Anisotropy of Fragments in the Neutron-Induced Fission of Pu^{240} , Pu^{242} , and $Pu^{241\dagger}$

J. E. SIMMONS, R. B. PERKINS, AND R. L. HENKEL

University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico

(Received 6 July 1964)

Experimental data are presented for the angular distribution of fragments in the neutron-induced fission of Pu²⁴⁰ and Pu²⁴², for a range of neutron energies from 0.65 to 8.25 MeV. The results are compared with other available data for even-even target isotopes by means of the parameter K_0^2 . It is concluded that excitation energy alone is not sufficient to describe the K_0^2 values for this class of fissioning nuclei. A correlation between K_0^2 and fissionability is obtained. Comparison is also made with data for the even-odd target nuclei U^{235} , U^{233} , and Pu²³⁹. It is concluded that the correlation with fissionability shown by the even-even targets could well explain the major part of the "anomalous spin effect" shown by the even-odd nuclei. Data for the anisotropy of fragments in the fission of Pu²⁴¹ are also recorded.

 \bigwedge^{\bullet} HE angular distribution of fragments $W(\theta)$ in fission induced by MeU neutrons is generally considered to depend primarily on the following two factors: L_m , the maximum value of orbital angular momentum, and the quantity K_0 , which is the standard deviation in the distribution of the projection of the total angular momentum vector on the nuclear symmetry axis at the saddle point. The quantity K_0 itself depends on the excitation energy E^* of the fissioning nucleus. The role of the target spin I_0 has generated considerable discussion, and is discussed below. The anisotropy¹ is conveniently given by the following approximation of Griffin, $\frac{2}{3}$

$$
W(\theta)/W(90) \cong 1 + L_m(L_m+2)/(8K_0^2). \tag{1}
$$

In the early theoretical development³ of the subject, the possible dependence of the anisotropy on the fissionability parameter Z^2/A was ignored, so that nuclei of a given class (even-even targets, for example) excited to the same excitation energy and bombarded by neutrons of the same value of L_m should show the same anisotropy. However, the work of Chaudhry, Vandenbosch, and Huizenga⁴ on the alpha fission of bismuth, lead, thallium, and gold has provided definite evidence of variation of K_0^2 with Z^2/A , in agreement with liquid-drop calculations of Cohen and Swiatecki.⁵ Recently, Griffin² has noted the possible existence of similar effects in the neutron-induced fission of U²³³ and Pu²³⁹.

This paper presents new experimental data for the angular distribution of fragments in the fission of Pu²⁴⁰ and Pu^{242} induced by neutrons in the energy range 0.65 to 8.25 MeV. Data taken at the same time for Pu²⁴¹ are recorded, but not discussed. We compare these new data with existing information available for other even-even target nuclei with the aim of looking for a possible dependence on Z^2/A . We find that the values of K_0^2 obtained for Pu²⁴⁰ and Pu²⁴² at fixed excitation energy, are significantly higher than those for uranium and thorium isotopes. The implications of these results are discussed in relation to the anisotropy data for Pu²³⁹, U²³³, and U²³⁵ in neutron-induced fission and the target spin question.

EXPERIMENT

The experimental arrangement is shown in Fig. 1. The counter is the same as that used previously,⁶ except for a modification described below. Monoenergetic neutrons were produced by means of the $T(p,n)He^3$ and $D(d, n)$ He³ reactions. The fission foil was contained in a multi-angle gas-filled counter, placed at a short distance from the neutron source. The angle-defining detectors were proportional counters covering angles from zero to ninety degrees. In the earlier work, the fissionable materials were deposited on thin foils, and backgrounds were minimized by coincidence counting between the fragment entering a given angle-defining counter and its recoil partner passing through a thin ionization region behind the foil. In the present experiment, the ionization region behind the foil was replaced by an ionization counter region of larger extent located between the foil and the proportional counters. Coincidences were then required between the two (dE/dX) signals from a single fragment in its passage through the ionization and proportional counters.

The foils of fissionable material⁷ were prepared by an electro-spray' technique. The active layers were deposited on a l-in. -diam circle on 0.005-in.-thick platinum. The Pu²⁴⁰ foil contained approximately 1.9 mg of PuO₂ containing less than 0.2% impurity. The Pu²⁴² foil contained 2.1 mg $PuO₂$ composed of 0.39% $Pu²³⁸$,

[[]Work performed under the auspices of the U. S. Atomic Energy Commission. '

¹ The anisotropy is here defined to mean the ratio of the zerodegree differential cross section to the cross section at 90 degrees, $W(0)/W(90)$, sometimes written $(0/90)$.

² James J. Griffin, Phys. Rev. 127, 1248 (1962); see also ac-

companying paper. 'References to earlier work may be found in the following review papers: I. Ha1pern, Ann. Rev. Nucl. Sci. 9, ²⁴⁵ (1959); R. Vandenbosch and J. R. Huizenga, in *Nuclear Reactions*, edited by P. M. Endt and P. B. Smith (North-Holland Publishing Company, Amsterdam, 1962), Vol. II, p. 42.
Company, Amsterdam, 1962), Vol. II, p. 42.
⁴ R. Chaudhr

⁶ Stanley Cohen and W. J. Swiatecki, Ann. Phys. (N. Y.) 19, 67 (1962); 22, 406 (1963).

⁶ J. E. Simmons and R. L. Henkel, Phys. Rev. 120, 198 (1960). ⁷ We are indebted to Dr. George Rogosa for supplying us with the Pu²⁴⁰ and to Dr. Paul Fields for the Pu²⁴².

D.J. Carswell and J. Milsted, J. Nucl. Energy 4, ⁵¹ (1957).

^a See Ref. 1.

 0.226% Pu²³⁹, 7.51% Pu²⁴⁰, 3.33% Pu²⁴¹, and the remainder (88.51%) Pu²⁴². The amount of Pu²⁴ available to us was appreciably smaller than for the other two isotopes. The Pu²⁴¹ foil contained 0.8 mg of the oxide, composed of 1.40% Pu²³⁹, 2.29% Pu²⁴⁰, 0.20% Pu²⁴², and 96.11% Pu²⁴¹.

RESULTS

The results of this experiment are given in Table I for the Pu²⁴⁰, Pu²⁴², and Pu²⁴¹ isotopes. The neutron energy E_N and half-energy spread $\Delta \overline{E}$ are listed in the first column. The following four columns give the experimental values of $W(\theta)/W(90)$, which defines the relative angular distributions of the fragments. The fifth column, labeled $(0/90)$ -Fit provides an extrapolation to zero degrees by an even Legendre polynomial function.⁶ The standard deviations associated with the data are predominantly statistical in origin. The errors listed under (0/90)-Fit are arbitrarily taken to be the experimental errors listed under the (10/90) column. There were two sources of backgrounds that were taken into account: At deuteron energies between 5.5 and

FIG. 1. Schematic diagram of the fission counter.

8.0 MeV, a measurable neutron flux is generated in the target assembly; this source produced an average background of 2.5% of the fission rate. Second, a spontaneous fission rate is present for Pu^{240} and Pu^{242} ; for Pu²⁴² the spontaneous fission contributed 2.5% to the total rate, and less for Pu^{240} .

Certain corrections must be applied to the data after background subtraction. Let $I(\theta)$ be the background-subtracted intensity at the angle θ , where θ is very closely the same for laboratory or c.m. coordinates. Then the relative angular distribution in the c.m. frame is defined to be

$$
\frac{W(\theta)}{W(90)} = \frac{I(\theta)}{I(90)} \times \frac{d\mu_L}{d\mu_{\text{c.m.}}} \times \frac{F_R}{F_{\Omega}}.
$$
 (2)

The correction factors entering this relation have the following meanings: The quantity $(d\mu_L/d\mu_{\rm o.m.})$ is the lab to center-of-mass (c.m.) solid angle reduction factor; at 8 MeV it attains its smallest value of 0.973. The F_R factor corrects for resolution effects⁶ which derive primarily from the nonuniform flux illumination of the fission foil, and secondarily from the finite angular opening of the counters. One set of values was used for all the data, namely $F_R = 1.0000, 1.0029, 1.0089, 1.0196,$ and 1.0196 at angles $\theta = 90, 67.5, 45, 22.5,$ and 10 degrees, respectively. F_{Ω} is the relative solid-angle factor which was measured experimentally. This factor measures any deviation of the solid angles of the apertures from their design values. In this experiment, it was measured in two ways: For the even-even isotopes, alpha particles were counted in the proportional counters with appropriate precautions taken for the high rates of decay. In the case of Pu²⁴¹, F_{Ω} was measured by using the thermal fission cross section to obtain a solid-angle calibration with very low-energy neutrons. This was done by surrounding the counter by two cubic feet of paraffin, as described previously, δ and bombarding the entire assembly by 2.5-MeV neutrons. The resulting increase in fission rate—^a factor of ⁸⁷—was assumed to be induced by very low-energy neutrons impinging on the foil from random directions in space. The values of relative solid angle factors are listed in Table II; it is

TABLE II. The relative solid angle correction factors F_{Ω} .

Isotope	Method	(67.5/90)	(45/90)	(22.5/90)	(10/90)
P_{11}^{241}	Paraffin run	0.9916	0.9698	0.9893	0.9803
P_{11}^{241}	Alpha decay	0.9911	0.9716	0.9917	0.9769
P_{11}^{240}	Alpha decay	0.9997	0.9825	1.0019	0.9730
P_{11} ²⁴²	Alpha decay	0.9999	0.9814	1.0024	0.9750

seen that there is agreement on the average to roughly ± 0.005 between measurements made by different methods and different isotopes.

It was estimated that the resolution and solid angle factors each contributed $\frac{1}{2}\%$ relative error to the data, and these were compounded with the statistical errors. It is to be noted that corrections for isotopic impurities in the foils were made in one case only: that of Pu^{242} at $E_N=0.65$ MeV, where the correction resulted in a rise of 5% for the anisotropy. The corrections at other energies and isotopes were less than the experimental errors and were not made.

Figure 2 displays the trend of the anisotropy for Pu²⁴⁰ and Pu²⁴² as function of neutron energy. The shape of the curves shows the usual behavior, i.e., almos constant values for neutron energies less than 5 MeV, followed by a rise caused by fission occurring after neutron emission. It may be noted that there is a lack of strong fluctuations in the anisotropy at low neutron energy, near the Gssion threshold. This behavior is in

FIG. 2. Fission fragment anisotropy in neutron-induced fission of Pu²⁴⁰ and Pu²⁴². Comparison to U²³⁸ is shown in the insert.

FIG. 3. Fission fragment anisotropy in the neutroninduced fission of Pu²⁴¹.

contrast to that of thorium and uranium even-even isotopes, which often show spectacular variations near threshold. The insert in Fig. 2 indicates the comparison to U^{238} . The values of anisotropy for Pu^{241} are shown in Fig. 3. The errors for this isotope were larger than for the other two owing to the smaller mass available on the foil. Ke do not comment further on the data for this isotope, except to note that the fission cross section has recently been measured⁹ for it.

DISCUSSION

Comparison of the angular distribution data for Pu²⁴⁰, Pu²⁴², and various other even-even $(e-e)$ isotopes are made by means of the parameter K_0^2 , which is related to the anisotropy by Eq. (1). The maximum value of orbital angular momentum is given by Leachman and Sanmann¹⁰ to be $L_m^2=4.2 E_N$, which we use here. The quantity K_0^2 is assumed to be a function of excitation energy, $E^* = E_N - E_F$, where E_N is the neutron energy and E_F is the neutron energy corresponding to the "fission threshold." The values of E_F are chosen to correspond to the 10% point on the fission excitation curve. This convention should provide consistent values to within a few hundred kilovolts; it corresponds to our earlier usage' for other ^e—^e targets. corresponds to our earlier usage⁶ for other e—e targets.
From excitation curves^{11,12} for Pu²⁴⁰ we choose $E_F{=}0.35$ MeV and from data¹³ for Pu²⁴² we choose $E_F = 0.45$ MeV. In Fig. 4, values of K_0^2 are plotted versus E^* for a number of e-e targets. The range of E^* varies from 1.5 MeV above the fission threshold to energies below the onset of n, n' fission. The values for the uranium isotopes come from Ref. 6. Recently, accurate data for U^{234} have become available from Lamphere¹⁴; however, they have not been included here. The Th²³² datum is

for bringing these references to our attention. '3 Daniel K. Butler, Phys. Rev. 117, 1305 (1960). '

derived from a measurement of Blumberg.¹⁵ Earlie data¹⁶ for Th²³² and data⁶ for Th²³⁰ have not been included because of their relatively large uncertainties. The straight lines appearing in the figure represent linear least-squares fits to the data of the various target isotopes, with the exception of $Th²³²$. The $K₀²$ values for the plutonium isotopes fall significantly higher than those for the uranium and thorium data. It had been noticed earlier that the uranium data seemed to group rather closely to one line; it is somewhat surprising that the Th²³² datum point falls close to the uranium data. In any case, the data of Fig. 4 indicate that excitation energy alone is not sufficient to correlate values of K_0^2 for this group of ^e—^e targets.

In Fig. 5, values of K_0^2 are plotted against Z^2/A of the compound nucleus at fixed E^* , to display a possible correlation with fissionability. The plotted points for the ^e—^e targets are taken from the straight-line fits of Fig. 4, at $E^*=4$ MeV, the highest excitation energy available to all the data. The errors were assigned to be $1/\sqrt{2}$ times the average of the nearest two experimental errors in K_0^2 . The plot suggests two groupings: that of the uranium and thorium data for which $K_0^2 \approx 16$, and that for the plutonium points for which K_0^2 is close to

FrG. 4. K_0^3 as function of excitation energy, $E^* = E_N - E_f$, for Pu²⁴⁰, Pu²⁴², and other even-even target nuclei. The uranium data are taken from Ref. 6. The Th²³² datum comes from Ref. 15. Straight-line fits have been made for the data of each nucleus and are shown in the figure.

^{&#}x27; H. L. Smith, R. K. Smith, and R. L. Henkel, Phys. Rev. 125, 1329 (1962).

¹⁰ R. B. Leachman and E. E. Sanmann, Ann. Phys. (N. Y.)

^{18, 274 (1962).&}lt;br>, " D. J. Hughes and R. B. Schwartz, BNL 325, 2nd ed., 1958 (unpublished).

^{1958 (}unpublished).
¹² V. G. Nesterov and G. N. Smirenkin, Zh. Eksperim. i Teor.
¹² Z. 35, 532 (1958) [English transl.: Soviet Phys.—JETP 8,
367 (1959)]; and At. Energ. 9, 16 (1960) [English transl.: Soviet
J. At. Ener

¹⁴ R. W. Lamphere, Nucl. Phys. 38, 561 (1962).

¹⁵ Leroy N. Blumberg, thesis, Columbia University, 1962 (unpublished), and accompanying paper of R. B. Leachman and L. Blumberg. The value of K_0^2 for Th²³² plotted in Fig. 4 was derived from $W(0)/W(90)$ as extrapolated to zero degrees by a leastsquares fit to the data which is illustrated in Fig. 12 of Blumberg's thesis.

¹⁶ R. L. Henkel and J. E. Brolley, Jr., Phys. Rev. 103, 1292 (1956).

FIG. 5. K_0^2 as function of (Z^2/A) compound at $E^*=4$ MeV for even-even and even-odd target nuclei. The ^e—^e points are taken from the straight-line fits of Fig. 4. Errors given with the points are discussed in the text. A linear function has been fitted to the $e-e$ discussed in the text. A linear function has been fitted to the e-e
data with the result $K_0^2 = (-117.7 \pm 5.9) + (3.8 \pm 1.7) \times (2^2 / A)$.
Note that the right-hand scale has been located such that the U¹⁸⁸ value of K_0^2 falls on the straight line through the e-e points.

23.5. Nevertheless, an attempt was made to fit these data by a linear function of Z^2/A . The result is

$$
K_0^2(E^*=4 \text{ MeV}) = (-177.7 \pm 5.9) +(3.8 \pm 1.7)(Z^2/A).
$$
 (3)

The value of χ^2 , with 4 degrees of freedom, for this fit is 5.4, corresponding to the probability of 25% that χ^2 could be larger than this value. The fit deserves our consideration even though χ^2 is on the high side. The probability that the data are fitted by a constant is much smaller; χ^2 becomes 12.5, with a 3.3% probability that x^2 is greater than this value.

The present results have a bearing on the target spin effect for even-odd targets, which has recently been investigated by Leachman and Sanmann.¹⁰ Bohr¹⁷ originally suggested that the presence of large spin in the target nucleus should tend to reduce the anisotropy of the fragments. The even-odd targets, Pu²³⁹, U²³³, and U^{285} with spins of $\frac{1}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$, respectively, are the usual examples in neutron-induced fission. The trend of the experimental data is reversed from that expected, since the anisotropy of Pu^{239} is smaller than that of U^{235} . In terms of K_0^2 , the value for Pu²³⁹ is largest and that of

U²³⁵ is smallest, at a given excitation energy. Leachman U^{235} is smallest, at a given excitation energy. Leachman and Sanmann,¹⁰ in their investigation of the process found indeed that the spin-induced effects were small. They ascribed part of the observed increase of K_0^2 to a dependence on Z^2/A , following the work of Chaudhry, Vandenbosch, and Huizenga.⁴ The major part of the increase in K_0^2 was explained by use of an angular momentum-dependent fission mechanism.

The slope of K_0^2 as a function of Z^2/A for the eveneven targets, given in Eq. (3), shows the same qualitative behavior as do the experimental values of K_0^2 for Pu²³⁹, U²³³, and U²³⁵. Taking Eq. (3) at face value, we assume that the slope represents even-odd targets as well as the even-even ones, from which it was obtained. Estimates of K_0^2 for Pu²³⁹, U²³³, and U²³⁵ have been made from the experimental data of Refs. 6, 15, and 18, at $E^*=4$ MeV. The procedure used was similar to that described above; however, the large number of points enabled the errors to be obtained from the least-squares fitting process. These estimates are entered in Fig. 5 as dashed vertical bars, plotted against a K_0^2 scale (on the right-hand side of the figure) which has been displaced vertically and normalized, so that the K_0^2 point for U^{233} falls on top of the straight line through the even-even data points. It is seen that the slope of Eq. (3) accounts for the major part of the change of K_0^2 from U^{235} to Pu²³⁹. This observation is intended to emphasize the possible role of fissionability with regard to the spin effect; however, the data do not allow a quantitative statement.

In conclusion, it is noted that the variation in K_0^2 with Z^2/A agrees qualitatively with that predicted by Cohen and Swiatecki.⁵ The parameter K_0^2 can be expressed¹⁹ in terms of the effective moment of inertia I_{eff} by $I_{\text{eff}}T = \hbar^2 K_0^2$, where T is the nuclear temperature, assumed to be constant. The variation in I_{eff} versus Z^2/A calculated by Cohen and Swiatecki shows about a 10% increase of I_{eff} per unit change in Z^2/A in the region $Z^2/A = 36$, compared to a $(20 \pm 9)\%$ increase in K_0^2 predicted by Eq. (3). Cohen and Swiatecki have pointed out that above $Z^2/A \approx 33.5$ (X = 0.67) the lack of a definite necking-in of the saddle-point shape gives rise to uncertainty in the final outcome of the fission process, as described by the liquid-drop theory. The comparison noted above must be qualified accordingly.

ACKNOWLEDGMENTS

We are indebted to Mrs. Patricia Stein for undertaking foil preparation for us.

 17 Aa. Bohr, in *Proceedings of the International Conference on the* Peaceful Uses of Atomic Energy, Geneva, 1955 (United Nations
New York, 1956), Vol. 2, p. 151.

 18 L. Blumberg and R. B. Leachman, Phys. Rev. 116, 102 (1959). ¹⁹ I. Halpern and V. M. Strutinski, in Proceedings of the Second United Nations International Conference on the Peaceful Uses of
Atomic Energy, Geneva, 1958 (United Nations, Geneva, 1958), Vol. 15, p. 408.