shown in Table II, the level energies follow quite closely a relation

$$E_{ex}=n^2\times E_0,$$

where $n$ is an integer and $E_0$ a constant energy. Except for the 1.12-, 1.53-, 1.92-, and 2.04-Mev levels, $E_0$ is $0.036\pm0.001$ Mev, with $n=3$, 4, 5, 6, and 7, while for the above-mentioned four levels $E_0$ is $0.031\pm0.001$ Mev, with $n=6$, 7, and 8. The 1.92–2.04 Mev levels seem to be "double." The same kind of regularity has been noted in the level structures of $Fe^{57}$, $Ni^{59}$, $Ni^{61}$, $Zn^{67}$, and $Zn^{65}$.[7]

TABLE II. Levels $E_{ex}=n^2\times E_0$ in $Ga^{69}$.

| $E_{ex}$ (kev) | $n$ | $E_0$ (kev) | |
|---|---|---|---|
| 324 | 3 | 36 | |
| 576 | 4 | 36 | |
| 878 | 5 | 35 | |
| 1120 | 6 | ... | 31 |
| 1340 | 6 | 37 | ... |
| 1530 | 7 | ... | 31 |
| (1720) | 7 | 35 | ... |
| 1920 | 8 | ... | 30 |
| 2040 | 8 | ... | 32 |


## ACKNOWLEDGMENTS

The authors are indebted to Professor Allan C. G. Mitchell for his advice and interest in this work.

We wish to thank Mr. George Bulbenko for the chemical separations of our sources, Dr. M. Sampson, and the cyclotron staff for the many bombardments, and Dr. H. J. van den Bold for his communications of preliminary results of a simultaneous investigation of $Ge^{69}$ in Utrecht, The Netherlands. One of the authors (R.H.N.) also gratefully acknowledges a Fulbright Travel Grant to the United States.


PHYSICAL REVIEW VOLUME 105, NUMBER 4 FEBRUARY 15, 1957

# Low-Energy Activation Functions for Photofission of $U^{238}$ and $Th^{232}$†

R. A. SCHMITT* AND R. B. DUFFIELD*
*University of Illinois, Urbana, Illinois*
(Received November 13, 1956)

Activation functions for symmetric and asymmetric photofission of $U^{238}$ and $Th^{232}$ have been determined over the energy range 4.5 Mev to 10 Mev. For both elements, the fission yield varied exponentially with betatron energy over the lowest energies covered and the data show the difficulty in establishing experimentally the existence of a threshold for photofission. The symmetric ($Cd^{117}$) photofission yield relative to the asymmetric ($Ba^{139}$) yield in $U^{238}$ showed a local maximum of 0.05% at 6-Mev betatron energy, while the corresponding symmetric yield in $Th^{232}$ rose steadily from <0.0003% at 6.5 Mev to 0.1% at 12-Mev betatron energy.

## I. INTRODUCTION

THE liquid drop model has been used to calculate the photofission thresholds of heavy nuclei.[1–4] Experimental determinations of the photofission thresholds[5,6] of several heavy nuclei have given values which in general were below the calculated ones and showed a less strong variation with $Z^2/A$ than that predicted by the model. The thresholds, obtained by use of fission ionization chambers, for $Th^{232}$, $U^{233}$, $U^{235}$, $U^{238}$, and $Pu^{239}$ in reference 6 were $5.40\pm0.22$, $5.18\pm0.27$, $5.31\pm0.25$, $5.08\pm0.15$, and $5.31\pm0.27$ Mev, respectively. Experimental values of the fission thresholds deduced from the behavior of the cross sections for neutron-induced fission of heavy nuclei show a similar disagreement with the calculated values (see Table I of reference 4).

In the present work we have attempted to remeasure the photofission thresholds of $U^{238}$ and $Th^{232}$ with increased sensitivity. This has been done by exposing samples in the circulating electron beam of the University of Illinois 24-Mev betatron in order to increase the beam intensity and separating radiochemically selected fission products from quite massive targets in order to measure the number of fissions. This method of measurement had the distinct disadvantage that it utilized a thick target bremsstrahlung spectrum which precluded the possibility of calculating accurately the cross sections in the low-energy region.

A second object of the present experiment has been to measure the difference in thresholds, if any, for asymmetric and symmetric fission. Previous research has shown[7,8] that the photofission of uranium, like

† This work was supported by the Office of Naval Research.
* Present address: General Atomic, San Diego, California.
[1] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
[2] Arakatu, Uemura, Sonoda, Shimiyu, Kimura, and Muraoka, Proc. Phys. Math. Soc. Japan **23**, 440 (1941).
[3] S. Frankel and N. Metropolis, Phys. Rev. **72**, 914 (1947).
[4] D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).
[5] H. W. Koch, Ph.D. thesis, University of Illinois, 1944 (unpublished).
[6] Koch, McElhinney, and Gasteiger, Phys. Rev. **77**, 329 (1950).
[7] R. A. Schmitt and N. Sugarman, Phys. Rev. **95**, 1260 (1954).
[8] Katz, Kavanagh, Cameron, Bailey, and Spinks, Phys. Rev. **99**, 98 (1955).

neutron-induced fission, becomes more symmetric as the excitation energy is increased. The data of Katz *et al.*[8] covered the energy region from 12 to 24 Mev for uranium. Recent data[9] on the angular anisotropy of the fragment distribution from photofission at low energies suggested that a correlation of this effect with the symmetric and asymmetric fission probabilities near threshold might be informative.

## II. EXPERIMENTAL PROCEDURE

### A. Probe and External Target Irradiations

Metallic slugs of natural uranium and thorium,[10] 1/4 in. in diameter by 5/8 in. in length, were exposed[11] in the circulating electron beam of the University of Illinois 24-Mev betatron. The electrons, entering the end of the slug, generated a thick target bremsstrahlung which sprayed the slug longitudinally in a rather broad cone. The escaping x-rays were monitored by an aluminum wall, air-filled ionization chamber placed two meters from the target. Irradiation times were usually twice the half-life of the isolated radioactive fission product.

At higher energies, irradiations were made in the external bremsstrahlung beam generated by a thin electron target. Twenty-gram samples of analytical reagent grade uranyl nitrate, wrapped with 0.015-in. cadmium foil, or 1/4-in. diam by 5/8-in. long metallic thorium slugs were exposed in the beam. The distance from the electron target to the fissionable material was 25 cm at 6.0 and 6.5 Mev and 99 cm at 8.0 Mev. The x-ray intensity was monitored by an aluminum wall ionization chamber calibrated against a 100-r Victoreen thimble in a Lucite cube 8 cm on an edge. The monitor was 100 cm from the electron target.

### B. Energy Calibration of the Betatron

The energy of the electrons at the time of x-ray production was controlled by an integrator circuit which has been shown to be stable to approximately 30 kev over long periods of time.[12] The integrator was calibrated using the $Cu^{63}(\gamma,n)Cu^{62}$ threshold at 10.73 Mev[13] and the $Be^{9}(\gamma,n)Be^{8}$ threshold at 1.66 Mev.[14] The $Cu^{63}(\gamma,n)Cu^{62}$ threshold was measured repeatedly during this work as a check on the energy stability. It was assumed that the energy varied linearly with the integrator setting between these two calibration points.

The betatron was usually operated so that orbit expansion at these low energies occurred at 30° or less of the magnetic cycle. If the expander circuit failed to fire, electrons of energy much higher than that chosen could strike the target due to self-expansion of the orbit later than 30°. In order to prevent this, a "tilt" circuit was built (by R. D. Bentley of the betatron staff) which shut off the injector after one miss of the expander. It was found, however, that the expander circuit was very reliable.

### C. Radiochemical Separations and Activity Determinations

#### *Uranium Probe Targets*

For the separation of the fission products, $Ba^{139}$ and $Cd^{117}$, the metallic uranium slugs were dissolved after irradiation in 25 ml of 11$N$ HCl. After dissolution, 5 ml of 15$N$ $HNO_3$ was added to effect a clear solution, followed by the addition of 20 mg each of barium and cadmium carriers. Standard radiochemical analyses[15] were performed, with slight modifications because of larger amounts of uranium.

For the separation of the fission products, $Br^{83,84}$, together with $Ba^{139}$, the uranium metal slugs were dissolved after irradiation in a flask (with reflux condenser) that contained 25 ml of 11$N$ HCl and 20 mg each of barium and bromide carriers. It was assumed that all of the bromide fission product was reduced to its lowest oxidation state by the metallic uranium. After dissolution, 5 ml of 15$N$ $HNO_3$ was slowly added to oxidize the bromide to the elemental state. A $CCl_4$ extraction removed the $Br_2$, after which standard radiochemical analyses[15] followed. For validity of the threshold determinations, it is only necessary that the recovery of bromide fission product be reproducible, not that it be quantitative. Reproducibility to 10% was established by repeated test runs at 6.0 Mev.

Uranyl nitrate targets were dissolved in dilute $HNO_3$ containing 20 mg of barium carrier. Standard radiochemical analyses were performed on the solution.[15]

#### *Thorium Probe Targets*

When $Sr^{91}$ and $Cd^{117}$ were isolated, the metallic thorium slugs were dissolved after irradiation in 25 ml of 11$N$ HCl and 0.5 ml of 0.2$N$ $(NH_4)_2SiF_6$. After dissolution, 20 mg each of strontium and cadmium carriers were added. The customary radiochemical analyses[15] followed.

For the separation of $I^{134}$, the thorium slugs were dissolved after irradiation in 25 ml of 11$N$ HCl, 0.5 ml of 0.2$N$ $(NH_4)_2SiF_6$, and 20 mg of iodide carrier. After dissolution, the solution was diluted to less than 3$N$ in HCl, and then the iodide was oxidized to its elemental state by $KMnO_4$. An extraction by $CCl_4$, followed by

[9] Winhold, Demos, and Halpern, Phys. Rev. **85**, 728 (1952); **87**, 1139 (1952); E. J. Winhold, Ph.D. thesis, Massachusetts Institute of Technology, 1953 (unpublished); Fairhall, Halpern, and Winhold, Phys. Rev. **94**, 733 (1954).

[10] These were supplied by the Argonne National Laboratory and with the cooperation of L. E. Glendenin and E. P. Steinberg.

[11] R. A. Becker, Rev. Sci. Instr. **22**, 773 (1951).

[12] B. M. Spicer and A. S. Penfold, Rev. Sci. Instr. **26**, 952 (1955).

[13] M. Birnbaum, Phys. Rev. **93**, 146 (1954).

[14] R. C. Mobley and R. A. Laubenstein, Phys. Rev. **80**, 309 (1950); Noyes, Van Hoomissen, Miller, and Waldman, Phys. Rev. **95**, 396 (1954).

[15] *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

the usual radiochemical procedures,[15] was performed. It was shown that approximately the same fraction (if any) of iodine was lost in the dissolution process for all the iodine experiments.

When $Sr^{91}$ and $Br^{83,84}$ were isolated, the thorium slugs were dissolved after irradiation in a flask (with reflux condenser) that contained 25 ml of 11$N$ HCl, 0.5 ml of 0.2$N$ $(NH_4)_2SiF_6$, and 20 mg each of strontium and bromide carriers. After dissolution, the solution was diluted to less than 3$N$ in HCl; excess $KMnO_4$ was then added to oxidize the bromide to the bromate state. Standard radiochemical procedures[15] followed this step.

*Radioactivity Determination*

The final precipitates of about 25 mg were filtered onto 1.8-cm diam filter paper disks, which were subsequently mounted on 1/16-in. aluminum plates. Cellophane of about 2.5 mg/cm$^2$ covered the precipitates. The radioactive samples, all beta emitters, were counted for about three half-lives by end-window, methane flow proportional counters which were operated at 4200 to 4500 v and encased in heavy cast iron shields. A low and stable background count of 10±0.5 per minute was observed.

The symmetric photofission yields of uranium and thorium, defined as the percentage of the total fissions that give two nearly equal masses, are compiled as a function of maximum betatron energy in Fig. 4. Above 7.5 Mev, a smooth curve which was derived from externally irradiated uranium samples has been drawn through the data. Below 7.5 Mev, the smooth uranium curve connects symmetric yields that are preponderantly probe target yields. All of the thorium symmetric yields from 6.4 to 12 Mev are derived from probe target irradiations. Downward directed arrows indicate upper limits.

The number of symmetric fissions produced in each irradiation was measured by isolating and counting $Cd^{117}$. Since the decay chain for mass number 117 is complex and not completely understood, it has not been possible to measure its fission yield directly. Therefore, it has been assumed that the yield in uranium fission of mass number 117 is 0.094% at a betatron energy of 12 Mev. This is the measured yield of $Cd^{115}$ at 12 Mev relative to $Ba^{139}$ at 6.0%; if the $Ba^{139}$ yield were 5.5%,[7,8,16] a systematic error of only 10% would be introduced. The normalization factor, obtained above for the determination of $Cd^{117}$ in uranium fission, is also applicable to the determination of the $Cd^{117}$ yield in thorium fission.

Uranium and thorium targets were irradiated for three hours and $Cd^{117}$ was chemically isolated two hours after the end of the irradiation. This schedule was adhered to as rigidly as possible through all the irradiations, so that in general, no time corrections were necessary in order to normalize the individual runs. Corrections to a few runs were made necessary by betatron scheduling or slight variations in the times required for chemical processing. These were small and were made by assuming that the genetic relation in the chain is: 2.95-hr $Cd^{117}\rightarrow$1.95-hr $In^{117}$. After subtraction of the long-lived $Cd^{115}-In^{115}$ activities, the samples were always observed to decay with an apparent half-life of 4.0±0.2 hr over a 6-hr interval beginning approximately one hour after the last cadmium-indium separation.

It should be noted that the number of asymmetric fissions in $Th^{232}$, represented by the fission product $I^{134}$, is actually due to $Te^{134}$, the parent of $I^{134}$. It was assumed that the independent fission yield of $I^{134}$ was constant over the energy region covered by this experiment. This is partially substantiated by the fact that the $I^{134}$ fraction of the mass-134 chain yield is 0.12 and 0.20 for thermal-neutron fission[17] of $U^{235}$ and 48-Mev x-ray fission[7] of $U^{238}$, respectively.

## D. Measurement of Number of Neutron-Induced Fissions

For the interpretation of some of the results presented below, it is necessary to know that the number of neutron-induced fissions is very small compared to the number of photofissions. This has always been a difficult point to establish conclusively in photonuclear reactions. At energies less than 7 Mev, neutrons can be generated only by the following processes:

(1) $U^{238}(\gamma,n)U^{237}$, threshold energy 6.0 Mev, with uranium target.

(2) $Th^{232}(\gamma,n)Th^{231}$, threshold energy 6.4 Mev, with thorium target.

(3) Photofission, threshold energy less than 6 Mev, or neutron-induced fission in uranium or thorium.

(4) Production of neutrons by $(\gamma,n)$ reactions on the following nuclei contained in the betatron structure; $H^2$, $Be^9$, $C^{13}$, and $O^{17}$. The amount of each of these in the beam is quite small; in particular, $Be^9$ is probably completely absent.

In order to estimate the number of neutron-produced fissions, we have done the following experiments.

A uranium metal slug was irradiated in the circulating electron beam at a betatron energy of 8 Mev and analyzed to determine the number of fissions and the number of neutron captures in $U^{238}$ that had occurred in the slug.[18] Neutron effects are expected to be relatively more important at 8 Mev than at lower energies. The number of fissions was obtained from the number of $Ba^{139}$ atoms produced; the number of neutron captures was obtained from the number of $Np^{239}$ atoms

[16] Gindler, Huizenga, and Schmitt, Phys. Rev. **104**, 425 (1956).

[17] L. E. Glendenin, Ph.D. thesis, Massachusetts Institute of Technology, Technical Report No. 35, July 29, 1949 (unpublished); A. C. Pappas and C. D. Coryell, Phys. Rev. **81**, 329 (1951).

[18] We are indebted to Professor N. Sugarman of the University of Chicago for suggesting this experiment.

produced. The number of neutron captures in $U^{238}$ was found to be 1% of the number of photofissions which had occurred. If we assume that the capture reactions are produced predominantly by thermal neutrons, we can estimate the maximum number of thermal-neutron-induced fissions in the natural uranium target (of $U^{235}$). The thermal-neutron capture cross section of $U^{238}$ is 2.8 barns and the thermal-neutron fission cross section of natural uranium is 3.8 barns.[19] It follows that less than 1% of the observed fissions were produced by thermal neutrons. Since there are strong resonances in both the $U^{238}$ capture cross section and the $U^{235}$ fission cross section in the epithermal energy region, this experiment probably showed that fissions produced by neutrons in this energy range were negligible compared to the photofissions.

Probe irradiations of uranium were made in which a metal bar was struck by the circulating electron beam as usual but, in addition, a second uranium metal bar was held parallel to the primary one and at a distance of 1.7 cm outside the electron orbit. This second bar was irradiated by a very small fraction of the x-rays produced in the primary target; however, both should have been exposed to approximately the same flux of thermal or epithermal neutrons. Also both bars should have been exposed to approximately the same flux of fast neutrons from the betatron structure. The number of fissions which occurred in both bars was determined in the usual way. At betatron energies of 4.9, 5.5, and 8.0 Mev, the ratios of the number of fissions in the outer bar to the number in the primary target were $<0.002$, 0.0026, and 0.0060, respectively.

This experiment was repeated with the outer slug wrapped in 0.015-in. thick cadmium foil to shield out thermal neutrons. The number of fissions in the outer bar to the number in the primary bar was found to be 0.0060 at 8.0 Mev. These experiments confirm that less than 1% of the observed fissions in the primary target were induced by thermal neutrons or by fast neutrons that were produced in the betatron structure exclusive of the target itself.

The number of neutrons produced per roentgen of x-rays per mole of element was measured for deuterium and for beryllium as a function of betatron energy up to 10 Mev. The yield of neutrons for these two elements was found to increase approximately as $E_0$, the betatron energy. The number of fissions per roentgen per mole of element for both uranium and thorium increases exponentially with $E_0$ between 4 and 6 Mev. This is taken as evidence that the neutrons generated in the betatron structure have a negligible effect on the experimental results.

An estimate of the number of fissions produced in the target by fast neutrons which are generated in the target itself by the ($\gamma$,fission) and ($\gamma$,$n$) reactions can be made from the geometry and the known neutron cross sections. It is necessary to make the following reasonable assumptions: (1) The neutron spectrum in the target is identical with that of fission neutrons. (2) Average neutron fission cross sections for fission spectrum neutrons in natural uranium and thorium are 0.54 and 0.13 barns, respectively.[20] (3) The average neutron path in the metal slug is 0.5 cm. (4) Recent experimental work[21] giving the total number of neutrons per uranium and thorium photofission is considered applicable. Using these assumptions, we calculate that the percentage of fissions which were due to fast neutrons produced in the targets were as follows: for $U^{238}$ at 6 and 8 Mev, a maximum of 5% and 8%, respectively; for $Th^{232}$ at 6 and 8 Mev, a maximum of 1% and 2%, respectively.

## III. EXPERIMENTAL RESULTS

The activation functions obtained from these experiments are shown in Figs. 1 to 3. In these graphs, the

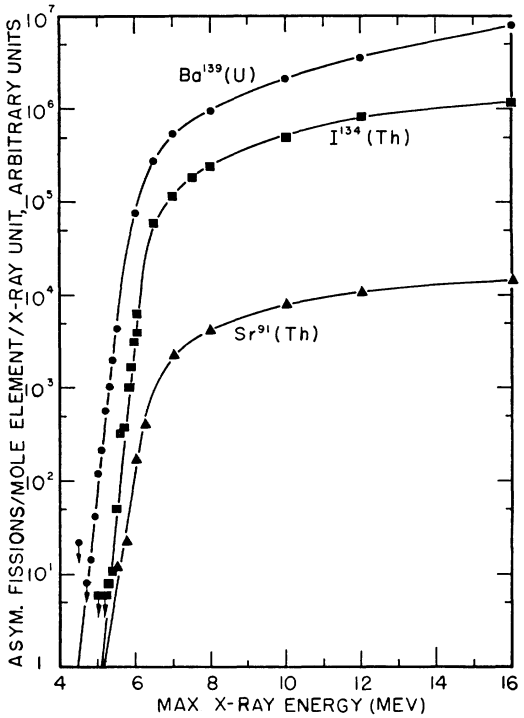


FIG. 1. Semilogarithmic plot of the asymmetric photofissions in natural uranium and natural thorium as a function of increasing betatron energy. ●, $Ba^{139}$ isolated from uranium probe target; ▲, $Sr^{91}$ and ■, $I^{134}$ isolated from thorium probe target. Arrows indicate upper limits. Ordinates for $Ba^{139}$, $Sr^{91}$, and $I^{134}$ are unrelated. See footnote 22 for absolute values of the fission yields, obtained from thin-target bremsstrahlung spectrum.

[19] D. J. Hughes and J. A. Harvey, *Neutron Cross Sections*, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).

[20] A. Turkevich and J. B. Niday, Phys. Rev. **84**, 52 (1951); Keller, Steinberg, and Glendenin, Phys. Rev. **94**, 969 (1954).

[21] Lazareva, Gavrilov, Valuev, Zatsepina, and Stavinsky, *Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, July 1–5, 1955, Session of the Division of Physical and Mathematical Sciences* (Akademia Nauk, S.S.S.R., Moscow, 1955) [translation by Consultants Bureau, New York, 1955], p. 217.

number of photofissions per mole of target element per unit of x-ray intensity has been plotted on a logarithmic ordinate *versus* the betatron energy.[22] Data are shown for the asymmetric, very-asymmetric, and symmetric modes of fission. For the $U^{238}$ target, these types of fission are represented by the fission products $Ba^{139}$, $Br^{83,84}$, and $Cd^{117}$, respectively; for $Th^{232}$ target, the corresponding fission products are $Sr^{91}$ and $I^{134}$, $Br^{33,84}$, and $Cd^{117}$, respectively. The over-all errors result from many independent parts of the experiment and are very difficult to determine quantitatively. It is felt that the over-all error is less than 10% for all of the points above 8 Mev but increases below 8 Mev due to the very low intensity; the over-all error may be as much as 50% for the lowest energy points on each curve.

It can be seen from the graphs that the sensitivity of the method is such that the yields of the fission products could be followed through a change by many orders of magnitude. For example, the asymmetric photofission of $U^{238}$ was found to increase by a factor of about $10^4$ as the betatron energy was increased from 4.8 to 6.0 Mev (Fig. 1). This sensitivity made it possible to follow the activation functions down to lower energies than had been possible in previous investigations.[23]

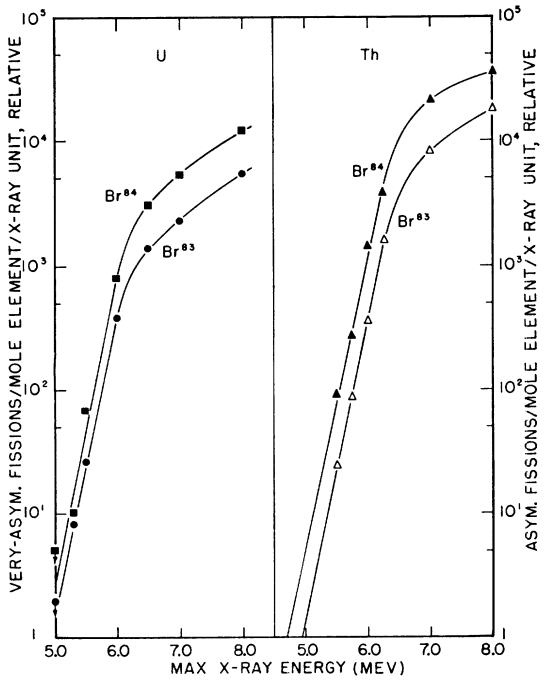


FIG. 2. Semilogarithmic plots of the very-asymmetric and asymmetric photofissions in natural uranium and natural thorium, respectively, as a function of increasing betatron energy. ●, $Br^{83}$ and ■, $Br^{84}$ isolated from uranium probe target; △, $Br^{83}$ and ▲, $Br^{84}$ isolated from thorium probe target. Arrows indicate upper limits. Uranium and thorium ordinates are unrelated.

[22] The absolute numbers of photofissions per mole element per 100 r of x-rays in uranium and thorium and at 8.0-Mev maximum thin target x-ray energy were $0.34\times10^8$ and $0.29\times10^8$, respectively.

[23] In a personal communication, we have learned that L. Katz and co-workers at the University of Saskatchewan have obtained photofission activation functions which resemble those reported here quite closely. The fissions were detected by means of a fission ionization chamber.

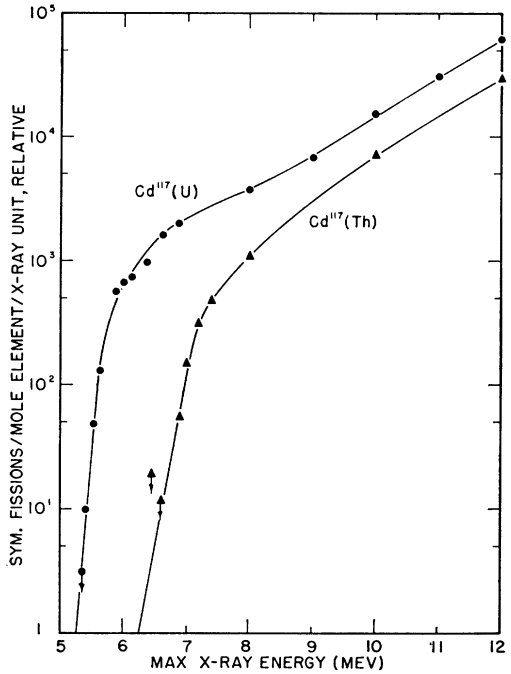


FIG. 3. Semilogarithmic plot of the symmetric photofissions in natural uranium and natural thorium as a function of increasing betatron energy. ●, $Cd^{117}$ isolated from uranium probe target; ▲, $Cd^{117}$ isolated from thorium probe target. Arrows indicate upper limits.

In Fig. 4 the symmetric photofission yields of uranium and thorium have been plotted in order to emphasize the change in probability of symmetric fission as a function of increasing betatron energy from 5.4 to 24 Mev. The ordinate is the logarithm of the percentage of the total number of fissions which produce the fission products $Cd^{115}$ or $Cd^{117}$ and the abscissa is the betatron energy (see previous section for a discussion of normalization of these two yields). The points, represented by crosses, from 12 to 24 Mev are for the photofission of $U^{238}$ and are taken from the paper of Katz *et al.*[8]

From this figure it can be seen that: (1) the probability of symmetric fission in $Th^{232}$ is vanishingly small at low betatron energies; (2) the probability of symmetric fission in $U^{238}$ is higher than in $Th^{232}$ at energies below 10 Mev; and (3) the probability of symmetric fission in $U^{238}$ has a local maximum at 6 Mev. These observations are discussed below.

## IV. DISCUSSION

### A. Observations of the Low-Energy Activation Functions

One of the reasons for initiating the series of experiments reported in this paper was to establish the photofission thresholds of $U^{238}$ and $Th^{232}$ by using as sensitive a detection method as possible. However, the data

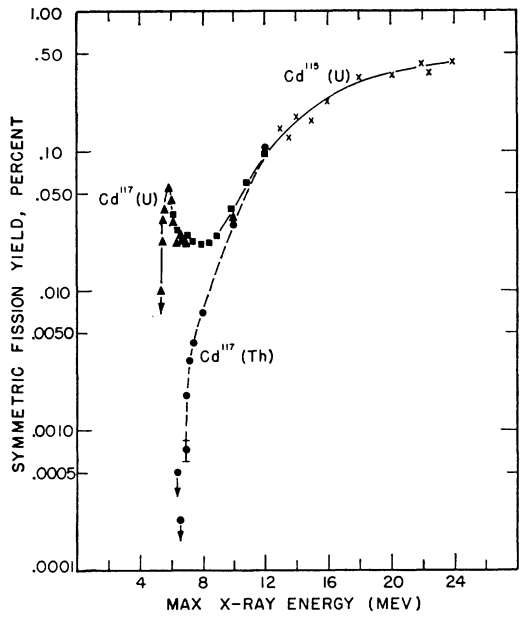


FIG. 4. Symmetric photofission yields of uranium and thorium relative to the 6.0% yields of the asymmetric mode of fission. ×, uranium external samples, $Cd^{115}$ and $Ba^{139}$ were separated by Katz *et al.* (reference 8); ■, uranium external samples, $Cd^{117}$ and $Ba^{139}$ were separated; ▲, uranium probe targets, $Cd^{117}$ and $Ba^{139}$ were separated; and ●, thorium probe targets, $Cd^{117}$ and $Sr^{91}$ were separated. All $Cd^{117}$ yields have a normalization factor which was calculated by assuming that the $Cd^{115}$ yield equals the $Cd^{117}$ yield for the 12-Mev x-ray fission of uranium. Downward directed arrows indicate absence of detectable activity and are the calculated upper limits. The 6.9-Mev thorium value of $0.00073\pm0.00008$ is the mean of two irradiations.

represented in Figs. 1 to 3 seem to show that this is not possible. The photofission cross sections for $U^{238}$ and $Th^{232}$ become very small below 5 Mev but the experiments do not indicate that they become equal to zero. On the contrary, within the lower energy region covered by the present experiments, it appears that the photofission yields continue to decrease exponentially as the maximum x-ray energy is decreased. Consequently, one may anticipate photofission at considerably lower energies than are reported here if a large increase in electron beam intensity were possible. It would seem worthwhile to pursue this question when higher current accelerators become available.

The activation functions shown in Figs. 1 to 2 have similar shapes. There appears to be a low-energy region in which the slope is very steep, then a fairly sharp turnover which is followed by a long region in which the yield increases very slowly with betatron energy. The yield appears to increase approximately by a factor of 10 in an interval of 0.3–0.4 Mev in the low-energy region and by a factor of 10 in an interval of 10 Mev in the high-energy region. The break comes at approximately the $(\gamma,n)$ threshold in each case, i.e., 6.0 Mev[24] for $U^{238}$ and 6.4 Mev[25] for $Th^{232}$. One possible explanation for the occurrence of the breaks is that they are produced by the onset of competition between photofission and the $(\gamma,n)$ reaction. It has been established by previous work[16,21,26] that the absorption of a photon of energy between 8 and 11 Mev in $U^{238}$ is followed by the $(\gamma,n)$ reaction or photofission in the ratio of approximately 4 to 1 in favor of the former. In $Th^{232}$, the ratio appears to be 5 to 1. Consequently, if the total photon absorption cross section varies smoothly with energy, it is to be expected that the onset of the $(\gamma,n)$ reaction should produce the observed effect.

Because of the thick-target bremsstrahlung spectrum used in these experiments at the lowest energies, it is very difficult to extract accurate cross sections from the activation functions. It would be desirable to repeat these observations with a thin-target spectrum from a high-intensity machine. We have attempted to calculate rough photofission cross sections at the low energies; the results are given in the following section.

The shapes of the activation functions for symmetric fission are less well established experimentally and appear to be more complicated (Fig. 3). For $Th^{232}$, the break appears to be more ambiguous than for $U^{238}$. From Fig. 4 it is apparent that for both $Th^{232}$ and $U^{238}$ the symmetric yields fall more sharply at very low energies than do the asymmetric yields.

Since these experiments do not establish a true threshold for the photofission process, they obviously do not give a difference between the thresholds for identical photofission modes in $U^{238}$ and $Th^{232}$ or for asymmetric *versus* symmetric photofission of either nucleus. Hill and Wheeler[4] have suggested that penetration of the fission potential energy barrier should be observed and that the probability, $P$, of barrier penetration should be given by the following expression:

$$P=1/[1+\exp(2\pi b)].$$

The variable $b$ is the energy deficit relative to the top of the barrier, divided by a characteristic quantum energy, $E_{\text{curv}}$, which is fixed by the curvature of the top of the barrier and by the effective mass associated with the fission mode of deformation. The photofission yields of $U^{238}$ and $Th^{232}$ increase exponentially with betatron energy over an energy interval of a few hundred kev at the lowest energies reached, as shown in Fig. 1. This is the behavior predicted by the above formula of Hill and Wheeler if the fissions observed at the lowest energies do occur by penetration of the fission potential energy barrier. According to this formulation, raising the betatron energy up to and beyond the barrier is expected to produce only a gentle break in the excitation function as the barrier is surmounted. The turnover of the activation functions in Fig. 1 may be

[24] Huizenga, Magnusson, Fields, Studier, and Duffield, Phys. Rev. **82**, 561 (1951).

[25] Magnusson, Huizenga, Fields, Studier, and Duffield, Phys. Rev. **84**, 166 (1951).

[26] R. B. Duffield and J. R. Huizenga, Phys. Rev. **89**, 1042 (1953).

partially due to this effect as well as competition from the $(\gamma,n)$ reaction.

Several points can be made about the probability of symmetric fission as a function of betatron energy as shown in Fig. 4.

(1) The percentage of symmetric fissions in $Th^{232}$ is very small at excitation energies below 7 Mev. The yield of $Cd^{117}$ at a betatron energy of 6.5 Mev is less than 0.0003%. This is considerably lower than the yield, 0.01%, from thermal-neutron-induced fission of $U^{235}$. It is also lower than the upper limit of 0.01% from the spontaneous fission[27] of $Cm^{242}$.

(2) The probability of symmetric fission in $U^{238}$ at betatron energies from 5.4 to 8 Mev is considerably higher than in $Th^{232}$. This observation may be correlated with the experiments of Winhold and Halpern[9,28] on the angular distribution of the fragments from photofission of $U^{238}$ and $Th^{232}$. The angular distribution was found to be of the form $a+b\sin^2\theta$, and the value of $b/a$ was larger for $Th^{232}$ than for $U^{238}$. They also found that the anisotropy in $Th^{232}$ and $U^{238}$ appears for photons whose energies are within a few Mev of the photofission thresholds. Moreover, the magnitudes of the ratio, $b/a$, for the asymmetric and symmetric modes in $Th^{232}$ at 16-Mev maximum electron accelerator energy were 0.4 and 0.0, respectively. The present experiments reinforce the observation that mass asymmetry and angular anisotropy are related phenomena.

(3) The peak in the probability for symmetric fission of $U^{238}$ at 6 Mev (Fig. 4) has not been predicted by any fission model. The possibility that it might be due to a spurious effect such as neutron-induced fission was realized and eliminated by the experiments reported above (see IID). The number of neutron-produced fissions is a few percent of the total and, therefore, cannot produce the effect. In addition, if the peak in the symmetric fission probability in $U^{238}$ were due to fast neutrons, it is expected that it would appear also in $Th^{232}$, since the fast-neutron fission cross sections are comparable.[19]

It may or may not be significant that the peak occurs approximately at the $(\gamma,n)$ threshold in $U^{238}$. Possibly the general tendency toward a higher probability for symmetric fission as the excitation energy increases is interrupted above 6 Mev by competition from the $(\gamma,n)$ reaction. This competition may occur more strongly with symmetric fission for reasons which are not now clear.

A model for the fission process has been proposed by Aage Bohr.[29] This model has been very successful in explaining the angular distribution of fission fragments from photofission and from neutron-induced fission. It proposes that the fissioning nucleus, passing over the potential barrier, has rotational levels which resemble quite closely those of the undistorted nucleus near its ground state. For an even-even nucleus, these levels are $0+$, $2+$, $4+$, etc., and also odd-parity states of $1-$, $3-$, etc. All levels are assumed to favor asymmetric fission from energy considerations, but because of the odd parity, the $1-$ and $3-$ levels lead only to asymmetric fission. Pure $E1$ photon absorption at low excitation energy in an even-even nucleus would then lead only to asymmetric fission. This presumably is the situation in $Th^{232}$. In order to explain the increase in the probability of symmetric fission in $U^{238}$ from 8 Mev down to 6 Mev as shown in Fig. 4, it is necessary to assume that $E2$ photon absorption becomes relatively more important. But it then becomes difficult to explain the observed decreasing probability of symmetric fission below 6 Mev.

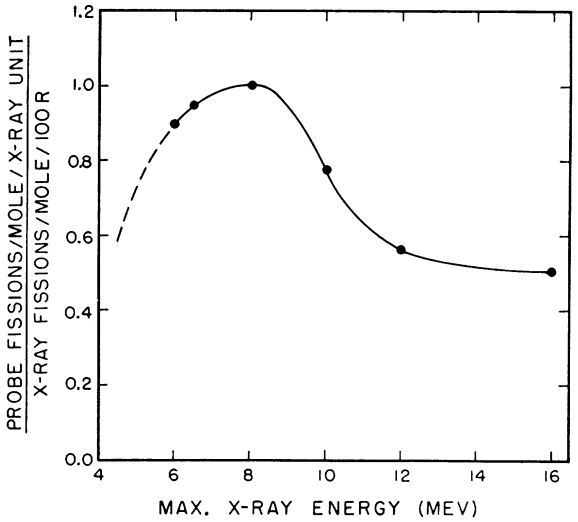


FIG. 5. Ratio of the total photofissions (internal probe target) per mole element per x-ray intensity to the total photofissions (external target) per mole element per 100 r as a function of betatron energy. ●, $Ba^{139}$ isolated from uranium. Ratios are normalized to unity at 8.0 Mev.

## B. Possible Existence of a Bump in Photofission Cross Sections at Low Excitation Energy

It has been stated earlier in this paper that the use of the thick-target bremsstrahlung spectrum for these experiments precludes the extraction of accurate cross sections from the observed activation functions because the shape of the spectrum is not known. We have attempted to use a crude approximation method in order to arrive at the general shape of the cross-section curves. The number of photofissions produced per mole of target per unit x-ray intensity was measured for betatron energies from 6 Mev to 16 Mev using both the

[27] E. P. Steinberg and L. E. Glendenin, Phys. Rev. **95**, 431 (1954).

[28] E. J. Winhold and I. Halpern, Phys. Rev. **103**, 990 (1956).

[29] Aage Bohr, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, August 8–20, 1955* (United Nations, New York, 1956), Vol. 2, *Physics: Research Reactors*, p. 151. We are grateful to Professor A. Bohr for correspondence on this subject and discussion of the present work.

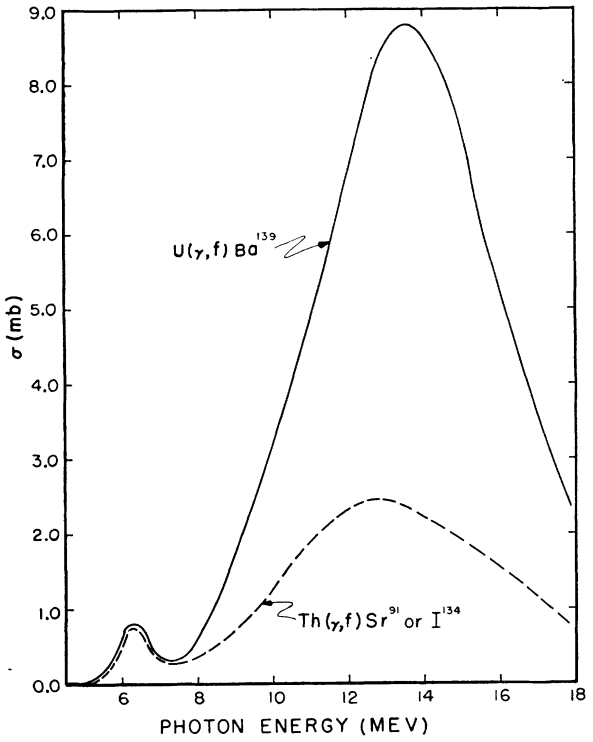


FIG. 6. Asymmetric photofission cross sections, expressed in millibarns, of natural uranium and natural thorium. $Ba^{139}$ was isolated in uranium fission and $Sr^{91}$ and $I^{134}$ were isolated in thorium fission. The relative low-energy cross sections, obtained in this work, were normalized to the absolute photofission cross sections (reference 16) at 10 Mev.

thin-target bremsstrahlung spectrum and thick-target spectrum of the probe target. The ratio of these two numbers is shown in Fig. 5 as a function of betatron energy. The measurements could not be made below 6 Mev with the thin-target spectrum. The ratio has been extrapolated along the smooth broken curve shown in the figure to 4.5 Mev and applied as a correction factor to the data of Fig. 1 in order to obtain a thin-target activation function. This thin-target activation function was treated by the photon difference method[30] in order to calculate the corresponding cross-section curve for asymmetric fission, shown in Fig. 6. Since the percentage yield of mass 139 in the photofission of uranium is known,[7,8] it was possible to obtain absolute values for the cross section by normalizing this curve to that of reference 16 at 10 Mev. The asymmetric photofission cross section for $Th^{232}$ was found in a similar manner. These cross-section curves show a local maximum at approximately 6 Mev, and then a rise to the maximum of the giant resonance. These curves are very similar in form to those found for the photofission of $U^{238}$ and $Th^{232}$ by Winhold and Halpern.[28] The shapes of the asymmetric cross-section curves in uranium and thorium are clearly due to the sharp change in slope of the activation functions near 6-Mev betatron energy. The exact shapes of the cross-section curves obtained here may not be correct because of the crude spectrum correction procedure that was used. However, it seems certain that the bumps at 6 Mev are really present in the curves because they can be removed only by making very drastic changes in the shapes of the experimental yield curves.

Cross-section curves for the fission products $Cd^{117}$ and $Br^{83,84}$ from $U^{238}$ and $Br^{83,84}$ from $Th^{232}$ have been calculated by a similar approximation procedure. Each of these shows a similar bump in the cross section near 6 Mev. The symmetric ($Cd^{117}$) photofission data of $Th^{232}$ are not available over a wide enough energy range and the existence of a maximum in the cross section at low energies cannot be established.

## ACKNOWLEDGMENTS

We wish to acknowledge the cooperation of Mr. T. J. Keegan and the 24-Mev Betatron Staff. Also, we are indebted to Dr. L. E. Glendenin and Dr. E. P. Steinberg of the Argonne National Laboratory for helpful discussions.

[30] L. Katz and A. G. W. Cameron, Can. J. Phys. **29**, 518 (1951). The supplement from 3 to 8 Mev for the original photon difference tables was kindly sent to us by L. Katz.