Symmetric-to-asymmetric mass ratios for proton-induced fission of ^{232}Th and ^{238}U

R. Eaker, H. S. Plendl, and A. F. Zeller
The Florida State University, Tallahassee, Florida 32306

Alex Clem, Luis Muga, and J. M. Nicovich
The University of Florida, Gainesville, Florida 32611
(Received 14 May 1979)

Fragment-energy and specific-luminescence spectra from the fission of ^{232}Th and ^{238}U were measured by a two-dimensional display of coincident signals from a solid state detector and a thin-film organic scintillator detector. Symmetric-to-asymmetric fission ratios and peak-to-valley ratios were determined as a function of bombarding energy. Discontinuities in the ratio curves, observed in some previous experiments but not in others, were mapped out in detail. The major discontinuities were found to occur near fission thresholds.

NUCLEAR REACTIONS: FISSION ^{232}Th (p,f), ^{238}U (p,f), $E=9.5$–17.0 MeV; measured E, specific luminescence spectra of single fragments; Si detector coin thin-film plastic scintillator detector; deduced symmetric-to-asymmetric mass ratio trends, competing reaction thresholds.

The excitation functions for proton-induced fission of ^{232}Th and ^{238}U, as well as of other uranium isotopes and of ^{239}Pu, have been determined by several groups at incident proton energies from 3 to 55 MeV.[1-6] Most symmetric-to-asymmetric (N_S/N_A) or peak-to-valley (P/V) ratios of fission fragments as a function of incident proton energy show distinct breaks at certain energies[1-3,5,6] that have been interpreted as the opening of another reaction channel, such as fission following emission of one or two neutrons (second- or third-chance fission). Some of the observed breaks, however, have been rather weak, and in one study[4] no definite breaks were observed at all over the range of incident energies investigated.

The objective of the present study was to determine the existence and position of such breaks with a more sensitive detector system than was previously used. The fission of ^{232}Th and ^{238}U induced by 9.5–17.0 MeV protons was studied. Ratio curves were determined from single-parameter spectra and contour plots of the coincident response of a thin-film detector (TFD) and a solid state detector (SSD) to the fission fragments.

Natural ^{232}Th and ^{238}U targets (0.3 mg/cm^2 $^{232}ThF_4$ and 0.01 mg/cm^2 $^{238}U_3O_8$ deposited on 0.1 mg/cm^2 nickel supporting foil) were bombarded with protons from the Florida State University FN tandem accelerator. The target chamber is described in Ref. 7. The TFD/SSD assembly used in this work was positioned at 140°, and the normal-to-the-target plane was 156° with respect to the beam. The beam current was held to 30–150 nA, resulting in TFD-SSD count rates of the order 1 to 10^3 counts per second, depending on proton energy and target material.

The basic features of the TFD have been described previously.[7-12] Its response measures the luminescence produced by an energetic ion passing through a thin plastic scintillator film. This response has been shown to be a function of nuclear charge and velocity.[9,10] The TFD's used in this experiment were made of NE-102 plastic scintillator laminated to a thickness of 100 μg/cm^2 and supported on a thin rectangular celluloid frame. The solid state detector was of the silicon surface-barrier type designed for fission fragment detection. Coincident TFD-SSD signals, as gated by a logic pulse generated by the SSD, were digitized and passed through a CAMAC interface to an on-line computer. Further details of the experimental setup and of the associated electronics are given in Ref. 13.

Dual-parameter plots of the TFD-SSD response to fission fragments were recorded as a function of incident proton energy from 9.5 to 17.0 MeV at 125 to 500 keV intervals. For comparison, ^{252}Cf spontaneous fission contour plots were recorded. Two different TFD's of comparable thickness were used and exhibited P/V ratios of 2.5 and 4.5, respectively, to ^{252}Cf fission fragments. Representative contour plots for ^{232}Th and ^{238}U fission induced by 18.0 MeV protons and for ^{252}Cf spontaneous fission[13] are shown in Figs. 1(a), 1(b), and 1(c), respectively. Note that the SSD signal response is proportional to fission fragment kinetic energy and the TFD signal response is a measure of the (ionic) fission fragment specific lumine-

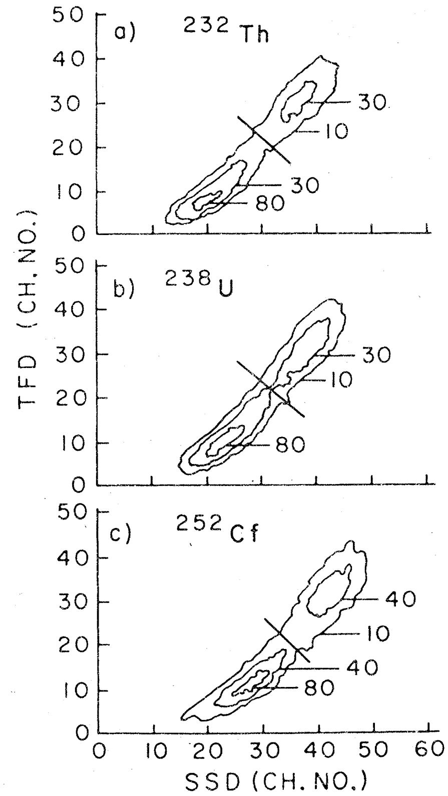

FIG. 1. Representative contour plots of TFD-SSD system response to fission fragments. The ~45° solid line follows the locus of minima between the two main peaks and is used as an arbitrary separation line between the purely asymmetric fission events (upper peak) and mixed symmetric/asymmetric fission events (lower peak). Note the change in peak position for the different isotopes. The contour lines are drawn at 10, 30, 40, and 80%, respectively.

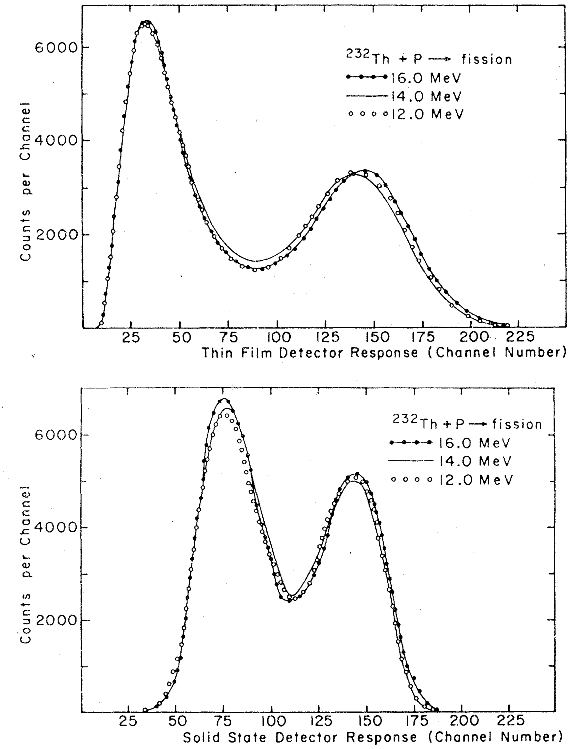

FIG. 2. (a) TFD and (b) SSD fission-fragment single-parameter spectra from ^{232}Th fission. The filling-in-of-the-valley region observed in going from 12 to 14 MeV in Fig. (a) is not as pronounced for the 16 MeV data and results from third-chance fission competition (see text).

scence. In the range of fission fragment energies, this latter parameter has been shown to be approximately proportional to the square of the fragment velocity and to increase with increasing nuclear charge.[10] Typical single-parameter TFD and corresponding SSD response data are shown in Fig. 2 for ^{232}Th fission. Similar data were obtained for ^{238}U fission.

In the single-parameter response data, the lower (left-hand) response peak is in general due to the less energetic heavy fragment and the higher response peak to the light fragment from asymmetric fission events. Fragments from symmetric fission are recorded in the valley between peaks and the slope just to its left and may be attributed largely to the lower *total* kinetic energy release for symmetric fission (~11 MeV less at 13 MeV proton energy[14]) that accompanies near-symmetric mass division. The P/V ratios taken from these spectra should, therefore, be indicative of the ratio of asymmetric-to-symmetric fission, even though, at first glance, little variation is evident over the 4 MeV range of incident proton energy.

A more sensitive measure of symmetric vs asymmetric fission competition may be derived from the two-parameter contour plots [e.g., Figs. 1(a) and 1(b)]. The locus of minima (solid, straight line) between the two peaks separates the asymmetric events (A_1) accumulated in the upper right-hand peak from the asymmetric plus symmetric events (A_2) registered in the lower left-hand peak. The criterion for this separation follows from the previously noted fact that symmetric fission events will be recorded predominantly in the lower response peak and valley region. Hence the number of symmetric events recorded is equal to the difference $A_2 - A_1$, and the number of asymmetric events registered during the same time is $2A_1$. A measure of the symmetric-to-asymmetric fission ratio is then given by

$$\frac{N_S}{N_A}=\frac{A_2-A_1}{2A_1}\ .$$

This ratio is not taken as an absolute measure of the symmetric-to-asymmetric fission ratio but is only used to indicate trends in the *relative* contributions. This limitation arises not only from the inherent instrumental resolution, but also from both the gradual transition between symmetric vs asymmetric mass division and the somewhat arbitrary manner of isolating the two modes on the contour plot. A comparison of the symmetric vs asymmetric trends as indicated by the P/V and N_S/N_A ratios is shown in Figs. 3 and 4.

Assuming a two-mode fission hypothesis,[15] the P/V and N_S/N_A ratios have been interpreted in terms of a symmetric vs asymmetric fission mode competition at different incident proton energies. Symmetric fission becomes more probable with increasing excitation energy,[16] thereby decreasing the P/V ratio and increasing the N_S/N_A ratio. Abrupt changes observed in the slope of the ratio curves at certain bombarding energies are interpreted as resulting from the onset of second- or third-chance fission, i.e. (p,nf) and $(p,2nf)$, or of competing reactions. Since those residual nuclei that undergo second- or third-chance fission do so from a lower excited state, *an increase in the P/V ratio* and *a decrease in the* N_S/N_A *ratio* is expected as the bombarding energy passes above the threshold leading to this process. In the case of competing reactions which do not lead to fission but which reduce the fractional fission cross section, one might expect a smaller effect. The discontinuities observed in this and in previous work are listed in Table I.

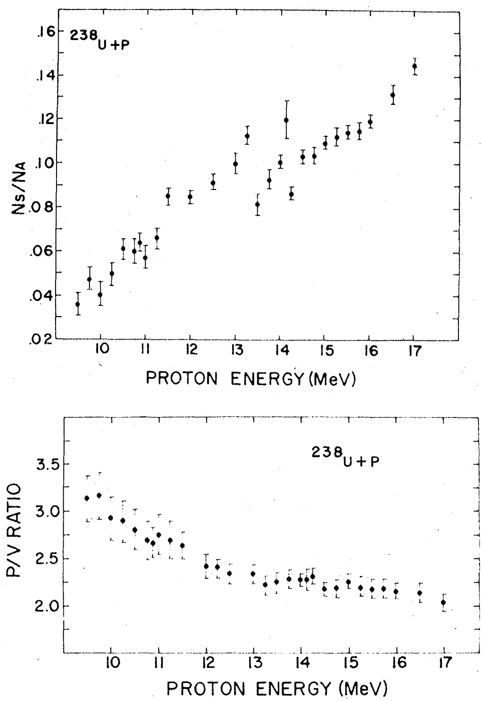

FIG. 4. (a) N_S/N_A ratios and (b) P/V ratios vs incident energy for $^{238}U+p$

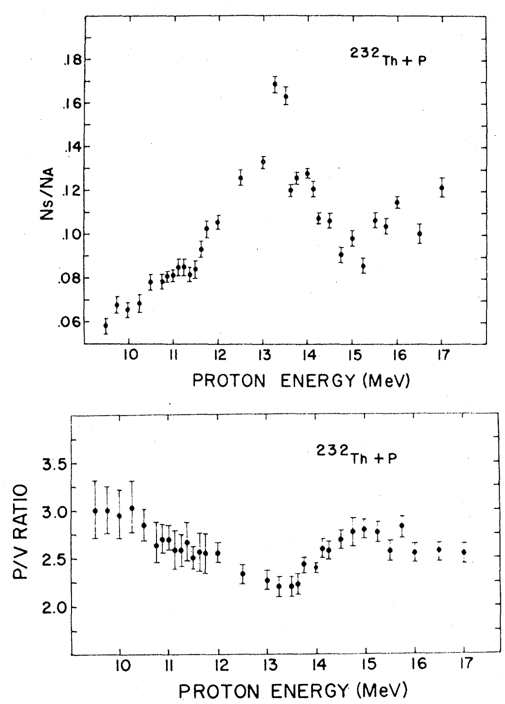

FIG. 3. (a) N_S/N_A ratios and (b) P/V ratios vs incident energy for $^{232}Th+p$.

For proton-induced fission of ^{232}Th, evidence for a major discontinuity is clearly present [decrease in N_S/N_A ratio, Fig. 3(a) and increase in P/V ratio, Fig. 3(b)], but it occurs at a lower energy (13.5 MeV) than the break previously measured by radiochemical methods (14.0 MeV).[1] This discontinuity is interpreted as arising from the

TABLE I. Observed reaction onsets, $E_P=9.5-17.0$ MeV.

	This work	Ref. 1	Refs. 2, 3, and 5
$^{232}Th+p$	11.25 ± 0.10 [a]	...	11.25 ± 0.2
	$13.5\ \pm0.2$	14.0 ± 0.5	c
	$16.2\ \pm0.3$ [b]	...	c
$^{238}U\ +p$	$10.5\ \pm0.2$ [b]	...	$10.6\ \pm0.1$
	$13.5\ \pm0.3$	13.0 ± 0.5	c

[a] Minor discontinuity.
[b] Inconclusive evidence.
[c] Outside of range of that experiment.

onset of third-chance fission. A detailed study was made in the energy range from 10 to 12 MeV in search for a decrease in the N_S/N_A curve at 11.25 MeV, where a break had been observed previously.[2,3] Only a slight decrease was found in the otherwise smoothly increasing curve. The authors of the previous studies considered the possibility that this discontinuity is due to the $(p,p'n)$ or $(p,3n)$ reaction onset.[2] But the Q value for the $(p,p'n)$ reaction is −6.43 MeV and the most probable kinetic energy for an emitted proton is 1.1 MeV,[17] so that the energy left for the proton is quite low compared to the Coulomb barrier. The $(p,3n)$ Q value is −13.6 MeV, which is too high to cause the 11.25 MeV break. A more likely candidate for that threshold would be a break that may be present near 16.2 MeV [Fig. 3(a)].

For ^{238}U proton-induced fission, similar though less distinct breaks occur in the N_S/N_A and P/V ratio curves (Fig. 4). A definite discontinuity is observed again near 13.5 MeV, confirming the existence of a discontinuity near 13 MeV suggested by radiochemical data and assigned to the onset of third-chance fission. A break in the vicinity of 13 MeV was also reported in Ref. 6. A previously reported minor break near 10.5 MeV is only weakly suggested in our Fig. 4 and also in the data of Ref. 4. As in the case of the 11.25 MeV break in the ^{232}Th$+p$ curves, the $(p,p'n)$ and $(p,3n)$ reaction onsets considered in Refs. 2, 3, and 5 (Q = −6.10 and −13.03 MeV, respectively) are unlikely causes of the discontinuity.

In comparing the data of Figs. 3(a) and 3(b) and also the data of Figs. 4(a) and 4(b), it is seen that for a given number of recorded events the N_S/N_A curve is more sensitive than the P/V curve for distinguishing trends in symmetric vs asymmetric fission yields and thus is a more precise measure for locating abrupt changes in this ratio.

The observed trend that the breaks in the ^{238}U data are consistently less clearcut than in the ^{232}Th data can be understood as follows: As the mass of the fissioning target nucleus increases, the positions of the two response peaks tend to converge, and the sensitivity of the P/V or N_S/N_A ratios for following two-mode fission patterns decreases correspondingly.

To summarize, the major breaks in both the ^{232}Th and the ^{238}U excitation functions for proton-induced fission from $E_p = 9.7$ to 17 MeV are ascribed to the onset of third-chance fission. (The first- and second-chance fission onsets[1,3] are below the lower limit of the present study.) The other observed breaks could not be assigned definitively to reaction thresholds.

This work was supported in part by the U.S. Department of Energy and the NSF.

[1] J. P. Butler, B. J. Bowles, and F. Brown, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1958 (United Nations, Geneva, *1958*), Vol. 15, Phys. Rev. 107, 751 (1957).
[2] J. R. Meriwether, Ph.D. dissertation, Florida State University, 1963 (unpublished).
[3] G. R. Choppin, J. R. Meriwether, and J. D. Fox, Phys. Rev. 131, 2149 (1963).
[4] G. L. Bate and J. R. Huizenga, Phys. Rev. B 133, 1471 (1963).
[5] A. T. Kandil, Ph.D. dissertation, Florida State University, 1969 (unpublished).
[6] J. R. Boyce *et al.*, Phys. Rev. C 10, 231 (1974).
[7] J. M. Nicovich *et al.*, Nucl. Instrum. Methods 157, 93 (1978).
[8] L. Muga *et al.*, Nucl. Instrum. Methods 111, 581 (1973).
[9] L. Muga and G. Griffith, Phys. Rev. B 9, 3639 (1974).
[10] L. Muga *et al.*, in *Proceedings of Third International Symposium on the Physics and Chemistry of Fission, Rochester, 1973* (International Atomic Energy Agency, Vienna, 1974), Vol. II, p. 451.
[11] L. Muga *et al.*, Nucl. Instrum. Methods 104, 605 (1972); L. Muga and G. Griffith, *ibid.* 109, 289 (1973); L. Muga and D. J. Burnsed, Rev. Sci. Instrum. 47 (No. 8), 924 (1976).
[12] W. J. McDonald *et al.*, Nucl. Instrum. Methods 115, 185 (1974).
[13] L. Muga *et al.*, Nucl. Instrum. Methods 119, 255 (1974).
[14] I. F. Croall and J. G. Cunninghame, Nucl. Phys. A125, 402 (1969).
[15] H. C. Britt, H. E. Wegner, and J. C. Gursky, Phys. Rev. 129, 2239 (1963).
[16] L. J. Colby *et al.*, Phys. Rev. 121, 1415 (1961).
[17] J. M. B. Lang and K. J. LeCouteur, Proc. Phys. Soc. London, A67, 586 (1954).