Angular Distribution of Fragments from Neutron-Induced Fission*†

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(Received January 18, 1954)

A parallel plate ionization chamber, in which one electrode served as a collimator, was employed to study the angular distribution of fission fragments referred to the neutron beam axis. Fragments from neutron bombardment of the following nuclei were examined: Th^{232}, U^{233}, U^{235}, U^{238}, and Np^{237}. When 14-Mev neutrons were used, a preponderance of fission along the neutron axis as compared with the orthogonal direction was noted. The average value obtained for the 0°/90° ratios was 1.26. Thermal neutron fission was found to be isotropic.

INTRODUCTION

THE Bohr-Wheeler liquid drop model[1] for the fission process would predict essentially no correlation between the direction of the incoming fast neutron and the direction of the fission fragments, apart from center-of-mass effects. According to this model, the energy of the neutron is quickly assimilated among the individual nucleons of the fissile nucleus and only later is this energy concentrated on a mode of deformation leading to fission. However, according to the recently developed collective model of the nucleus[2,3] as applied to the fission process, energy from the capture of a fast neutron may be divided promptly between nucleonic excitation and the vibrational excitation of the nuclear surface. When the beam neutrons have an energy of several Mev, the vibrational excitation will be predominantly such as to distort the nucleus along the direction of the neutron beam, leading preferentially to fission in this direction. Also, on the basis of the collective model, the presence of a nuclear quadrupole moment might be expected to influence the angular distribution.

We have measured the angular distribution of the fragments from the 14-Mev neutron-induced fission of Th^{232}, U^{233}, U^{235}, U^{238}, and Np^{237} to provide a test between these two pictures of the fission process. We have observed also the angular distribution of fragments from the thermal neutron-induced fission of U^{233} and U^{235}, principally to provide a check of our apparatus since no anisotropy would then be expected.

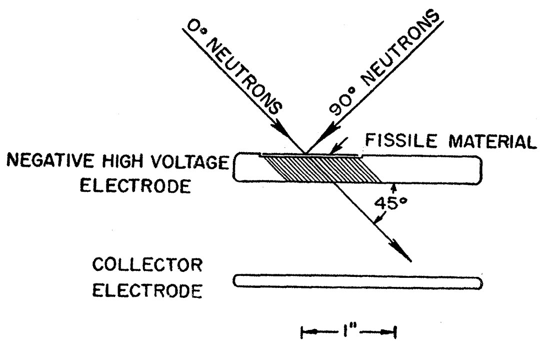

FIG. 1. Schematic representation of the fission chamber.

APPARATUS AND PROCEDURE

The source of fast neutrons was the $T(d,n)He^4$ reaction. An atomic beam of 250-kev deuterons from the Los Alamos Cockcroft-Walton accelerator is magnetically analyzed, collimated through a $\frac{1}{4}$-in. diaphragm and intercepted by a thick zirconium-tritium target. The tritium is absorbed in a zirconium film which is deposited on a thin tungsten disk.[4] The direction of observation was at an angle of 74° with respect to the deuteron beam. In this direction the neutrons have an energy of 14.3±0.1 Mev, assuming a reaction energy of 17.6 Mev for the $T(d,n)He^4$ reaction. A collimated beam of thermal neutrons was obtained from the Los Alamos homogeneous reactor.

TABLE I. Approximate analyses of foils.

Principal isotope	Percentage abundance
Th^{232}	100
U^{233}	96 (balance normal U)
U^{235}	96 ($\frac{1}{2}$% U^{234}, balance U^{238})
U^{238}	100 (16 parts/million U^{235})
Np^{237}	100

A cross section of the fission chamber is shown schematically in Fig. 1. The negative high-voltage electrode served as a collimator for the fission fragments; 0.04-in. holes were drilled on an hexagonal matrix to fill a circle 1 in. in diameter. These passages were inclined 45° with respect to the normal in order that the geometry would be in all essentials identical for the 0° and 90° angular settings. The extreme angular resolution of the collimator was ±6° and the average angle of emission of the fragments with the axis of collimation was $2\frac{1}{2}$°. The foils of fissile material were 1 in. in diameter and at a distance of 6.25 in. from

* This document is based on work performed under the auspices of the U. S. Atomic Energy Commission.
† A preliminary report of these findings was presented at the meeting of the American Physical Society, New York, January, 1953 [Phys. Rev. 90, 388 (1953)].
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[4] Graves, Rodriguez, Goldblatt, and Meyer, Rev. Sci. Instr. **20**, 579 (1949).

the Zr—T target. Since the largest projected dimension of the target seen by the foils was $\frac{5}{16}$ in., the extreme angle that a neutron could make with the target-foil axis was 6° and the average angle with this axis was $3\frac{1}{2}$°. No corrections to the final data due to angular spread were necessary.

A gas filling of 95 percent argon and 5 percent CO_2 was used at a pressure of 45 cm Hg in order that the fastest fission fragments would be stopped shortly before reaching the collector. A field of 118 volts per cm was applied between plates, no attempt being made to attain saturation. The rise time of the electron collection pulses was about 0.5 μsec. The chamber was mounted so that it could be rotated by means of an indexed rotary table about an axis containing a diameter of the fissile layer. Angular error of position probably did not exceed 0.5°. For the fast neutron studies with U^{233} and U^{235} a cadmium jacket surrounded the chamber.

Thin layers of ThO_2, U_3O_8, and NpO_2 were prepared by painting solutions of the nitrates on 0.01-in. Pt disks and baking at 800°C until the oxides were formed.[5] Deposits ranging from 100 μg/cm² to 5 mg/cm² were used. Table I gives approximate analyses of the foils.

TABLE II. 0°/90° fission intensity ratios (center-of-mass system) for 14-Mev neutrons.

Isotope	Type	0°/90° ratio
U^{233}	Even-odd	1.32±0.05
U^{235}	Even-odd	1.27±0.08
U^{238}	Even-even	1.31±0.05
Np^{237}	Odd-even	1.15±0.04

A preamplifier with cathode follower output was attached to the back of the fission chamber so as to form a shielded unit. Fission pulses from a linear pulse amplifier with rise time of 0.5 μsec and *RC* clipping time of 2 μsec were fed to two scalers. The discriminator setting of one scaler was such as to accept all fission pulses while that of the second scaler was such as to accept principally only pulses due to the higher energy (light) fragments. In this way it was hoped that angular distributions could also be obtained for the light fragments alone. However, later work with an 18 channel pulse-height analyzer indicated that the two fission ionization peaks were not sufficiently separated to make this feasible.

For each foil an integral bias curve was first taken to determine discriminator settings.[6] About 500 counts were then taken at each angular setting and this was repeated until adequate counts were accumulated.

The integrated fast neutron flux was monitored by counting a known fraction of the alpha particles generated in the $d+T$ reaction by means of a proportional counter mounted 22 in. from the target in a geometry permitting a view of the entire target area. Also a long counter directly monitored the neutron flux. Since in some instances the ratio of the counts in these two counters fluctuated by as much as 2 percent, the alpha counter was always used as the standard monitor. The thermal neutron flux from the homogeneous reactor was maintained at a constant value within 0.5 percent throughout the measurements.

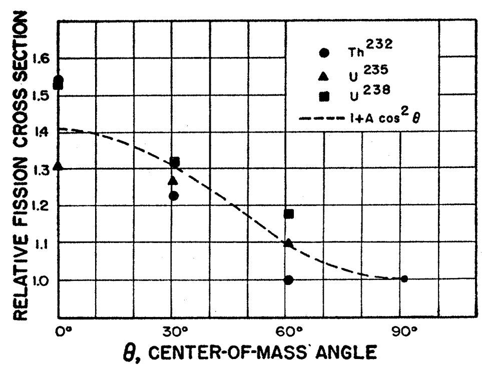

FIG. 2. Angular distribution of fragments from the 14-Mev fission of Th^{232}, U^{235}, and U^{238}.

RESULTS

The 0°/90° fission intensity ratios for thermal neutrons on U^{233} and U^{235} were determined to be 1.00±0.04 and 0.99±0.05, respectively.[7] The intensity measured at 45° was no different within statistical error from that at 0°.

Table II gives the 0°/90° fission intensity ratios as measured for 14-Mev neutrons. In all cases foils of 1 mg/cm² or thinner were used.[8] Angular distributions with points at 0°, 30°, 60°, and 90° were taken for Th^{232}, U^{235}, and U^{238} using 5-mg/cm² foils. The experimental points are plotted in Fig. 2. Although these measurements are of a more qualitative nature it is seen from Fig. 2 that a curve of the form $1+A\cos^2\theta$ satisfactorily fits the points, where θ is the angle between the fragment direction and the neutron beam.

Winhold, Demos, and Halpern[9] have measured the angular distribution of fission fragments in the 16-Mev photofission of Th^{232} and found a $(1+A\sin^2\theta)$ dependence corresponding to a higher intensity of frag-

[5] We are indebted to Dr. John Povelites of this laboratory for the preparation of the foils.

[6] Because of the center-of-mass effect for 14-Mev neutrons, a fission fragment emitted at 90° has about $3\frac{1}{2}$ percent less energy than if it were emitted at 0°. The rather large energy loss of the fragments in the collimator of the fission chamber results in a magnification of this effect. Thus, for a 1-mg/cm² foil, the ionization peaks were observed to shift downward about 10 percent going from the 0° to the 90° angular setting. The lower discriminator setting was always such as to count essentially all fragments at both angular settings.

[7] Errors quoted in this paper are standard deviations resulting from statistics only. Systematic errors are thought to be small by comparison.

[8] Anisotropy of about the same amount was observed for Th^{232} but because of the necessity of using a 5 mg/cm² foil to boost the counting rate we do not at present quote a 0°/90° intensity ratio for this isotope.

[9] Winhold, Demos, and Halpern, Phys. Rev. **87**, 1139 (1952).

ments at 90° to the incident photon beam. The experimental results for both photofission and fast neutron fission are at least qualitatively compatible with the collective model picture of the fission process[10] whereas they are in disagreement with predictions to be obtained from the simple liquid drop model.

We express our thanks to Dr. J. H. Coon and the Los Alamos Cockcroft-Walton group for making available to us their accelerator and necessary auxiliary equipment required to monitor the neutron beam. We also thank the Los Alamos homogeneous reactor group for the use of their reactor. We are indebted to Dr. Keith Boyer for suggesting the type of collimator used in these experiments and to Dr. D. L. Hill for several stimulating interpretative discussions on the subject of the collective model.

[10] For discussion refer to reference 3, page 1116.

PHYSICAL REVIEW VOLUME 94, NUMBER 3 MAY 1, 1954

Inner Bremsstrahlung of Cs^{131}†

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(Received January 13, 1954)

The continuous gamma-ray spectrum (inner bremsstrahlung), accompanying orbital electron capture in Cs^{131}, has been observed by the method of scintillation spectrometry. The absolute intensity of the gamma radiation has been measured and compared with the total disintegration rate. The end point of the gamma-ray distribution was found to be 320 ± 10 kev. The experimentally determined shape of the spectrum, as well as the intensity of the continuum, is compared with theoretical calculations. The disagreement exceeds considerably that which might be expected from a consideration of the probable errors of the observations. The growth of the 12-day isomer of Xe^{131} was not detected.

INTRODUCTION

THE continuous gamma radiation (inner bremsstrahlung) associated with the electron capture process was first investigated theoretically by Morrison and Schiff.[1] More recently, Jauch[2] has reviewed and extended the earlier studies. In addition to experimental comparisons of the spectral distributions which have been computed theoretically, this particular disintegration process can also provide information concerning the disintegration energy released in electron capture.

The gamma-ray continuum has been previously detected,[3–7] and the spectral distributions determined for the cases of Fe^{55}, A^{37}, and Ge^{71}. However, it appears that the extent of gamma-ray emission per disintegration has not been measured for any of the above-cited examples. The radioactive chain $Ba^{131}\rightarrow Cs^{131}\rightarrow Xe^{131}$ was first reported by Yu, Gideon, and Kurbatov.[8] The results of several subsequent investigations showed that neither gamma rays nor conversion electrons are emitted.[9–11] Cheng and Kurbatov[12] have reported the growth of the 12-day Xe^{131m} from Cs^{131}, whereas Canada and Mitchell[13] obtained a contrary result. To study the properties of the inner bremsstrahlung and to investigate further the possible formation of Xe^{131m}, the radiations of Cs^{131} have been reinvestigated.

CHEMICAL PROCEDURE

Eighty-eight grams of $Ba(NO_3)_2$ were exposed to neutrons in the Oak Ridge pile for a period of four weeks. From an aqueous solution of this $Ba(NO_3)_2$ with its cesium daughter element, $BaCl_2\cdot H_2O$ was precipitated by ether and HCl, thus removing barium alone. Small portions of inactive $Ba(NO_3)_2$ were repeatedly added and precipitated as $BaCl_2\cdot H_2O$ to extract all of the active barium from the cesium solution. The cesium solution was evaporated to small volume and again scavenged by $BaCl_2\cdot H_2O$ precipitation. Finally, after adding approximately one milligram of cesium carrier—$CsClO_4$ was precipitated.

The first separation of cesium from the neutron irradiated barium disclosed the presence of a 660-kev gamma ray in the cesium fraction which decayed with a half-period of approximately seven days and may, therefore, be associated[14] with Cs^{132}.

† Assisted by the joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.
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