energetically possible unless the residual nucleus (after emission of the proton) has sufficient excitation to allow neutron emission. Actually most of the reactions leave the residual nucleus with considerably higher excitation than this minimum.

Therefore, these experiments show quite conclusively that neither the first nor the fourth proposals listed in the introduction of this paper are the correct explanations for the excessive emission of charged particles in nuclear reactions. On the other hand, they provide a new and clear-cut demonstration of that effect in an energy region where uncertainties in nuclear temperatures and in the method of correcting for thresholds cannot be important factors.

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Angular Distributions of Fission Fragments from 22-Mev Proton-Induced Thorium Fission

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The angular distributions of barium, strontium, zirconium, ruthenium, and silver fission products from thorium fission induced by 22-Mev protons were measured using the internal, circulating beam of the ORNL 86-inch cyclotron. Within the accuracy of the measurements, all angular distributions are symmetric about 90° and may be well fitted to $I(\theta)=a+b\cos^2\theta$. For Ba, Sr, Zr, Ru, and Ag fission products, for which the fission mass ratios are 1.53, 1.52, 1.37, 1.19, and 1.04, the anisotropy (b/a) is 0.26, 0.25, 0.19, 0.15, and 0.10, respectively.

INTRODUCTION

THE angular distributions of fragments from fission induced by thermal neutrons should be isotropic according to the Eisner-Sachs-Yang rule,[1] and the fact that they are, has been experimentally confirmed.[2] It has commonly been assumed that angular distributions are also isotropic for fission induced by high-energy particles, because fission proceeds by a compound-nucleus interaction in which the number of intermediate and final states is sufficiently large that the effects of individual levels, which are the usual cause of anisotropic angular distributions, would be expected to average out. This expectation, however, has not been verified by experiment.

The first reported measurements of this type was the work of Winhold, Demos, and Halpern[3] on thorium photofission; their data indicate angular distributions of the form $I=\alpha+\beta\sin^2\theta$, where β/α, which is a measure of the anisotropy, reaches values as large as 1.2. They interpret the phenomenon as due to the effects of dipole absorption. However, the recent theoretical work of Wheeler and Hill[4] has shown that compound nuclei undergo large oscillations which lower the Coulomb barrier in specified directions and thereby explain, at least qualitatively, the results of reference 3. At their suggestion, Dickinson and Brolley[2] measured the 0°/90° intensity ratios of fragments from 14-Mev neutron-induced fission of Th^{232}, Np^{237}, U^{233}, U^{235}, and U^{238}. In all cases, they found these ratios to be greater than unity (1.2−1.5), which is in qualitative agreement with the Wheeler-Hill prediction for particle-induced fission.

To throw further light on this problem, a program for the measurement of angular distributions of the products of *proton*-induced fission was undertaken by utilizing the internal, circulating beam of the ORNL 86-inch cyclotron. This approach has the advantage of providing very large incident particle currents, and is especially timely because methods of measuring angular distributions with this beam have recently been studied in considerable detail.[5] In this paper, we describe measurements of angular distributions of Ba, Sr, Zr, Ru, and Ag fission products from 22-Mev proton-induced thorium fission. The work is now being extended to other target elements, and eventually will be extended to other bombarding energies.

EXPERIMENTAL PROCEDURE AND RESULTS

The target assembly is similar to that pictured in reference 5, except that a more accurate method of

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[1] E. Eisner and R. G. Sachs, Phys. Rev. **72**, 680 (1947); C. N. Yang, Phys. Rev. **74**, 764 (1948).

[2] W. L. Dickinson and J. E. Brolley, Phys. Rev. **90**, 388 (1953).

[3] Winhold, Demos, and Halpern, Phys. Rev. **87**, 1139 (1952); I. Halpern and E. J. Winhold, U. S. Atomic Energy Commission Report AECU-2494 (unpublished).

[4] D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).

[5] B. L. Cohen and R. V. Neidigh, Rev. Sci. Instr. **25**, 255 (1954).

positioning the detector foil has been incorporated. The target itself is a 0.001-inch platinum foil with a narrow strip of $\frac{1}{2}$ mg/cm^2 thorium oxide plated on one side. The fission fragments are collected in a 7 mg/cm^2 aluminum detecting foil covered with a 1.3 mg/cm^2 aluminum wrapper to exclude thermally evaporated activities which are present in the cyclotron. Since the range of fission fragments[6] in aluminum is about 2.5 to 4.0 mg/cm^2, practically all fragments should be caught in the detecting foil. Independent tests indicated that about 6 percent were stopped in the wrapper, and about $\frac{1}{2}$ percent went completely through the foil. Both of these quantities vary with angle because of the energy variations due to center-of-mass motion, so they require a small correction which will be discussed below.

After bombardment, the aluminum foil is cut into equal sized pieces (these are weighed and corrections are made for the small differences in their sizes) and processed chemically to isolate the various fission products. These are then counted under end-window Geiger counters to determine their activities, and later weighed to determine chemical yields. The specific activities in the various foils are thus proportional to the intensity of fission fragments emitted at the angles at which the foils were located during the bombardment. The activity measurements are corrected for variation of solid angle in the center-of-mass system and for small ($\sim$1 percent) known variations in the target-to-collector distance. For purposes of studying the experimental problems, the angular distributions thus obtained were fitted by least squares methods to a formula of the type

$$I = a + b\cos^2\theta, \tag{1}$$

and the parameter b/a, which is essentially a measure of the anisotropy of the angular distribution, was determined.

In considering the effects of various types of uncertainties in the experiment on the resulting value of b/a, it is necessary to distinguish between random and systematic errors. Random errors, which are by far the greater in any single run, are evidenced by large and uncorrelated variations in the data which are not reproduced in successive runs. The rms fractional error of this type, δ, can be determined by comparing intensity measurements at adjacent angles and correcting for the true change in intensity between these angles as determined from the final results of the experiment. (In all cases, the true change is small.) Analysis shows that the absolute error in the determination of b/a is 1.1δ to 1.4δ.

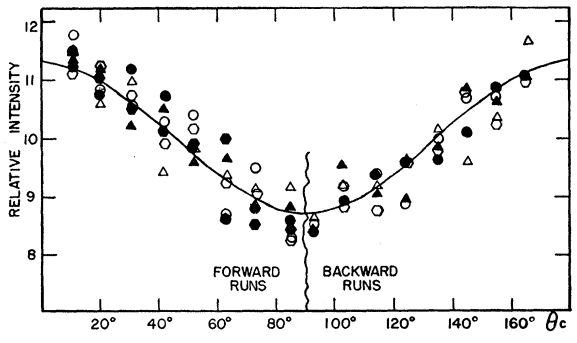

FIG. 1. Angular distribution of Ba^{139} fission fragments from the first set of runs described in text. There is no significance in the use of the same type pointers in the forward and backward direction. The internal consistency of this set is better than for any other.

[6] Katcoff, Miskel, and Stanley, Phys. Rev. **74**, 631 (1948).

The principle contributor to δ is error in the chemical processing and yield determination which introduces an rms error of 6–10 percent. (Actually it varied as the chemical processes were changed in an effort to reach the best compromise between chemical purity and time requirements. For ruthenium, the process was never perfected, the average value of δ being about 0.17 percent.) Other sources of random error in the relative intensity at various angles, and their estimated magnitudes are

Counting statistics	1–3 percent,
Counter geometry	1 percent,
Beta self-absorption	1–4 percent,
Target alignment	2 percent,
Detector foil positioning	2 percent.

While the total random error in a given determination of b/a is quite large, these average out in a large number of runs, so that systematic errors are at least equally important. The estimated errors in the alignment of the thin target and the estimation of the effects of finite beam size would change b/a by about ± 0.02. The maximum uncertainties in determination of the angles and in the corrections for center-of-mass solid angle would not change it by as much as ± 0.01. None of these would cause any error in the relative values of b/a for the various fission products, or for the forward and backward directions.

One source of systematic error of the latter type is the variation with angle of the fraction of the fragments stopped in the thin wrapper covering the collector foil due to energy differences arising from center-of-mass motion. This effect, combined with any other systematic errors between the forward and backward directions, was investigated by carrying out an extensive series of runs on the angular distributions of Ba^{139} and Sr^{91,92} fragments; these are emitted in the same reactions (assuming 2 or 3 neutrons per fission) and hence must be emitted in opposite directions. The values of b/a were

Barium forward	0.277 ± 0.025
Strontium backward	0.240 ± 0.030,
Barium backward	0.232 ± 0.027
Strontium forward	0.260 ± 0.026.

The systematic errors thus caused the forward values to be larger by 0.037 ± 0.040 and 0.028 ± 0.038 in the two cases; the average of these is 0.033 ± 0.027. This agrees satisfactorily with a direct determination of 2 ± 3 percent arrived at by processing the aluminum wrappers to determine the relative amounts stopped in them at various angles.

The corrected value of b/a for Ba forward (or Sr backward) can be taken as the average of the first two values, or 0.258 ± 0.019; similarly, the corrected value for Ba backward (or Sr forward) is 0.246 ± 0.019. It thus appears that the angular distributions are symmetric about 90° within the accuracy of this experiment. In the determinations of angular distributions of all other fission fragments, the difference between b/a in the forward and backward directions differed by amounts within experimental error of the difference for barium and strontium; they were thus assumed to be symmetric about 90° and the value of b/a was taken as the average of the values in the forward and backward directions, with the standard deviation taken as $\sqrt{2}/2$ times the rms deviations of the two. The values obtained in this way are shown in Table I. This procedure probably underestimates the standard deviation of the result, but was adopted as the most practical approach. If there is actually a small asymmetry about 90° so that the angular distributions are of the form

$$I(\theta)=a+a_1\cos\theta+b\cos^2\theta, \qquad (2)$$

the values of b/a given here are still correct for that formula. From the barium-strontium data, $a_1/a=0.006\pm0.013$ or, more to the point, $a_1/a<0.02$.

In the first set of runs, which were made primarily to test the feasibility of the project, the angular distributions were determined for barium only. This set included six runs for the forward direction and five for the backward, with determinations at eight angles on each run. After chemical processing, each sample was counted at least four times and the data were then plotted to determine the 85-minute activity of barium-139 in each sample. The results are shown in Fig. 1. Concurrently, tests were made to ascertain (by range considerations) that the activities actually were fission fragments and were coming from the thorium. Also, the aluminum wrappers were processed to determine their necessity (they were very necessary in most runs) and to obtain an estimate of the center-of-mass effect described previously.

Since the results seemed satisfactory, the project was enlarged to include determinations of barium, strontium, and silver, still at eight angles in each direction. To conserve time, two counters were used for each element. A single sample (No. 1) was placed in the first of the two, and each of the other seven (No. 2 –No. 8) was counted in turn in the second counter to determine the ratios of their activities to that of sample No. 1. To determine the relative efficiencies of the two counters, sample No. 2 was counted in the first counter and sample No. 1 in the second. The counting process was repeated at least twice. This method automatically corrects for decay and thus eliminates the necessity of plotting decay curves. A few samples of each element were counted exhaustively to ascertain that the half-lives were correct; this serves as a check on the adequacy of the chemical processing.

TABLE I. The values of b/a obtained by fitting the data to Eq. (1). The forward and backward data are not corrected for variation of energy with angle due to center-of-mass motion; since this correction is equal and opposite for the two cases, the average does not require this correction. The mass ratios are calculated assuming three neutrons per fission. The errors listed are standard deviations due to statistics only.

Element	Mass ratio	Measured b/a Forward	Backward	Average
Ba^{139}	1.53	0.285 ± 0.02	0.248 ± 0.02	0.264 ± 0.015
$Sr^{91,92}$	1.52	0.260 ± 0.025	0.240 ± 0.03	0.250 ± 0.020
$Ag^{112,113}$	1.04	0.105 ± 0.025	0.095 ± 0.025	0.100 ± 0.015
Zr^{97}	1.37	$0.24\ \pm0.025$	$0.15\ \pm0.03$	0.195 ± 0.020
Ru^{105}	1.19	$0.19\ \pm0.08$	$0.11\ \pm0.08$	$0.15\ \pm0.06$

A total of twenty-eight runs, 18 forward and 11 backward, were made in this program. The values of b/a were determined for each run and plotted against time to determine whether secular variations were present. Since there was some evidence of such a variation for silver, further data on that element were taken in the next set of runs, described below. It was finally concluded, however, that the apparent secular variation was due to statistical fluctuations. No secular variations were noticeable in the barium and strontium data.

The values of b/a for barium and strontium on the same runs were plotted against each other. The resulting "scatter" diagram showed no significant correlation between the two, thus indicating that there were no major errors due to such things as target alignment, beam characteristics, etc.

The values of b/a for each element and for each direction (i.e., forward and backward) were plotted on probability paper as shown in Fig. 2. This tests how well the values can be fitted to a Gaussian error function (for a perfect fit, the points should lie on a straight line) and determines the most probable value of b/a (the abscissa for which the best line through the data crosses the ordinate "50") and the standard deviation of a single determination (the difference between the most probable value and the abscissa for which the line crosses the ordinate "16"). The fits to the Gaussian error function are generally satisfactory; the standard deviations of a single determination of b/a average about 0.09 for barium and strontium and about 0.08 for silver. The expected standard deviation, assuming that it is due only to random errors, was calculated by determining δ by the method discussed above; in this set of runs, the mean value of δ was about 0.06 to 0.07. Analysis shows that the standard deviation in b/a should be about 1.1δ for silver and about 1.2δ

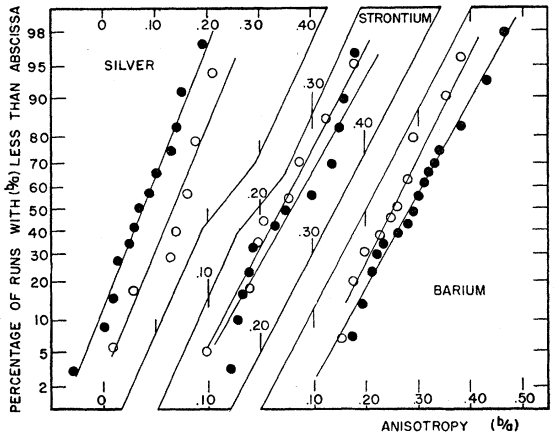

FIG. 2. Probability paper plot of anisotropies (b/a) in angular distributions of Ba, Ag, and Sr fission fragments from first and second sets of runs. The probability paper scale is such that when the percentage of runs with b/a less than each value is plotted *vs* that value, the points lie on a straight line if the data follow a Gaussian distribution. The abscissa at which the line crosses ordinate 50 is the most probable value; the difference between this and the abscissa at which the line crosses ordinate 16 is the standard deviation of a single determination, and the standard deviation of the mean is that divided by the square root of the number of runs. Solid circles represent forward direction runs and open circles represent backward direction runs. Data are not corrected for variation of fragment energy with angle due to center-of-mass effects. The correction reduces the anisotropy for the forward runs and increases it for the backward runs.

for barium and strontium. Thus the random errors account for the standard deviation in the results which supports the assumption that there are no appreciable secular variations.

The barium-strontium comparisons mentioned previously were carried out in this set of runs. The agreement between the values of b/a for barium and strontium lent confidence to the general method.

For purposes of plotting and further analysis, each run was normalized to the same total intensity and the data were grouped into series of 5 or 6 runs in chronological order. The average intensity at each angle for each group is shown plotted *vs* angle in Figs. 3, 4, and 5. One set of points in Fig. 3 (solid circles) represents the average of the data from Fig. 1. No correction was made for the center-of-mass variation of energy with angle in the plotting.

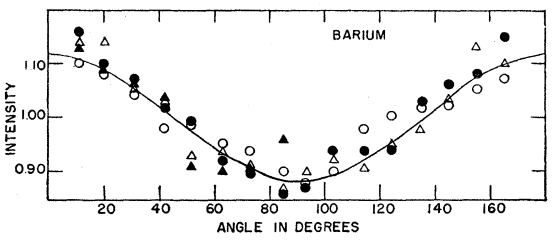

FIG. 3. Composite angular distribution of Ba^{139} fission fragments. Each set of points represents the average of a group of five or six runs. Data are not corrected for the variation in fragment energy with angle due to center-of-mass motion. The correction makes the data more nearly symmetric about 90°.

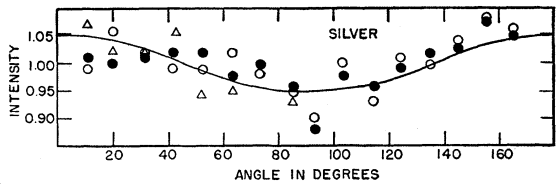

FIG. 4. Composite angular distribution of $Ag^{112,113}$ fission fragments. See caption for Fig. 3.

The average data were also fitted by least-squares methods to

$$I(\theta)=a'+b'\cos^2\theta+d'\cos^4\theta. \quad (3)$$

The values of d'/a' thus determined showed very poor reproducibility, so that it was concluded that the data is not sufficiently accurate for this type of analysis. In all cases, however, the sum of $b'/a'+d'/a'$ was nearly equal to b/a from Table I; the quantity $b/a-(b'/a'+d'/a')$ was 0.012 ± 0.007 for the forward runs and 0.000 ± 0.007 for the backward runs. Thus, if the anisotropy A is defined as

$$A=\frac{\frac{1}{2}[I(0°)+I(180°)]-I(90°)}{I(90°)}. \quad (4)$$

A is the same regardless of the method of analysis.

A third set of runs was undertaken primarily to extend the data to other fission products. The new elements chosen were zirconium and ruthenium, and due to the previously mentioned uncertainty in the silver data, further data on silver were taken concurrently. The procedure was also simplified by discontinuing intensity determinations between 25° and 65°, and between 115° and 155° since these are of little importance in determining the anisotropy; the number of foils was thus reduced to four for each run. A further simplification was achieved when it was realized that no significant information had been obtained from counting each sample several times, since, in the end, a simple average was always taken. For this set of runs, therefore, each sample was counted only once but for an extended time. A total of 19 runs, 11 backward and 8 forward, were included in this set, and least-squares fits to Eq. (1) were made to determine b/a.

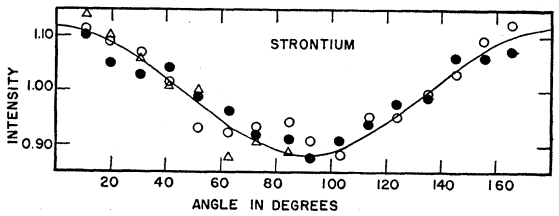

FIG. 5. Composite angular distribution of $Sr^{91,92}$ fission fragments. See caption for Fig. 3.

Before comparing these values directly with the previous data, it is necessary to determine whether the four angles omitted in this set contribute to b/a in a systematic way. An analysis of the previous data omitting these four angles was therefore carried out; it was found that b/a obtained from eight angles was greater than b/a obtained from the four by 0.026 ± 0.009 in the forward direction, and 0.000 ± 0.005 in the backward direction. The discrepancy in the forward direction is somewhat disturbing; however, the values of $(b'/a'+d'/a')$, which are probably the most accurate index of the anisotropy, lie midway between the values of b/a as calculated by the eight-angle and four-angle methods. Also, the agreement between all three methods is excellent in the backward direction. There thus seemed to be no alternative to considering the values of b/a as calculated by the eight and four angle methods as equivalent, with the realization that there may be a small systematic error introduced in the process.

The values of b/a obtained from this last set of runs are shown plotted on probability paper in Fig. 6, and their averages are listed in Table I. The entry for silver in Table I is an average over the two sets of silver runs.

CONCLUSIONS AND DISCUSSION

Two principle new quatitative conclusions can be drawn from this data. Firstly, the angular distributions are symmetric about 90° well within the experimental error, and secondly, the anisotropy is much greater for asymmetric fission than for symmetric fission. The latter point is demonstrated in Fig. 7 where the anisotropy is plotted against the mass ratio of the fission products. In considering the absolute values of the anisotropy, one should allow for possible systematic errors of about ±0.03 in the data from Table I or Fig. 7. In using the *relative* values for the various mass ratios, the systematic error is probably not much larger than ±0.01.

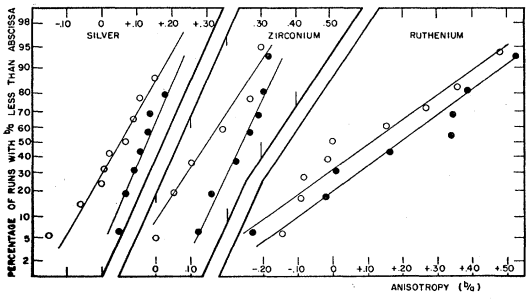

FIG. 6. Probability paper plot of anisotropies in angular distributions of Ag, Zr, and Ru fission products from third set of runs. See caption for Fig. 2. The increased slope of the forward with respect to the backward data for silver and zirconium represent an improvement in accuracy (smaller standard deviation) due to changes in the chemical processing methods. The shallow slope for ruthenium is due to difficulties in the chemical processing that were never overcome.

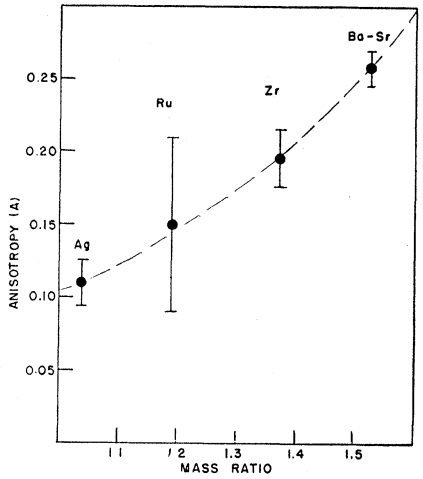

FIG. 7. Anisotropy in fission fragment angular distributions *vs* mass ratio (mass of heavy fragment divided by mass of light fragments). The data are taken from Table I.

The Wheeler-Hill theory, so far as is known, has not been applied to predict numerical values of the anisotropy. Also nothing is said in reference 4 about a possible dependence on mass ratio.

It is important to note that it has been demonstrated both theoretically[7] and experimentally[8] that, due to straightforward angular momentum considerations, angular distributions from compound nucleus reactions exhibit a monotonic decrease in intensity from 0° to 90° with symmetry about 90°. This is due to the fact that the angular momentum imparted to the compound nucleus by the orbital angular momentum of the incident particle is polarized in the sense that its component in the direction of incidence is zero; thus the spin of the compound nucleus is polarized in the same sense, and when it breaks up, this polarization must be shared in some fashion between the orbital angular momentum of the outgoing particles and their spins. The polarization of the outgoing orbital angular momentum causes the asymmetry in the angular distribution. The effect is sensitive to the distribution of spins in the final nuclei, so that some estimate of this would be necessary to determine the magnitude of the asymmetry. It is interesting to note, however, that this asymmetry is of about the same magnitude as the asymmetry in the angular distributions of neutrons from (α,n) reactions induced by 30-Mev alpha particles (from reference 8, $A=0.21$). The (α,n) case cannot be a Coulomb effect (such as the Wheeler-Hill effect) since neutrons are unaffected by Coulomb barriers, and the observed asymmetry is independent of atomic number.

The authors would like to acknowledge the assistance and encouragement of R. S. Livingston and J. L. Fowler in carrying out these experiments.

[7] L. Wolfenstein, Phys. Rev. **82**, 690 (1951).
[8] B. L. Cohen, Phys. Rev. **81**, 632 (1951).