolve the thorium anomaly.

The experimental information available on this isotope in the subbarrier energy region can be summarized as follows:

(1) No fission isomer has been observed to date for this nucleus.

(2) Khan and Knowles[13] have measured subbarrier photofission cross sections of ^{232}Th and found evidence for a slight peak near 5.5 MeV of excitation. Such a structure has also been observed in photofission cross sections of ^{232}Th by Rabotnov *et al.*[14] and to a much lesser extent by Dickey and Axel.[15] A slight kink in the fission probability of ^{232}Th near this energy is also evident in the measurements of Back *et al.*[16] through the reaction ^{230}Th$(t,pf)^{232}$Th.

(3) The deep subbarrier photofission cross section on ^{232}Th reported recently by Bowman *et al.*[17] and by Zhuchko *et al.*[18] shows the presence of a "shelf" in the cross section (see Fig. 3) meaning an abrupt decrease in the slope attributed to delayed fission on the assumption[19] that at these energies γ deexcitation to the fission isomer (ground state in the second well) is competitive with decay by fission from the second well. These cross-section data are available down to about 3 MeV of excitation above the ground state of ^{232}Th, and the presence of the shelf provides the first and the only available evidence so far for the possible existence of a fission isomer in this nucleus at an excitation energy near 3 MeV.[17]

As photofission has long been recognized[20–22] as a very useful means of obtaining information regarding the details of the fission barrier shapes at low excitations, it is evident that a satisfactory analysis of the above subbarrier fission characteristics of ^{232}Th in terms of a three hump fission barrier consistent with the suggestions of Möller and Nix[4] should provide a convincing quantitative evidence in favor of resolving the thorium anomaly along the lines suggested by these authors.

II. FISSION PENETRABILITY CALCULATION

The penetrability through a three-hump fission barrier in ^{232}Th has been calculated in WKB approximation. The potential barrier has been parametrized by smoothly joining five parabolas and is given by

$$V(\epsilon)=E_i \pm \tfrac{1}{2}\mu\omega_i^2(\epsilon-\epsilon_i)^2\,, \tag{1}$$

where the plus sign applies for $i=2$ and 4 and the minus sign for $i=1$, 3, and 5. E_i represent the maxima and minima of the potential, $\hbar\omega_i$ their respective curvature parameters, and ϵ_i the locations of extrema on the deformation axis. $V(\epsilon)$ is taken to be zero at $\epsilon=0$. μ is the inertial mass parameter, assumed to be constant for all values of ϵ and has the dimensions of the moment of inertia,[23] as ϵ, the distortion parameter, is dimensionless. The value of μ used in the calculation is

$$\mu=0.054\,A^{5/3}\hbar^2\ \mathrm{MeV}^{-1}\,. \tag{2}$$

Following the suggestion of Möller and Nix[4] for the possible resolution of the thorium anomaly, we have used a barrier shape (Fig. 1) similar to those suggested by Bhandari[9] earlier by assuming that Back *et al.*[16] have actually determined the parameters of the second and third saddles for the nucleus ^{232}Th from an analysis of their (t,pf) fission data. We have accordingly taken parameters similar to theirs for that part of the three-hump barrier and have fixed the first saddle approximately 2–3 MeV lower than the third saddle as predicted by the microscopic calculations. The bottom of the third well (E_4) has been taken to be rather high so as to be consistent with the experimental evidences[5–8] favoring a rather shallow third minimum and its curvature ($\hbar\omega_4$) has been chosen such as to reproduce a slight peak structure in fission cross section at 5.5 MeV of excitation. The bottom of the second well (E_2) and its curvature have been chosen so as to give an isomeric state at 2.9 MeV, consistent with the isomeric excitation energies of the most actinide nuclei (2–3 MeV) and also with the more recent data of Bowman *et al.*[17]

The details of the penetrability calculation through a three-hump fission barrier in the WKB approximation have been reported earlier.[9] In Fig. 1, as the first saddle is rather low, in effect only a double-humped barrier is encountered in the penetrability calculations in the entire energy range, and we therefore give only such relevant formulas in the following: Defining P_A, P_B, and P as the respective penetrabilities for the inner barrier alone, outer barrier alone, and the entire barrier, it has been shown[24,11] that

$$P=P_AP_B/\{[1+((1-P_A)(1-P_B))^{1/2}]^2\cos^2\nu_2 + [1-((1-P_A)(1-P_B))^{1/2}]^2\sin^2\nu_2\}\,, \tag{3}$$

where the individual penetrabilities (P_A and P_B) in the WKB approximation are given[25] as

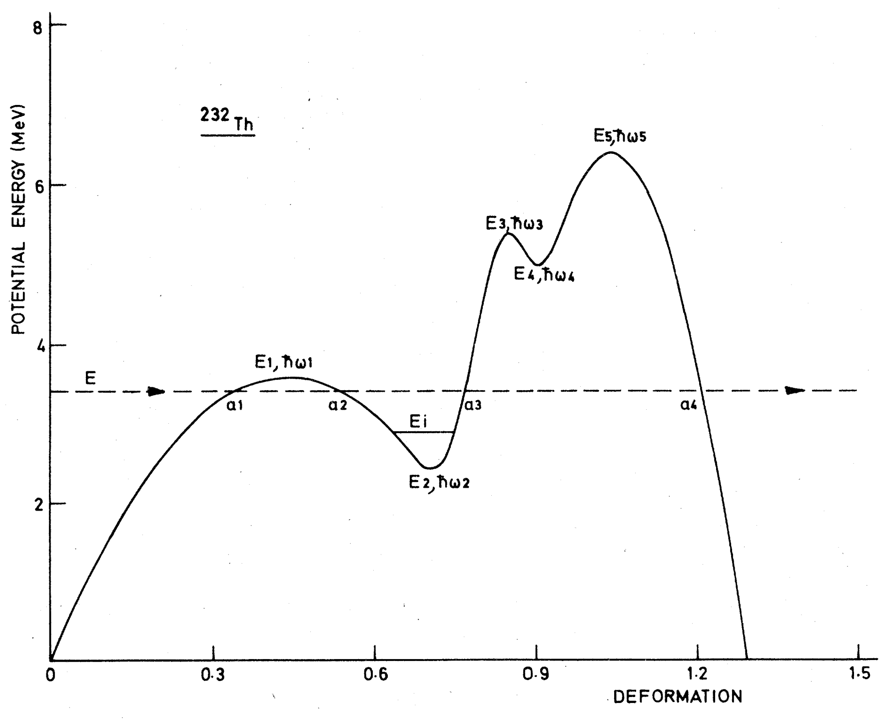

FIG. 1. Three-hump fission barrier in ^{232}Th. The solid horizontal line in the second well corresponds to the shape-isomeric state (E_i). Other symbols are described in the text. The barrier parameters are $E_1=3.6$ MeV, $\hbar\omega_1=0.28$ MeV, $E_2=2.4$ MeV, $\hbar\omega_2=1.0$ MeV, $E_3=5.4$ MeV, $\hbar\omega_3=1.2$ MeV, $E_4=5.0$ MeV, $\hbar\omega_4=1.0$ MeV, $E_5=6.4$ MeV, $\hbar\omega_5=0.67$ MeV, $E_i=2.9$ MeV.

$$P_A = [1 + \exp(2\nu_1)]^{-1}$$

and

$$P_B = [1 + \exp(2\nu_3)]^{-1} \tag{4}$$

for energies below the top of the barriers, and

$$P_A = [1 + \exp(-2|\nu_1|)]^{-1}$$

and

$$P_B = [1 + \exp(-2|\nu_3|)]^{-1} \tag{5}$$

for energies above the top of the barriers. The quantities ν are the integrals in respective regions, as shown in Fig. 1, of the phase

$$K_1(\epsilon) = \{2\mu[E - V(\epsilon)/\hbar^2\}^{1/2} = iK_2(\epsilon)\,, \tag{6}$$

for example,

$$\nu_1 = \int_{a_1}^{a_2} K_2(\epsilon)d\epsilon\,,\ \nu_2 = \int_{a_2}^{a_3} K_1(\epsilon)d\epsilon\,,$$
$$\nu_3 = \int_{a_3}^{a_4} K_2(\epsilon)d\epsilon\,. \tag{7}$$

The classical turning points a_1, a_2, a_3, and a_4 are as shown in Fig. 1 for an excitation energy E.

The competition due to γ deexcitation to the isomeric state (which consequently fissions) has been simulated in the present work by including an absorptive (imaginary) part in the potential in the region of the second well. The calculation again uses the WKB approximation. Using the terminology of Eqs. (3–7) with the added feature that the phase ν_2 now has added an imaginary part ($i\delta$), expressions for the penetrability through the double barrier and for the absorption in the second well are found[27] to be

$$P = [P_A P_B / \{e^{2\delta} + 2[(1-P_A)(1-P_B)]^{1/2}\cos 2\nu_2 + (1-P_A)(1-P_B)e^{-2\delta}\}] \tag{8}$$

and

$$L = P\left(\frac{e^{2\delta}}{P_B} - \frac{(1-P_B)}{P_B}e^{-2\delta} - 1\right), \tag{9}$$

respectively. For a complex potential ($V + iW$) in the region of the second well, the phase factor δ is given as

$$\delta = -\left(\frac{\mu}{2\hbar^2}\right)^{1/2} \int_{a_2}^{a_3} \frac{W(\epsilon)}{[E - V(\epsilon)]^{1/2}} d\epsilon\,. \tag{10}$$

As expected, Eq. (8) reduces to Eq. (3), and L vanishes for $\delta = 0$ ($W = 0$). The flux absorbed in the second well is redistributed in different available channels and contributes a "delayed" fission term indistinguishable from the prompt fission in the measurements of photofission cross sections.[17] The total penetrability of the barrier is then given as

$$P' = P + L\left(\frac{P_B}{P_A + P_B + P_{\gamma_2}} + \frac{\kappa P_{\gamma_2}}{P_A + P_B + P_{\gamma_2}}\right), \tag{11}$$

where P_{γ_2} is the γ deexcitation probability to the shape-isomeric state in the second well and κ is a fraction[18] representing the average ratio of the probabilities of the spontaneous fission and the radiative decay to the first well for the isomeric state. We have accordingly chosen κ as

$$\kappa = \frac{\tau_i^\gamma}{\tau_i^{sf}}\,, \tag{12}$$

where τ_i^γ and τ_i^{sf} are the respective half-lives for γ decay and for spontaneous fission of the shape isomeric state and have been calculated for our static potential shape (Fig. 1) using the expressions given by Nix and Walker.[26]

In Fig. 2 we have shown a sample calculation of L, P, and P' which displays the shelf of Refs. 17–19. For energies greater than approximately 4.7 MeV, $P_B \gg \kappa P_{\gamma_2}$, and the predominant contribution to the observed fission yield is made by the prompt fission penetrability; the difference in the dashed and the solid curve in this energy region is mainly due to the first term in the parentheses in Eq. (11). However, for energies well below 4.7 MeV, the second term in the parentheses in Eq. (11) dominates, as here $P_B \ll \kappa P_{\gamma_2}$, and the predominant contribution to the observed fission yield now is made by the delayed fission penetrability. This results in the shelf in the fission penetrability, as shown in Fig. 2. The magnitude of the penetrability (cross sections) on the shelf is largely determined by the value of the fraction κ, as shown in Fig. 2, and, therefore, defining κ in terms of Eq. (12) puts rather severe constraints on the arbitrariness in the variation of the barrier parameters and leads to a somewhat self-consistent calculation of various observables such as fission half-lives and cross sections. The absorptive part (W) of the potential in the second well has a parabolic shape with respect to the deformation parameter (similar to the real part V) and increases linearly with the excitation energy. The functional form of $W(\epsilon)$ in the region of the second well used in this calculation is

$$W(\epsilon) = -\tfrac{1}{10}[E - V(\epsilon)]\,. \tag{13}$$

III. PHOTOFISSION CROSS-SECTION CALCULATION

The photofission cross section below the neutron threshold is related to the photoabsorption cross section by the expression

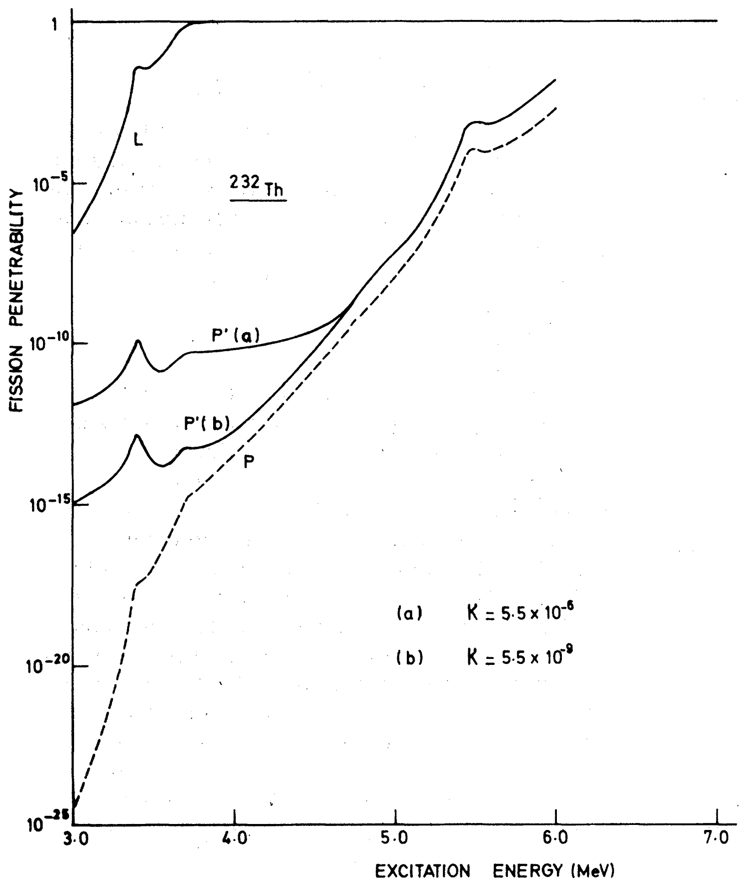

FIG. 2. Fission penetrability versus excitation energy for a three-hump barrier shown in Fig. 1. The symbols L, P, and P' are defined in the text. The curve (a) for P' corresponds to κ defined by Eq. (12) of the text, while, for comparison, the curve (b) corresponds to κ equal to 10^{-3} times its value in (a).

$$\sigma_{\gamma,f}(E)=\left\langle\frac{\Gamma_f(E)}{\Gamma_f(E)+\Gamma_{\gamma_1}(E)}\right\rangle\sigma_{\gamma,\mathrm{abs}}(E)\,. \tag{14}$$

$\Gamma_f(E)$ and $\Gamma_{\gamma_1}(E)$ are the widths for fission and radiative decay in the primary well and are related to the transmission coefficients (P_i) in their respective channels by

$$\langle\Gamma_i\rangle=\langle D\rangle\frac{P_i}{2\pi}\,, \tag{15}$$

where $\langle D\rangle$ is the average spacing between levels of a given total angular momentum and parity, and i represents the individual channel for deexcitation of the excited state in the primary well. Thus, in order to calculate the photofission cross sections, one needs to know the photoabsorption cross section and the relative strength in the fission channel.

Reliable data on the cross section for dipole and quadrupole photoabsorption at low energies are not available. However, Axel[28] has estimated the total photonuclear dipole absorption cross section of heavy elements near 7 MeV excitation as

$$\sigma_{\gamma,\mathrm{dipole}}(E)=5.2\times(E_\gamma/7)^3\times(0.01A)^{8/3}\ \mathrm{mb}\,, \tag{16}$$

and below 3 MeV as

$$\sigma_{\gamma,\mathrm{dipole}}(E)=3.8\times(E_\gamma/7)^2\times(0.01A)^{7/3}\ \mathrm{mb}\,. \tag{17}$$

In Eqs. (16) and (17), E_γ is the photon energy in MeV and A is the mass number of the nucleus. Huizenga and Britt[29] have compared the values obtained from Eq. (16) with the absorption cross sections obtained by extrapolation to lower energies from the photon cross sections of ^{232}Th and ^{238}U, measured by Veyssière *et al*,[30] and find reasonable agreement although the extrapolated cross sections are slightly smaller than those deduced from Eq. (16). The ratio of the quadrupole to dipole absorption cross section is approximately equal to 0.02 for low energy γ rays[29,31]. Therefore, one might expect the photoabsorption cross section to be given by the dipole absorption cross section in the absence of any significant enhancement of the quadrupole component (e.g., a quadrupole giant resonance) in the energy region of our interest. We have therefore used Eqs. (16) and (17) to represent the total absorption cross section.

The relative strength in the fission channel is determined in terms of the penetrabilities corresponding to fission and γ deexcitation channels [Eqs. (14) and (15)]. The fission penetrability is given by Eq. (11) and the radiative penetrabilities (P_{γ_1} and P_{γ_2}) have been calculated using a semiempirical expression given by Bowman[19]:

$$P_\gamma = 2\pi \times 4.1 \times 10^{-7} \times \exp(1.6E_\gamma)\,. \tag{18}$$

This expression gives a radiative penetrability consistent with the dipole radiative transmission coefficient calculated in Ref. 32. We have therefore restricted ourselves only to the dipole channel corresponding to $J^\pi = 1^-$ in the present calculation and have also not included the competition between $K=0$ and $K=1$ channels for the simple reason that most of this calculation is in the deep-subbarrier region where only the lowest barrier corresponding to $K=0$ will be most significant. This has been shown recently in Ref. 15, where the fission transmission coefficient for ^{232}Th corresponding to $K=1$ channel is approximately 2 to 3 orders of magnitude smaller than that for $K=0$ channel.

IV. RESULTS AND DISCUSSION

Using a suitable three-hump fission barrier (Fig. 1) in ^{232}Th, we have calculated "deep" subbarrier photofission cross sections as well as the fission half-lives. Figure 3 shows that the observed shelf in the cross sections is reasonably reproduced. For energies greater than 5 MeV, the calculated cross sections are somewhat larger than those measured. This is expected, as the neutron competition has not been included in our calculation. The calculated curve shows resonance structure in the cross sections near 3.4 and 5.5 MeV of excitation. The absorptive part of the potential in the second well region does broaden these peaks somewhat, as to be expected, but the structure should nevertheless be readily observable on

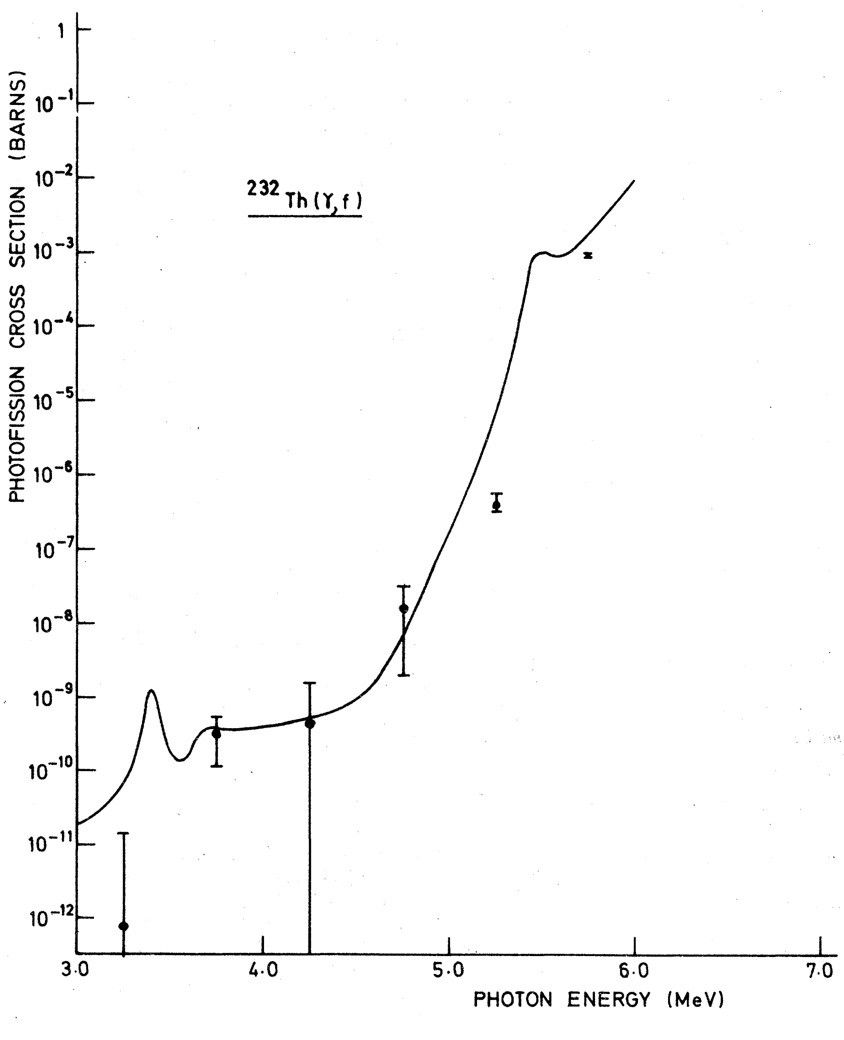

FIG. 3. A comparison of the calculated (solid line) subbarrier photofission cross sections of ^{232}Th with those measured by Bowman *et al.* (Ref. 17).

top of the shelf. This is because when the excitation energy is appropriate for the transmission resonance, there is a large amplitude for the wave function in the second well which augments both fission and γ decay equally. Thus the fission output, whether prompt or delayed, is amplified at the resonance energy. A precise and good-resolution angular anisotropy measurement in the energy region of the shelf might help in isolating the resonance structure corresponding to low-lying vibrational states in the second well. Such a measurement may also yield useful information on the complicated concept of a "transition-state nucleus" in the framework of multihump fission barriers and may help in answering whether K is preserved during the barrier penetration.

We have also calculated ground-state spontaneous fission half-life and the isomeric half-life using the three-hump fission barrier (Fig. 1) in ^{232}Th. These estimates for the fission half-lives are, however, only of qualitative value as these have been calculated using a static potential shape with a constant mass parameter. It has been shown by Pauli and Ledergerber[33] that significantly different results could be obtained when dynamical effects are taken into account. Assuming a curvature ($\hbar\omega$) of 1.2 MeV for the primary potential well the value obtained for the ground state spontaneous fission half-life corresponding to the three-hump barrier of Fig. 1 in our calculation is equal to 1.5×10^{28} yr. There is no precise information available in the literature on the measured ground state spontaneous fission half-life of ^{232}Th. Flerov *et al.*[34] found it to be $>10^{21}$ yr.

Similarly, assuming an isomeric state at 2.9 MeV, the static potential shape shown in Fig. 1 leads to the following values for different components of the isomeric half-life:

$$\tau_i^{\gamma}=76.9\times10^{-9}\ \mathrm{s}\,,$$

$$\tau_i^{sf}=13.9\times10^{-3}\ \mathrm{s}\,,$$

$$\tau_i^{total}=76.9\times10^{-9}\ \mathrm{s}\,.$$

These estimates clearly suggest that the shape-isomeric state in ^{232}Th decays predominantly through γ-decay to the first well. This may explain why no fission isomer has been observed to date for this nucleus.

In conclusion, we can state that the observed sub-barrier fission characteristics of ^{232}Th can be reasonably explained in terms of a three-hump fission barrier consistent with the suggestion of Möller and Nix,[4] and this work therefore provides a further quantitative evidence in favor of resolving the thorium anomaly along the lines suggested by these authors.

V. ACKNOWLEDGMENTS

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