

## Angular Distribution of Fission Fragments from 22-Mev Proton-Induced Fission of $U^{238}$ , $U^{235}$ , $U^{233}$ , $Th^{232}$ , and $Th^{230}$

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Angular distributions of several fission fragments from 22-Mev proton-induced fission of  $U^{238}$ ,  $U^{235}$ ,  $U^{233}$ ,  $Th^{232}$ , and  $Th^{230}$  were measured. The data are fitted to  $N(\theta) = a + b \cos^2\theta$  where  $b$  ranges from  $0.08a$  to  $0.28a$ . The coefficients for possible "cos $\theta$ " terms (i.e., terms asymmetric about 90 deg) are no larger than  $0.01a$ . In every case, asymmetric fission gives more anisotropic angular distributions than symmetric fission. The anisotropy in symmetric fission is about the same for all target elements; in asymmetric fission, the thorium isotopes have larger anisotropies than the uranium isotopes. Among the latter, the anisotropies are quite similar for  $U^{238}$  and  $U^{235}$ , but somewhat smaller for  $U^{233}$ . Explanations for various aspects of the phenomena are proposed.

IN a previous paper (A),<sup>1</sup> the importance of studies of angular distributions of various fission fragments from proton-induced fission was discussed, and results were presented for fragments of five different masses resulting from 22-Mev proton-induced fission of  $Th^{232}$ . In this paper, results are presented for similar work on  $U^{238}$ ,  $U^{235}$ ,  $U^{233}$ , and  $Th^{230}$ , and the data for all of these and for  $Th^{232}$  are extended to include a greater range of fragment masses. The four-angle method, described in detail in A, was used throughout. To recapitulate briefly, the target assembly consists essentially of a one-mil platinum foil target,\* plated on one side with a narrow strip of fissionable material about  $0.4 \text{ mg/cm}^2$  thick, and placed at the center of a 3.5-inch radius semicircle, along the circumference of which is placed a one-mil aluminum foil. The internal, circulating, 22-Mev proton beam from the ORNL 86-inch cyclotron passes through the target and induces fission reactions; the fission fragments are stopped in the aluminum foil. After the bombardment, the foil is removed and cut into several pieces, and each piece is radio-chemically processed for various fission products. These are then counted under end-window Geiger counters to determine their activities, and weighed to determine chemical yields. The specific activities in the various foils are proportional to the intensity of fission fragments emitted at the angles at which the foils were located during the bombardment. Separate runs are necessary for the forward (0 to 90 deg) and backward (90 to 180 deg) directions. It was concluded in A that the data could be satisfactorily fitted by an angular distribution of the form

$$N(\theta) = a + b \cos^2\theta, \quad (1)$$

and that the principal result to be obtained is the ratio  $b/a$  which is a measure of the anisotropy. It was decided that this could be determined most efficiently by measurements at only four angles (12, 20, 75, and

83 deg for the forward runs, and 163, 155, 102, and 93 deg for the backward runs). The data at these angles were fitted by least-squares methods to Eq. (1) to determine  $b/a$ . Since separate runs were made in the forward and backward directions, independent determinations of  $b/a$  from the  $0^\circ/90^\circ$  and  $180^\circ/90^\circ$  ratios were obtained so that the symmetry about 90 deg was checked.

The principal change with respect to A was the inclusion of data for bromine fission products which gives a very important point on the anisotropy *vs* mass ratio curve. The data analysis was slightly different, in that run results were thrown out when the determinations at one of the two pairs of adjacent angles differed by more than three S.D.'s,<sup>2</sup> or when determinations at each of the two pairs differed by more than two S.D.'s.

One important question to be investigated is the symmetry of the angular distributions about 90 deg. This is inextricably tied up with the determination of asymmetries introduced by the experimental methods, so that the two (hereafter termed "true" and "experimental") must be determined simultaneously. The difference between  $b/a$  measured in the forward and backward directions is shown in Table I for various target materials and fragment masses. For symmetric fission (represented by Ag) there can be no true asymmetry, and for asymmetric fission, the true asymmetries for light fragments (represented by Zr,

TABLE I.  $b/a$  from  $0^\circ/90^\circ$  ratio minus  $b/a$  from  $180^\circ/90^\circ$  (multiplied by 100).

Target	Fission fragment				
	Ag <sup>112,113</sup>	Zr <sup>97</sup>	Sr <sup>91,92</sup>	Br <sup>83</sup>	Ba <sup>139</sup>
$Th^{232}$	$1 \pm 3$	$-5 \pm 4$	$2 \pm 3$	$10 \pm 5$	$2 \pm 3$
$U^{238}$	$-2 \pm 4$	$0 \pm 2$	$7 \pm 4$	$-7 \pm 4$	$-1 \pm 3$
$U^{235}$	$-4 \pm 2$	$1 \pm 3$	$-1 \pm 3$	$-5 \pm 5$	$\dots$
$U^{233}$	$0 \pm 3$	$6 \pm 5$	$7 \pm 6$	$-4 \pm 6$	$1 \pm 11$
Average	$-1.3 \pm 1.5$	$0.5 \pm 2$	$3.8 \pm 2$	$-1.5 \pm 2.5$	$0.5 \pm 2$

<sup>1</sup> Cohen, Jones, McCormick, and Ferrell, Phys. Rev. **94**, 625 (1954), hereafter referred to as "A".

\* Note added in proof.—The target was so oriented that the fragments studied were emitted at angles less than  $45^\circ$  from the normal to the surface.

<sup>2</sup> "S.D." denotes "standard deviations". In this paper, all errors quoted are one standard deviation, determined from the reproducibility of the results.

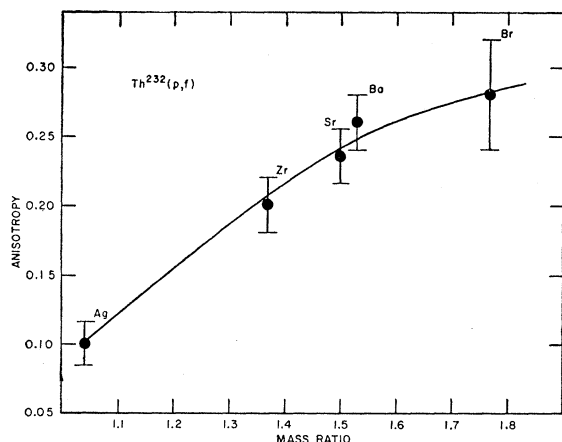


FIG. 1. Anisotropy in angular distributions of fragments from 22-Mev proton-induced fission of  $\text{Th}^{232}$  vs mass ratio (mass of heavy fragment divided by mass of light fragment). Values plotted are average of  $0^\circ/90^\circ$  and  $180^\circ/90^\circ$  ratios. Errors shown are one standard deviation as determined by reproducibility.

Sr, and Br) must be equal and opposite to those for heavy fragments (represented by Ba). From the symmetric fission, the experimental asymmetry is  $-0.013 \pm 0.015$ . From the asymmetric fission, the experimental asymmetry is  $0.007 \pm 0.015$ , and the true asymmetry is  $0.004 \pm 0.025$ .

Another method of determining the true asymmetry, as discussed in A, is by comparing anisotropies of two fragments emitted in the same reaction. Three cases of this type are available. In  $\text{U}^{238}$  fission, Ba and Zr correspond to the same mass ratio; the asymmetry favors the light fragment going forward with a difference in anisotropies of  $0.003 \pm 0.025$ . In  $\text{U}^{235}$  fission, Ba and Sr correspond to the same mass ratio; the light fragment going forward has a larger anisotropy by  $0.03 \pm 0.05$ . For  $\text{Th}^{232}$ , Ba, and Sr have approximately the same mass ratio; the heavy fragment going forward is favored with an anisotropy difference of  $0.025 \pm 0.03$ .

Both methods therefore indicate no significant true asymmetry about  $90^\circ$  deg within a standard deviation of

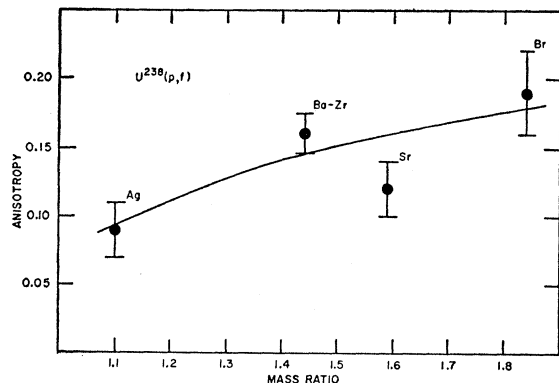


FIG. 2. Anisotropy in angular distributions of fragments from 22-Mev proton-induced fission of  $\text{U}^{238}$  vs mass ratio. See caption for Fig. 1.

about 0.02. This corresponds to a coefficient of a possible  $\cos\theta$  term in (1) no larger than about  $0.01a$ .

The experimental asymmetry also appears to be quite small; averaging the two above determinations gives  $-0.002 \pm 0.01$ . Evidently the experimental asymmetry<sup>7</sup> discussed in A was overestimated, although the preliminary values of  $0.03 \pm 0.03$  and  $0.02 \pm 0.03$  found by two different methods in that paper are not in disagreement with this result.

Since it has been concluded that the angular distributions are truly symmetric about  $90^\circ$  deg, values of anisotropies obtained by forward direction and backward direction runs were averaged, and the S.D. of the average is estimated from the agreement between the two and their separate S.D.'s (note that this is a somewhat more conservative procedure than used in A). The separate S.D.'s were determined strictly from the reproducibility of the results.

The results for  $\text{Th}^{232}$ ,  $\text{U}^{238}$ ,  $\text{U}^{235}$ , and  $\text{U}^{233}$  are shown in Figs. 1 to 4 and summarized in Fig. 5. For  $\text{Th}^{230}$ ,

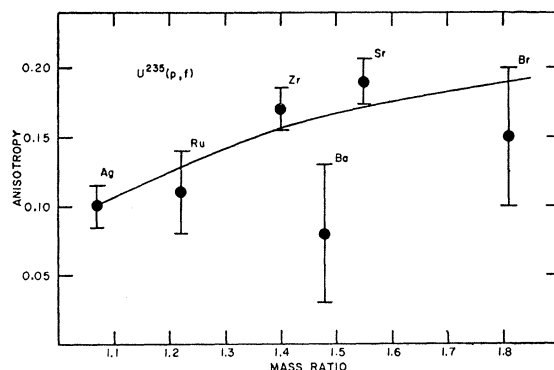


FIG. 3. Anisotropy in angular distributions of fragments from 22-Mev proton-induced fission of  $\text{U}^{235}$  vs mass ratio. See caption for Fig. 1.

considerably less data were available so that a plot is not shown; the anisotropies for that element are given in Table II.

In general, the variations in standard deviations are due to the number of runs carried out, although the larger errors for the bromine points are generally due to poorer reproducibility. A ruthenium point for  $\text{Th}^{232}$  was given in A, but is not included here because of the very poor reproducibility and resultant large S.D.

Qualitatively, the data are similar for all target elements in that the anisotropy for asymmetric fission is larger than for symmetric fission. The anisotropy for symmetric fission is about the same for all elements studied; for asymmetric fission it seems to be appreciably larger for the thorium than for the uranium isotopes. Among the uranium isotopes,  $\text{U}^{233}$  seems to show the least variation of anisotropy with mass ratio; within the accuracy of the experiment it may have no such dependence. The curves through the  $\text{U}^{235}$  and  $\text{U}^{238}$  data could easily be identical. An important difference

between these curves and the curve drawn through the meager data of A is that the bromine points show very definitely that the curves are concave toward the mass-ratio axis.

As pointed out in detail in A, the anisotropic angular distributions can be qualitatively explained by the Wheeler-Hill collective model<sup>3</sup>; but they can also be adequately explained by straightforward application of conservation of angular momentum in the statistical theory of nuclear reactions.<sup>4,5</sup> No quantitative calculations have been made on either model, but since the latter explanation is more conventional and has been experimentally shown to explain similar angular distributions in cases where the Wheeler-Hill model could have no effect (angular distributions of neutrons from  $(\alpha, n)$  reactions<sup>5</sup>), it will be adopted in the ensuing discussion. Actually, there is some indication that it is the correct model; in accordance with it, the larger anisotropies in asymmetric fission could be explained<sup>6</sup> by the fact that there is more energy available in these

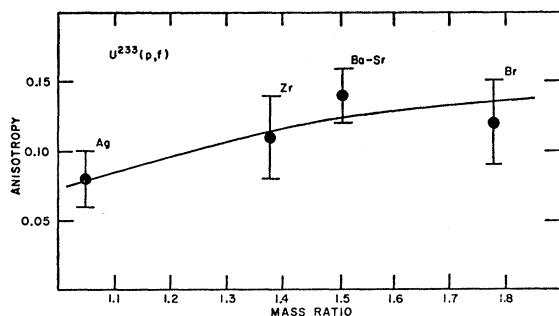


FIG. 4. Anisotropy in angular distributions of fragments from 22-Mev proton-induced fission of  $U^{233}$  vs mass ratio. See caption for Fig. 1.

than in symmetric fission.<sup>7</sup> On the other hand, the Wheeler-Hill model assumes that the anisotropies are caused by Coulomb effects, and these would be stronger in symmetric fission. The increase in anisotropy with asymmetry would therefore be difficult to explain.

The larger anisotropies for the thorium isotopes may be correlated with the fact that the fission cross section is smaller in these.<sup>8</sup> For the uranium isotopes, fission occurs in practically every nuclear reaction; this probably includes many cases where a  $(p, n)$  reaction is

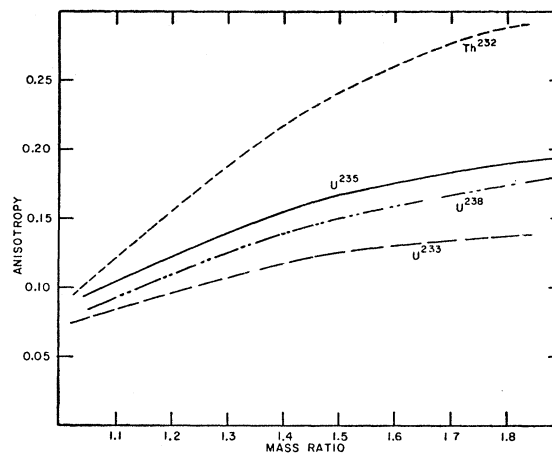


FIG. 5. Anisotropy in angular distributions of fragments from 22-Mev proton-induced fission of various target isotopes. Curves are taken from Figs. 1 to 4.

TABLE II. Anisotropies for  $Th^{230}$  fission.

Fission product	Mass ratio	Anisotropy
$Sr^{91,92}$	1.48	$0.39 \pm 0.08$
$Zr^{97}$	1.35	$0.19 \pm 0.04$
$Ag^{112,113}$	1.02	$0.22 \pm 0.08$

followed by fission. In such cases, the energy available is quite low, so that the angular distributions would be more isotropic. Since low-energy fission is largely asymmetric, the symmetric fission angular distributions would not be affected. In thorium, on the other hand,  $(p, n)$  reactions are quite frequently followed by further neutron emission to give  $(p, 2n)$  or  $(p, 3n)$  reactions rather than fission,<sup>8,9</sup> so that initial fission (i.e., not fission following neutron emission) makes up a larger share of the reactions.

Additional evidence for this point of view may be obtained from fission mass distribution studies.<sup>10</sup> Symmetric fission has been found to be more probable in  $Th^{232}$  than in  $U^{233}$  and  $U^{235}$ . This larger probability for asymmetric fission in the latter could be explained by the lower excitation energies available when fission follows emission of a neutron.

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<sup>9</sup> H. A. Tewes and R. A. James, Phys. Rev. **88**, 860 (1952).

<sup>10</sup> Jones, Timnick, Paehler, and Handley, Phys. Rev. (to be published). Other work is also summarized there.

<sup>3</sup> D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).

<sup>4</sup> L. Wolfenstein, Phys. Rev. **82**, 690 (1951).

<sup>5</sup> B. L. Cohen, Phys. Rev. **81**, 632 (1951).

<sup>6</sup> B. L. Cohen, unpublished report NP 1621, 1950 (unpublished).

<sup>7</sup> P. Fong, Seminar at Oak Ridge National Laboratory, 1954 (unpublished).

<sup>8</sup> G. H. McCormick and B. L. Cohen, Phys. Rev. **96**, 722 (1954); W. H. Jones (private communication).