Correlation between angular anisotropy and fragment mass in 15 MeV proton-induced fission of ^{232}Th

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(Received 26 May 1981)

The angular distributions of 15 fission fragments were radiochemically obtained in the 15 MeV proton-induced fission of ^{232}Th. It was found that the angular anisotropies of symmetrically divided fission fragments were smaller than those of asymmetrically divided fragments. With the assumption of two kinds of saddle points, one leading to a symmetric mass division and the other to an asymmetric division, the observed angular anisotropies could be well reproduced.

[NUCLEAR REACTIONS, FISSION Angular distribution in 15 MeV proton-induced fission of ^{232}Th, Bohr's channel theory, two kinds of saddle point configurations, mass division.]

I. INTRODUCTION

According to Bohr[1] the angular distribution of fission fragments are determined by the quantum states at the saddle point with the assumption that (i) the K quantum number is conserved during the descent from saddle to scission and (ii) the fission fragments separate along the nuclear symmetry axis. Angular distributions of fission fragments have been extensively studied,[2] and, by now, Bohr's theory may be considered to be well established. Therefore, if final mass divisions or, at least, the mode of mass division, namely, symmetric or asymmetric, are closely related to the saddle point configuration, there must exist some correlation between the angular distributions of fission fragments and fragment masses. This correlation, however, can be observed only when there are meaningful differences in the quantum states at the saddle, each corresponding to a different mass splitting. Vandenbosch *et al.*[3] reported that no dependence of angular anisotropies on fragment mass could be observed. A similar result is also reported within the experimental error by Vorob'eva *et al.*[4] for the 3 MeV neutron-induced fission of ^{232}Th. Flynn *et al.*[5] investigated the alpha particle-induced fission of ^{209}Bi, and found no mass dependence of the angular anisotropy. However, they found that the anisotropy of ^{83}Br was 10% smaller than those of other masses, and they conjectured that this anomalous behavior of ^{83}Br would be related to the 50N shell and also to the higher excitation energy required to form ^{83}Br. Cohen *et al.*[6] reported in 1955 that the angular distributions of fission fragments in 22 MeV proton-induced fission of ^{232}Th were dependent on fragment masses. In this system both symmetric and asymmetric fission products exist in comparable magnitude. In the so-called medium energy fission, however, there are possibilities of multiple chance fissions, and the analysis of the data becomes more complicated.

Möller and Nilsson[7] suggested that there might be two kinds of saddle configurations in the fission of ^{236}U, one with reflection symmetry with respect to the axis perpendicular to the nuclear symmetry axis and the other with reflection asymmetry, and that the fission barrier height for the symmetric saddle would be a few MeV higher than the one for the asymmetric saddle. They also hinted that the final mass division mode could be closely related to those two kinds of saddle point configurations. If there truly exist two kinds of saddles and if they control the final mass division mode, fragment mass dependence of angular anisotropies should be observable in such a fission system where (i) both symmetric and asymmetric fission products are equally observed and (ii) an appreciable difference in anisotropies is expected from Bohr's channel theory for the fragments that have experienced different kinds of saddles.

In the present work the correlation between the angular distribution of fission fragment and fragment mass was examined in a wide fragment mass range for the 15 MeV proton-induced fission of ^{232}Th, which is theoretically predicted to have two kinds of saddle configurations. (Compound nuclei in the present system would be $^{231-233}$Pa which are odd-even or odd-odd nuclei and there is no theoretical prediction, although it exists for even-even nuclei around $Z=91$.[8]) We also investigated in a separate paper[9] detailed excitation functions of (p,xn) reactions and those of fission fragments, and deduced by statistical calculation the degree of the contributions of multiple chance fission. The angular anisotropies observed in the present work were compared with the theoretical prediction with the assumption of two kinds of saddle points.

II. EXPERIMENTAL

Angular distributions of fission fragments in 15 MeV proton-induced fission of ^{232}Th were measured by the recoil-catcher foil method. A thorium target was prepared by electroplating onto a 10 μm-thick nickel foil from isopropanol solution at 900 V.[10] The thickness of the thorium target ranged from about 280 to 350 $\mu g/cm^2$. The target and the catcher foil assembly is schematically drawn in Fig. 1. An aluminum catcher foil of 8 mg/cm^2 thickness was put on the wall of a semicylindrical brass holder of 113 mm in length and a 41.5 mm radius. The thorium target was attached to the target holder which was inclined at an angle of 45° to the beam direction in order to lower the energy loss of fission fragments within the target when they were emitted at right angle to the beam direction. The proton beam was collimated with a 5 mm-diameter graphite collimator. The collimator, the target holder, and the brass holder with an aluminum catcher foil were screwed up on a flat 1 cm-thick aluminum plate, and set in the scattering chamber. The proton-bombardment was performed for 5–12 h at the cyclotron of the Institute of Physical and Chemical Research. The beam intensity was monitored with a Faraday cup equipped with a current integrator in order to estimate the saturation factor in genetic relationships. The beam current was about 1 μA. Thorium of about 500 μg was attached to the back wall of the brass holder at 3 cm above the proton beam line in order to examine the effect of fission induced by neutrons. It was found that no appreciable fission was induced by neutrons.

After bombardment the aluminum catcher foil was taken out and sliced to strips of appropriate widths corresponding to some solid angles. The mean angle of each strip is presented in Table I, where $\Delta\theta$ corresponds to the width of the strip, namely, the difference between the geometrically possible maximum and the minimum angles. For θ smaller than 45°, the catcher-foil portion corresponding to the azimuthal angles of $\phi=0$ to π was used, whereas for θ larger than 45°, only the portion corresponding to the angles $\phi=0$ to $\pi/2$ was employed, since the energy loss of the fission fragment within the target might become appreciable for the larger ϕ. The strips of the aluminum catcher foil were first weighed in order to correct for the error caused by imprecise cutting of the catcher foil. It was found that the cutting ambiguity was not so large, say, a few percent. Then the strips were folded to the same area so that the geometrical efficiencies are not varied with samples in the direct γ-ray spectrometry. After the direct γ-ray measurement, chemical separation[11,12] was performed for the determination of low fission-yield products.

The γ-ray spectrometer system was based on a 40-cm^3 Ge(Li) detector equipped with a 2048 channel pulse height analyzer except for the case of ^{136}Cs. For the measurement of γ rays from ^{136}Cs, an extremely low-background counter system of the Institute for Cosmic Ray of the University of Tokyo was employed.[13] The measured radioactivi-

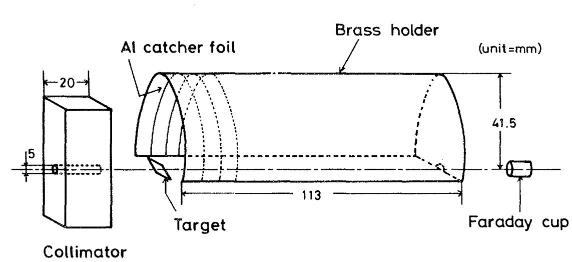

FIG. 1. Target and catcher foil assembly used in the measurement of angular distributions of fission fragments in 15 MeV proton-induced fission of ^{232}Th.

TABLE I. Mean angles in the measurement of angular distributions of fission fragments in 15 MeV proton-induced fission of ^{232}Th.

Angle (θ) (deg)	Angle width ($\Delta\theta$) (deg)
4.3	8.7
10.0	7.7
14.3	6.8
18.1	7.0
22.3	7.3
26.2	8.0
30.0	8.0
33.7	9.5
36.9	8.3
39.6	8.9
42.5	9.7
45.9	10.5
49.6	11.4
53.8	12.2
58.5	13.1
63.7	13.8
69.4	14.3
75.6	14.6
82.1	14.4

ties were corrected for decay, genetic relationships, chemical yield, and cutting ambiguity so that the relative radioactivities at the end of bombardment could be obtained. The experiments were repeated eight times to obtain precise angular distributions.

III. RESULTS AND DISCUSSION

The relative radioactivity of a nuclide in each strip was normalized to give a fraction of the total radioactivity of the nuclide so that results of different runs could be directly compared to each other. The normalized fractions were smoothed by three-points smoothing assuming that the form of angular distributions of fission fragments is $W(\theta)=a+b\cos^2\theta$. The shape of $W(\theta)=a+b\cos^2\theta$ was fitted to the smoothed angular distribution of each fission product by the weighted least-squares method. The results are shown in Fig. 2, and the curves fitted by the least-squares method are given by dashed lines. In this figure, the error of each point is not shown in order to avoid complexity, but a typical one is presented by a vertical line.

Although the data of ^{105}Ru, ^{127}Sb, and ^{136}Cs were rather scattered as shown in Figs. 2(f), (i), and (l) due to the large counting errors, there were only a few points that deviated from the fitted curve by more than one standard deviation (1σ), and the degree of fitting seemed fairly good. The results of fitting are tabulated in Table II, where the angular anisotropy is defined as $1+b/a$, that is, $W(0°)/W(90°)$. The angular anisotropy is plotted as a function of fragment mass number in Fig. 3, which clearly shows the dependence of anisotropy on the mass number (A) of fission products. The anisotropy has a minimum near $A=115$, namely, the anisotropy decreases with mass number up to $A=115$, and then increases with A. However, the increasing trend is likely to level off at large mass numbers. When the asymmetry degree of mass division is defined as the ratio of the mass of a heavy fragment to the mass of the complementary light fragment, the correlation between the angular anisotropy and the fragment mass can be shown as in Fig. 4. It was assumed that the number of neutrons emitted from primary fragments was two.[6,14] From this figure, it is observed that the anisotropies of symmetrically divided fission products are considerably smaller than those of asymmetrically divided ones, while the anisotropies are of intermediate value for the intermediate mass division. The anisotropy of symmetrically divided products is about 1.06 ± 0.03, which is the average value of $^{115}Cd^g$ (1.08 ± 0.02) and ^{112}Pd (1.03 ± 0.02), and the anisotropy for asymmetrically divided products is about 1.38 ± 0.05, which is the average of ^{92}Sr (1.36 ± 0.02), ^{141}Ce (1.42 ± 0.04), ^{143}Ce (1.36 ± 0.06), and ^{147}Nd (1.37 ± 0.06). The anisotropies of ^{91}Sr and ^{88}Kr were appreciably smaller, 1.20 ± 0.01 and 1.28 ± 0.02, respectively, compared with the average value for asymmetrically divided products. The reason is possibly due to the fact that krypton isotopes in forward angles escape from the aluminum catcher foil by thermal diffusion[15] because of heating of the aluminum catcher foil at forward angles by scattered protons. Krypton isotopes are precursors of strontium isotopes in fission, and the reason for the effect of migration in ^{91}Sr being larger than that in ^{92}Sr is perhaps the difference in half-lives of ^{91}Kr ($T_{1/2}=8.6$ sec) and ^{92}Kr ($T_{1/2}=1.84$ sec). Owing to the shorter half-life, ^{92}Kr might have decayed to ^{92}Rb before migration, thus giving a correct anisotropy.

It is worth noting that the anisotropy of ^{136}Cs is nearly equal to those of the neighboring nuclides, although ^{136}Cs is one of the shielded nuclides. The shielded nuclide ^{136}Cs produced in thermal neutron fission of ^{233}U and ^{235}U is known to have smaller

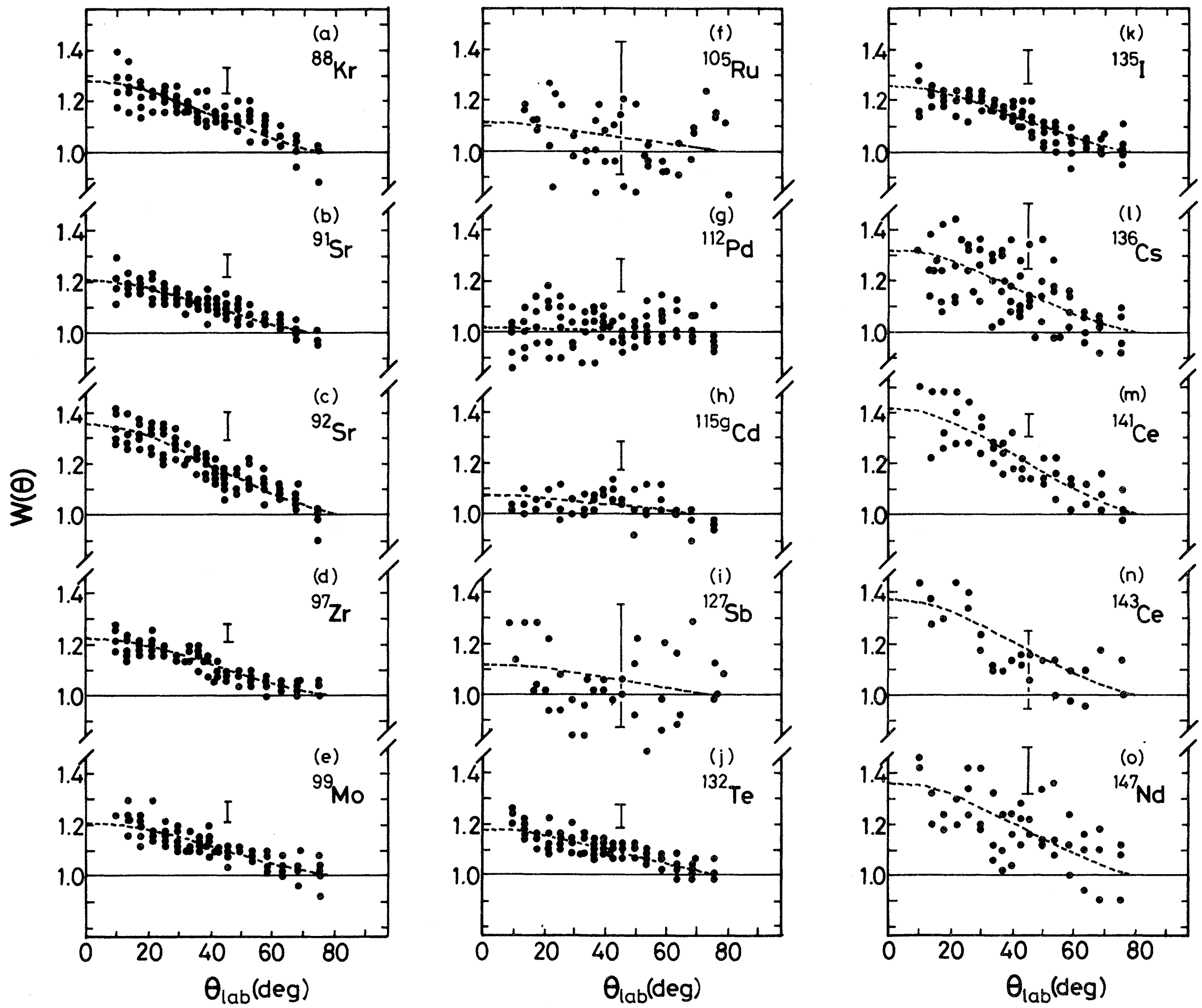

FIG. 2. Angular distributions of fission fragments produced in 15 MeV proton-induced fission of ^{232}Th. Dashed lines are the curves fitted by the least-squares method assuming $W(\theta)=a+b\cos^2\theta$. Vertical lines are the typical examples of errors.

kinetic energies than the most-probable nuclide of the $A=136$ isobaric chain by $4-8$ MeV.[16] It is generally thought that only the primary fission fragments with a large amount of excitation energy can reach to the shielded nuclides after emission of a large number of neutrons.[16] This large excitation energy is considered to be stored in the form of deformation energy at the time of scission, the larger deformation resulting in smaller Coulombic repulsion energy. The fact that the anisotropy of ^{136}Cs is similar to those observed for the neighboring nuclides may, therefore, be interpreted as indicating that the fragment anisotropy is rather insensitive to the degree of deformation and to the static deformation energy of the two touching nuclides at scission. A similar discussion is applicable to ^{147}Nd, although in this case ^{147}Nd is not a shielded nuclide. It is more asymmetrically divided products, and the reaction Q value to form ^{147}Nd is smaller than those of typical asymmetrically divided products, such as ^{92}Sr, ^{141}Ce, etc. The fission events leading to the production of ^{147}Nd are generally deemed to have experienced more deformed configurations at time of scission than those leading to the formation of asymmetric peak products in the mass yield curve since they give smaller kinetic energies[17] and a larger number of neutron emission.[18] The fact that the anisotropy of ^{147}Nd is similar to those of typical asymmetrically divided products also suggests that the anisotropy is insensitive to

TABLE II. Angular anisotropies of fission fragments obtained in 15 MeV proton-induced fission of ^{232}Th.

Nuclide	Anisotropy
^{88}Kr	1.28±0.02
^{91}Sr	1.20±0.01
^{92}Sr	1.36±0.02
^{97}Zr	1.22±0.01
^{99}Mo	1.21±0.02
^{105}Ru	1.10±0.05
^{112}Pd	1.03±0.02
^{115}Cd^{g}	1.08±0.02
^{127}Sb	1.12±0.05
^{132}Te	1.18±0.01
^{135}I	1.24±0.02
^{136}Cs	1.32±0.04
^{141}Ce	1.42±0.04
^{143}Ce	1.36±0.06
^{147}Nd	1.37±0.06

the static potential energy at scission. In this work the observed mass dependence of angular anisotropies was interpreted by assuming the existence of two kinds of saddle points, one leading to the mode of asymmetric mass division and the other to the mode of symmetric mass division.

In the 15 MeV proton-induced fission of ^{232}Th, however, multiple chance fissions (p,nf) and $(p,2nf)$ are energetically possible. If the contributions of higher-order-chance fissions are not negligible, the effect of the contributions has to be taken into consideration. In the higher chance fissions the excitation energies are smaller than that of the first chance fission (p,f) because of neutron emission prior to fission.

If the excited levels in the transition nucleus are described by statistical theory at moderate excitation energies, the angular distribution of the fission fragment is expressed by[2]

$$W(\theta)\propto\sum_{I=0}^{\infty}\frac{(2I+1)^2T_I\exp[-(I+\frac{1}{2})^2(\sin^2\theta)/4K_0^2]J_0[i(I+\frac{1}{2})^2(\sin^2\theta)/4K_0^2]}{\operatorname{erf}[(I+\frac{1}{2})/(2K_0^2)^{1/2}]}\,, \quad (1)$$

where the target spin is assumed to be zero and T_I denotes the transmission coefficient for the partial wave I. The J_0 is the zero order Bessel function, erf(x) is the error function, and K_0^2 is given by $I_{\rm eff}(t/\hbar^2)$. The quantity $I_{\rm eff}$ is equal to $I_\perp I_{||}/(I_\perp-I_{||})$, where $I_\perp$ and $I_{||}$ are nuclear moments of inertia around an axis perpendicular and parallel to the nuclear symmetry axis, respectively, and t is the temperature of the nucleus at the saddle point.

If the excitation energy is small, the nuclear pairing effect has to be taken into considera-

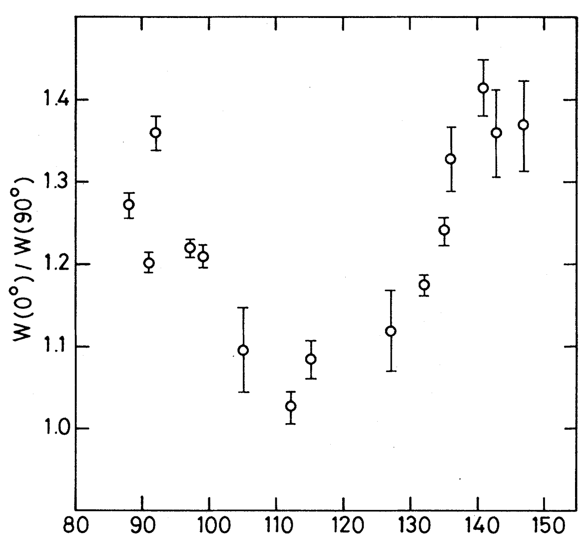

FIG. 3. Angular anisotropy of fission fragment as a function of fragment mass.

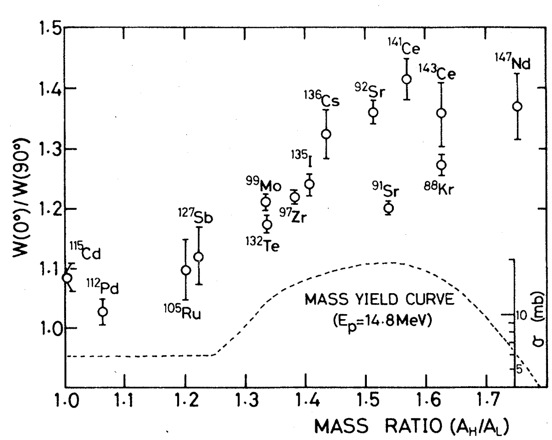

FIG. 4. Dependence of angular anisotropy on fragment-mass ratio in 15 MeV proton-induced fission of ^{232}Th. Dashed curve is the mass yield curve for 14.8 MeV proton-induced fission of ^{232}Th (Ref. 9).

tion.[19–22] Griffin[23] used the BCS prescription for the analysis of the anisotropy of fission fragments from the neutron-induced fission on ^{239}Pu. According to him, at low excitation energy, a superfluid nucleus will have a value of K_0^2 significantly less than its noninteracting counterpart, and at an excitation energy exceeding the critical energy there is no reduction of K_0^2:

$$K_0^2=\frac{I_\perp}{I_\perp-A(t/t_c)I_{\parallel}^{\text{rigid}}}\frac{t_c I_{\parallel}^{\text{rigid}}}{\hbar^2}\frac{t}{t_c}A(t/t_c)\ , \qquad (2)$$

where $I_{\parallel}^{\text{rigid}}$ represents the rigid body moment of inertia. It is known that the moment of inertia of a nucleus can be approximated by that of a rigid body as the deformation becomes large.[24] The notation t_c is the critical temperature, and $A(t/t_c)$ is a temperature-dependent integral. The critical temperature can be obtained from a gap energy,[23] and the numerical value of $A(t/t_c)$ is given by Vandenbosch and Huizenga.[2]

For the interpretation of the observed angular anisotropies, two kinds of saddle points were assumed. This assumption is supported not only by the argument described above, but also by the excellent fit of the observed trough-to-peak ratio as reported in a separate paper.[9] The contributions of multiple chance fissions and the excitation energies E_{ex} at the corresponding saddle points in the 15 MeV proton-induced fission of ^{232}Th were also evaluated in Ref. 9, some results of which are tabulated in Table III together with nuclear temperature t evaluated by $E_{\text{ex}}=at^2$. The moments of inertia are given by Cohen *et al.*[25] They calculated rigid-body moments of inertia for the symmetric saddle point shape predicted by the liquid-drop model as a function of the fissionability parameter χ. The moments of inertia are presented in Table IV for ^{233}Pa, ^{232}Pa, and ^{231}Pa. Although the moments of inertia for the asymmetric saddle point shape may be slightly different from those for the symmetric saddle point shape, here they were assumed equal. The neutron energy gaps of ^{233}Pa and ^{231}Pa were estimated from the neighboring even-even nuclides,[26] and evaluated as 1.214 MeV for ^{233}Pa and 1.520 MeV for ^{231}Pa. It turned out that the effect of the nuclear pairing had to be taken into account only for the asymmetric third chance fission, where the ratio of t to t_c was 0.627. For each multiple chance fission, the K_0^2 values were evaluated and are presented in Table V. The corresponding anisotropy can be evaluated by means of Eq. (1), and it is weighted by the contribution of each chance fission in order to give an apparent angular anisotropy. The results of the calculation are that the angular anisotropy of fission products that have experienced the asymmetric saddle is 1.30, whereas the anisotropy of those that experienced the symmetric saddle is 1.06. These values are close to the experimental results, 1.38 ± 0.05 and 1.06 ± 0.03 for asymmetric and symmetric fission products, respectively. The anisotropies in the intermediate mass region may be interpreted as the results of mixing of the symmetric and asymmetric fission modes in various degrees according to the degree of contribution from each mode.

Cohen *et al.*[6] reported in 1955 that the angular distribution of the fission fragment in the 22 MeV proton-induced fission of ^{232}Th is dependent on fragment mass, although the experimental errors were fairly large. Halpern and Strutinski[27] gave an explanation of their results. They assumed that the symmetric mass division required an extra energy in comparison with the asymmetric mass division, and that the asymmetric fission fragments were mostly produced in the second chance fission while the symmetric fragments were produced in

TABLE III. Excitation energy at saddle point and contribution of multiple chance fission at $E_p=15$ MeV from Ref. 9.

Mode of fission		E_{ex} (MeV)	t (MeV)	Contribution (%)
Asymmetric fission	(p,f)	14.3	0.69	39
	(p,nf)	7.5	0.50	5
	$(p,2nf)$	2.1	0.27	56
Symmetric fission	(p,f)	11.4	0.58	98
	(p,nf)	5.0	0.39	2

TABLE IV. Moments of inertia of ^{233}Pa, ^{232}Pa, and ^{231}Pa at saddle point configuration given by Cohen *et al.* (Ref. 25).

Nuclide	χ	$I_{\parallel}$	$I_{\perp}$	I_{eff}
^{233}Pa	0.764	0.568	1.90	0.808
^{232}Pa	0.765	0.570	1.89	0.816
^{231}Pa	0.766	0.571	1.88	0.820

the first chance fission. According to them, the excitation energy at the saddle point becomes higher for the symmetric mass division than that for the asymmetric. Hence, the value of K_0^2 for the symmetric mass division is larger, and consequently, the anisotropy of symmetrically divided fission products is smaller. They did not, however, explain the nature of the "extra energy" required for the symmetric mass division. Wilkins *et al.*[28] attempted to interpret the radium threshold anomaly reported by Konecny *et al.*[29] by the potential energy associated with the scission configuration. According to Wilkins *et al.*, if the minimum potential energy associated with scission configuration lies above the saddle-point energy for a particular mass split, then a fission threshold at some excitation energy above the saddle-point barrier will appear for that mass split. This kind of interpretation may be applicable only to the fission system when the saddle-point configuration is close to the scission configuration. If the extra energy assumed by Halpern and Strutinski is sought to be explained by the difference between the potential energy of the scission configuration and the saddle-point barrier energy, the former has to be roughly constant for all the mass divisions of $A_H/A_L \simeq 1.0-1.2$ and, moreover, it has to also be nearly constant for the mass divisions of $A_H/A_L > 1.5$, and the difference smaller for $A_H/A_L > 1.5$ than for $A_H/A_L \simeq 1.0-1.2$ in order to explain the present anisotropy data shown in Fig. 4. From the present knowledge of the fragment shell structure at scission,[28,30] it is hard to understand such a constancy for a wide range of mass splittings. Another source of the extra energy may be the extra threshold energy required for the dynamical motion to proceed through a path from a saddle to a symmetric scission configuration. However, no theoretical work has been reported on this nature of the extra threshold energy yet. In this paper, the extra energy was taken to be the energy difference between the symmetric and asymmetric barrier heights as originally proposed by Möller and Nilsson,[7] and experimental data observed in this work and those reported in a separate paper[9] could be consistently explained by the assumption of two types of saddle configurations. However, this fact does not necessarily exclude the possibility of other models equally explaining the data. Detailed calculations based on other models have to be carried out to verify their validity.

It has been established recently that there exists a double-humped fission barrier. If the first barrier height (at smaller deformation) is higher than the second barrier height (at larger deformation), the fission probability is mainly controlled by the first saddle, whereas the angular distribution may be determined at the second saddle. In this case the excitation energy (and hence, the nuclear temperature) deduced from the fission probability may not be related to the angular distribution of the fission fragment. However, for ^{233}Pa, ^{232}Pa, and ^{231}Pa the second barrier heights are predicted to be higher than the first barrier heights,[31] and, therefore, in the case of proton-induced fission of ^{232}Th the second saddle at which the angular distribution

TABLE V. Theoretically predicted K_0^2 values and angular anisotropies in 15 MeV proton-induced fission of ^{232}Th.

Mode of fission		K_0^2	$W(0°)/W(90°)$	Apparent anisotropy
Asymmetric fission	(p,f)	71	1.05	
	(p,nf)	51	1.07	1.30
	$(p,2nf)$	7	1.50	
Symmetric fission	(p,f)	59	1.06	1.06
	(p,nf)	40	1.09	

is determined is also expected to control fission probability. Thus, the analysis of the data described above may be justified.

Consequently, the agreement between the theoretical prediction and the observed anisotropies strongly suggests that there are two kinds of saddle points with different barrier heights, and that the final mode of mass division is essentially determined by which saddle point a nucleus has experienced in the course of the collective motion to fission. The present result is also qualitatively in agreement with the theoretical prediction by Maruhn and Greiner,[32] which states that the effect on mass division of the dynamical descent from the saddle to scission is rather small and that the final mass division is already determined at the stage shortly after the completion of barrier penetration. Two kinds of saddle points deduced in this work may be related to the reflection asymmetric and symmetric saddles predicted by Möller and Nilsson.[7]

IV. CONCLUSION

The angular distributions of 15 fission fragments were radiochemically measured in the 15 MeV proton-induced fission of ^{232}Th. It was found that the angular anisotropies of symmetrically divided fragments (1.06 ± 0.03) were smaller than those of asymmetrically divided fission fragments (1.38 ± 0.05). Using the results of the calculation of the evaporation-fission competition and Bohr's channel theory including the correction for nuclear pairing (BCS model), the angular anisotropy could be deduced theoretically assuming two kinds of saddle point configurations with different barrier heights. Theoretical angular anisotropies were in good agreement with the experimental ones for the fragments produced by typical asymmetric and symmetric mass divisions. Therefore, it is suggested that the gross feature of the final mass division in fission process is determined by which saddle the nuclear collective motion would proceed through.

The authors would like to express their grateful acknowledgment to Prof. K. Yamakoshi of the Institute for Cosmic Ray of the University of Tokyo for use of the low-background Ge(Li) detector for the measurement of ^{136}Cs.

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