Intermediate Structure in the Photofission Cross Section of ^{232}Th

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Intermediate structure has been observed in the photofission cross section of ^{232}Th measured with a photon energy resolution < 500 eV. The gamma rays, variable in energy, were obtained from the (p,γ) reaction on several nuclei. The average spacing of the observed photofission resonances at an excitation energy of 6.16 MeV is 1.6 ± 0.4 keV. The average areas of the resonances are compared with theoretical expectations for a double-humped and a triple-humped barrier.

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The properties of the fission barriers of actinide nuclei have been, in large measure, derived from studies of structure in the fission cross section at subbarrier excitation energies.[1] Much attention has been given to the question of the shape of the barrier in thorium isotopes, where barrier calculations have been in disagreement with experimental data.[1] The existence of undamped vibrational resonances in the ^{230}Th(n,f) and ^{232}Th(n,f) cross sections at excitation energies above 5.8 MeV has implied, within the confines of the double-hump model, an inner barrier height $E_A \approx 6$ MeV and a secondary well with a minimum at $E_{\rm II} \approx 4.5$ MeV. On the other hand, theoretical calculations, generally successful in predicting barrier heights for heavier actinides, have yielded[2] $E_A \approx 4$ MeV for thorium isotopes. This discrepancy has been known as the thorium anomaly.

A solution was suggested by Möller and Nix[2] who obtained in their calculations for light actinides a low inner barrier and a shallow third well at the deformation of the outer barrier. Experimental evidence for such a triple-humped barrier has been provided[3] by investigations of the structure within the vibrational resonances in ^{231}Th and ^{233}Th.

Subbarrier structure has been observed in many nuclei, mainly in high-resolution measurements of neutron-induced fission. However, such measurements cannot be done when the neutron separation energy exceeds the height of the barrier. This is the case in ^{232}Th, a nucleus whose barrier structure is expected[2] to be similar to that of its neighbors. A way to reach subbarrier energies in ^{232}Th is provided by photofission.

Photon-induced fission takes place only from states which can be reached by electric dipole and, with a much smaller probability, electric quadrupole excitations. This makes identification of spins and parities much easier than in the case of particle-induced fission where a much wider range of angular momenta is possible.

The main difficulty in photofission experiments is in obtaining monochromatic photons of variable energy and adequate intensity. Compton scattered as well as direct gamma rays from slow neutron capture, bremsstrahlung, and tagged bremsstrahlung have been used.[4,5] The last technique in its improved version provides an energy resolution of 12–14 keV for 6-MeV photons.

In this paper we present results of measurements of the photofission cross section of ^{232}Th carried out with a photon energy resolution < 500 eV. These measurements for the first time clearly reveal narrow intermediate structure resulting from the excitation of compound states in the second well of the fission barrier of ^{232}Th. Similar structure has been found in neutron and charged-particle induced fission of various nuclei.[1] Intermediate structure has not been observed before in photofission.

A novel technique in which the photons are obtained from resonances in the (p,γ) reaction has been employed.[6] The experimental setup is shown schematically in the inset of Fig. 1. A collimated proton beam of average current ~ 150 μA from our Dynamitron strikes a water-cooled target mounted in a vibrating target assembly. The emerging gamma rays are allowed to fall on sandwiches of 30-mg/cm^2 thorium foils and 8-μm Kimfol films which serve as track recorders of the photofission fragments. The sandwiches are placed on a cylindrical surface coaxial with the proton beam. The energy of the gamma rays varies with the angle θ relative to the beam as a result of the Doppler shift. The average energy dispersion is ~ 200 eV/deg. An efficiency-calibrated Ge(Li) detector (not shown) located at $\theta = 90°$ serves to determine the absolute intensity of the gamma rays.

The developed[7] Kimfol films are scanned for fission tracks by a vidicon camera[8] which determines the positions of the tracks.

The photofission cross section is shown in Fig. 1 versus the gamma-ray energy. To compute the cross section, the efficiency of the Kimfol film[7] for counting fission fragments as a function of the depth in the foil from which the fragments emerge and the angle at which they enter the film was determined. The efficiency was folded with the angular distribution[9] of the fragments.

Resonances in ^{29}Si, ^{42}Ca, ^{34}S, and ^{25}Mg, yielding gamma rays of 6180, 6172, 6140, 6073, and 5871 keV at 90° to the proton beam were used. The proton energies were well below the (p,n) thresholds of all targets. The intensity of photons from undesired branching did not exceed 10% in any of the cases. To match the spectra taken at the average photon energies of 6180 and 6172 keV it was important to measure accurately the energy difference between these two lines. This was found to be 7.7 ± 0.5 keV. The uncertainties in the magnitude of the cross section at the matching point do not exceed 15%. Unfortunately, with the present setup it was impossible to obtain an overlap of the two spectra. The instrumental gamma-ray energy uncertainty was ~ 200 eV in the runs with ^{42}Ca and ^{34}S targets and < 500 eV in the remaining two runs. The natural width of the ^{29}Si resonance is[10] 179 ± 5 eV. The widths of the other resonances are unknown but an upper limit of 1 keV can be obtained from their excitation curves. From the observed structure in Fig. 1 the overall photon energy resolution is seen to be better than 500 eV.

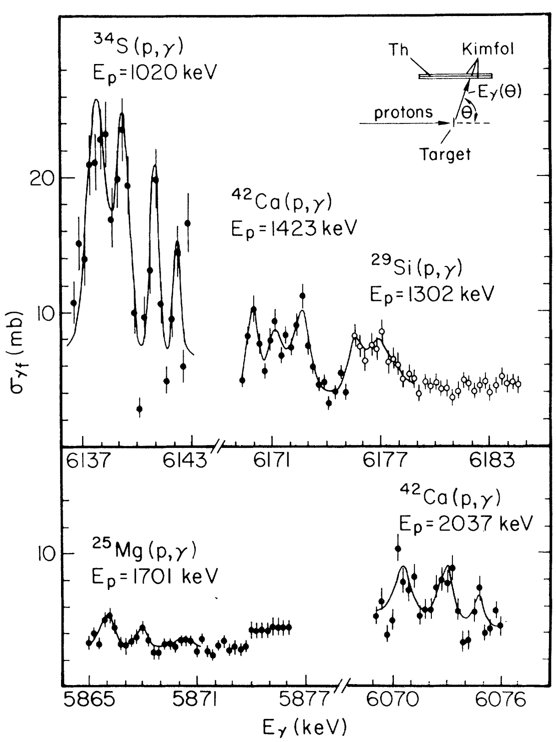

FIG. 1. Photofission cross section of ^{232}Th. The vertical bars represent counting statistical errors only. The solid lines were obtained by Gaussian fitting. The inset in the right upper corner shows the geometrical setup of the experiment.

The average values of the cross sections over the photon energy range for each proton resonance are in good agreement with the results of Dickey and Axel[4] and Caldwell *et al.*[11] They are somewhat higher than those of Knowles *et al.*[5]

Well-established peaks appear in all spectra. The probability that these peaks represent resonances in the photon absorption cross section is negligible given the fact that this would require for each resonance a ground-state radiative transition width ~ 50 times larger than the average. It should also be noted that no structure has been found at higher excitation energies.[6] We therefore assume that the underlying states are compound levels of spin and parity 1^- in the second well of the barrier.

The average spacing of the nine peaks at 6.16 MeV is 1.6 ± 0.4 keV. From the constant temperature level-density formula with parameters given by Bjornholm and Lynn[1] we find the energy of the second minimum $E_{\rm II} = 2.8 \pm 0.1$ MeV, in good agreement with previous determinations.[4,12]

The average widths W of the observed resonances at each excitation energy can be computed from the energy dependence of the fission probability.[13] The results listed in Table I are consistent with the experimental widths within the limits determined by the photon energy resolution.

The average area of a class-II compound resonance at an excitation energy below the neutron

TABLE I. Average areas and widths of resonances.

E (MeV)	A (b·eV) Expt	Calc[a]	Calc[b]	W (eV) Calc[a]	Calc[b]
6.17	5.1 ± 1.7	5.0	4.7	257	240
6.14	13.0 ± 4.0	5.7	11.0	304	493
6.07	3.1 ± 1.5	7.2	7.9	421	434
5.87	0.9 ± 0.4	1.3	1.2	95	97

[a]Double-humped barrier.
[b]Triple-humped barrier.

separation energy can be written in the form[1]

$$A=\sigma_\gamma \pi^{1/2} D_{\rm I}\Gamma_{{\rm II}f}\Gamma_{{\rm II}c}(\pi\Gamma_{\rm II}^2\Gamma_\gamma^2+2D_{\rm I}\Gamma_{{\rm II}c}\Gamma_{{\rm II}f}\Gamma_\gamma)^{-1/2}S,$$

where σ_γ is the average gamma-ray absorption cross section, $D_{\rm I}$ is the average level spacing of class-I compound states, $\Gamma_{{\rm II}c}$ and $\Gamma_{{\rm II}f}$ are the coupling width and fission width of class-II compound states respectively ($\Gamma_{\rm II}=\Gamma_{{\rm II}c}+\Gamma_{{\rm II}f}$), Γ_γ is the average total gamma-decay width of class-I compound states, and S is the width fluctuation factor.

If there are several fission channels of a given spin and parity J^π differing in the spin projection on the fission axis K and having incompletely damped vibrational resonances in the second well at excitation energies $E_{{\rm II}v}^{(K)}$, the widths $\Gamma_{{\rm II}c}$ and $\Gamma_{{\rm II}f}$ are[13]

$$\Gamma_{{\rm II}c,f}=(D_{\rm II}/2\pi)\sum_K \Gamma_{A,B}^{(K)}\Gamma_W[(E-E_{{\rm II}v}^{(K)})^2+(\Gamma_{{\rm II}v}^{(K)}/2)^2]^{-1},$$

where $D_{\rm II}$ is the class-II compound level spacing, $\Gamma_{A,B}^{(K)}=T_{A,B}^{(K)}\hbar\omega_{\rm II}^{(K)}/2\pi$, $\hbar\omega_{\rm II}^{(K)}$ is the level spacing of the vibrational states, Γ_W and $\Gamma_{{\rm II}v}^{(K)}$ are their damping and total widths, respectively, $T_A^{(K)}$ and $T_B^{(K)}$ are the penetrabilities for channel K of barriers A and B, respectively.

If the barrier for channel K has a third well with a vibrational resonance at $E_{{\rm III}v}^{(K)}$, then $T_B^{(K)}$ in the expressions above has to be replaced by

$$T_{BC}^{(K)}=\Gamma_{{\rm III}B}^{(K)}\Gamma_{{\rm III}C}^{(K)}[(E-E_{{\rm III}v}^{(K)})^2+(\Gamma_{{\rm III}v}^{(K)}/2)^2]^{-1},$$

where $\Gamma_{{\rm III}B,C}^{(K)}=T_{B,C}^{(K)}\hbar\omega_{\rm III}^{(K)}/2\pi$, and $\Gamma_{{\rm III}v}^{(K)}$ and $\hbar\omega_{{\rm III}v}^{(K)}$ are the width of the resonance, and the level spacing of the vibrational states in the third well, respectively.

For parabolic barriers $T_i^{(K)}=\{1+\exp[2\pi(E_i^{(K)}-E)/\hbar\omega_i^{(K)}]\}^{-1}$ where $E_i^{(K)}$ and $\hbar\omega_i^{(K)}$ are the heights and curvatures of the first ($i=A$), second ($i=B$), and third ($i=C$) barriers for channel K.

The average areas of the observed resonances at the four excitation energies are compared in Table I with the values calculated for a double-humped and a triple-humped barrier. The calculations were done with the barrier parameters listed in Table II and[4] $\sigma_\gamma=25$ mb and $\Gamma_\gamma=31$ MeV at 6 MeV.

The information on the gross structure in the fission cross section, which is necessary for the area calculations, is somewhat uncertain. A broad resonance at 6 MeV has been well established.[4,5] In addition, Knowles *et al.*[5] reported resonances at 5.92 and 6.11 MeV, each ~ 50 keV wide. These resonances cannot be clearly discerned in the spectra of Dickey and Axel[4] and those of Janszen *et al*,[15] the latter obtained with an energy resolution of 17 keV. Therefore, in the calculations with the double-humped barrier we assumed a single resonance with a damping width of 0.2 MeV.

The results are in agreement with the experimental values when the areas at 6.14 and 6.17 MeV are averaged together and represented by a single value at 6.16 MeV, reflecting the possibility that the difference between the observed areas at the two energies could be due to width fluctuations.

The barrier parameters listed in Table II are close to the values obtained by fitting the measured[4,7] photofission cross sections between 5.5 and 11.5 MeV. It should be noted, however, that fits to average cross sections do not, in general, allow a unique determination of barrier parameters. The present results provide direct evidence for an inner barrier substantially higher than the one predicted by Möller and Nix. (The uncertainties in the 0^- barrier heights are ± 0.2 MeV.) They also provide a unique determination of the depth of the second well.

In the calculations with the triple-humped barrier the narrow resonances reported by Knowles *et al.* were taken into account and assumed to correspond to vibrational states in the third well. To obtain a better fit to our data we increased the energy of the higher resonance to 6.12 MeV. The broad 6-MeV resonance was assumed to be located as before in the second well.

Agreement with the measured areas is somewhat better than in the case of the double-humped barrier. The increase in the resonance areas at 6.14 MeV can be understood as resulting from enhance-

TABLE II. Fission barrier parameters.

	Barrier heights (MeV)				
	Double-hump[a]		Triple-hump[b]		
K	E_A	E_B	E_A	E_B	E_C
0^-	6.15	6.55	6.15	6.55	6.9
1^-	6.55	6.85	6.55	6.95	7.5

[a] $\hbar\omega_A=\hbar\omega_{\rm II}=0.9$ MeV; $\hbar\omega_B=0.65$ MeV for both K barriers. These values supersede the slightly different ones reported earlier (Ref. 14).

[b] $\hbar\omega_A=\hbar\omega_{\rm II}=0.9$ MeV; $\hbar\omega_B=1.4$ MeV; $\hbar\omega_{\rm III}=1.0$ MeV; $\hbar\omega_C=1.2$ MeV for both K barriers.

ment of the fission widths of the class-II compound states because of their coupling to the 6.12-MeV resonance. However, because of the limited extent of the present data and large errors of the measured areas, it is impossible to conclude with certainty whether the observed increase confirms the 6.12-MeV resonance and, consequently, whether a third well exists in the fission barrier of ^{232}Th. More extensive measurements at subbarrier energies, using properly chosen (p,γ) resonances, are necessary to reach such a conclusion.

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