

## Relative Probability of Ternary Fission with Emission of Long-Range Alpha Particles in the Thermal-Neutron Fission of $U^{235}$ †

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The ratio of binary to ternary fission in  $U^{235}$  has been measured using solid-state detectors and aluminum absorbers, for two reactor beams, having cadmium ratios of 30 to 1 and 200 to 1, and for incident neutron energies of 0.003, 0.0253, 0.08, and 0.3 eV. Furthermore, a new method of measuring this ratio without the use of absorbers is presented. The results obtained show that the binary-to-ternary fission ratio in  $U^{235}$  is a constant to a precision of 3% in the energy region between 0.003 and 0.3 eV. The measurements performed with the two reactor beams, having cadmium ratios of 30 to 1 and 200 to 1, gave for this ratio  $518 \pm 13$  and  $512 \pm 14$ , respectively. The measurement performed without an absorber gave a value of  $490 \pm 20$  for the 30-to-1 cadmium-ratio beam. It is concluded that the binary-to-ternary fission ratio in  $U^{235}$  is a constant in the energy region considered to within the precision of the present experiment.

### I. INTRODUCTION

THE relative probability of ternary fission with emission of long-range alpha particles in the thermal neutron fission of  $U^{235}$  has been measured by several investigators.<sup>1-10</sup> These data are not in good agreement and there is a considerable spread in the reported results. It has been suggested<sup>6</sup> that this variation could be due to the fact that the probability for this process varies with the energy of the incident neutron.

The purpose of the present experiment was both to look for a possible variation, with incident neutron energy, of the relative probability of ternary fission with emission of long-range alpha particles, that is, of the binary-to-ternary fission ratio, in the thermal neutron fission of  $U^{235}$ , and to measure this ratio to a higher precision than had hitherto been obtained. The measurements performed fall into three groups:

- Determination of the ratio of binary to ternary fission for two reactor beams having different cadmium ratios.
- Search for a possible variation of this ratio in the thermal region by performing a relative measurement of the binary-to-ternary fission ratio in the neutron energy region between 0.003 and 0.3 eV.
- Measurement of the binary-to-ternary fission ratio

for reactor neutrons by a new method which permits the simultaneous determination of this ratio and of the energy distribution of the long-range alpha particles without having to correct for the energy loss in air, aluminum, or other stopping material.<sup>11</sup>

### II. RATIO OF BINARY TO TERNARY FISSION FOR REACTOR NEUTRONS

The ratio of binary to ternary fission in  $U^{235}$  has been measured using two neutron beams, from the Brookhaven National Laboratory graphite reactor, with cadmium ratios of 30 to 1 and 200 to 1.<sup>12</sup> Care was taken in both sets of measurements to maintain the same geometry.

#### A. Apparatus and Experimental Procedure

The neutron beam was incident on a 1 in.  $\times$  1 in.  $U^{235}$  target containing 0.5 mg/cm<sup>2</sup> of highly enriched<sup>13</sup>  $U^{235}$  deposited<sup>14</sup> on a 0.015-in.-thick aluminum plate. A 300  $\Omega$ -cm resistivity surface-barrier gold-silicon detector was mounted 1 cm from the source and out of the neutron beam. (A 300  $\Omega$ -cm resistivity detector was employed for the purpose of counting ternary alpha particles and not for determining the shape of the energy distribution.) A 0.696-mil aluminum foil could be placed between the counter and the source and 4 mm from the former. The purpose of this foil was to stop all natural alpha particles and fission fragments from reaching the detector, while alpha particles of energy greater than 6.2 MeV were detected by the counter. The pulses from the detector were fed into a low-noise charge-sensitive

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<sup>2</sup> P. Demers, *Phys. Rev.* **70**, 974 (1946).

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<sup>4</sup> Tsien San-Tsiang, Ho Zah-Wei, R. Chastel, and L. Vigneron, *J. Phys. Radium* **8**, 165 and 200 (1947).

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<sup>8</sup> C. B. Fulmer and B. L. Cohen, *Phys. Rev.* **108**, 370 (1957).

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<sup>12</sup> D. J. Hughes, *Pile Neutron Research* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1953), p. 62.

<sup>13</sup>  $U^{234}$  (0.06%),  $U^{235}$  (99.76%),  $U^{238}$  (0.06%), and  $U^{238}$  (0.09%).

<sup>14</sup> B. Cohen and D. E. Hull, Columbia University Report CU-A-1235, 1944 (unpublished).

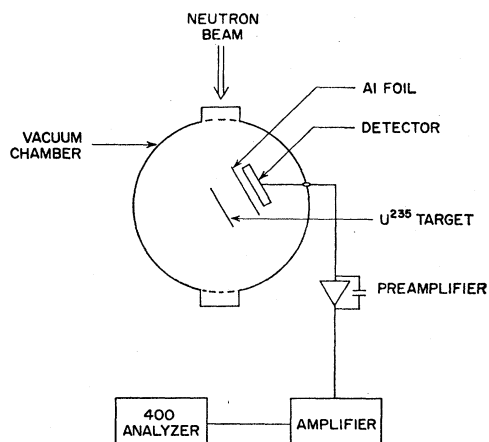


FIG. 1. Experimental arrangement and block diagram of the associated electronics used to measure the binary-to-ternary fission ratio with an aluminum absorber.

preamplifier,<sup>15</sup> amplified, and recorded on a 400-channel pulse-height analyzer. The counter and source arrangement are shown in Fig. 1 together with a block diagram of the associated electronics.

The mode of operation was as follows: With the 0.696-mil aluminum foil removed, the binary fission events were recorded in the analyzer for a preset time. The aluminum foil was then inserted between the counter and the source. Ternary alpha particles were then recorded for a preset time. The aluminum between the counter and the source was then removed and a new cycle started. The cycle was repeated until the desired statistics were obtained.

Before and after a ternary fission run, the natural alpha spectrum from the source was recorded. This measurement served two purposes. It calibrated the energy scale of the system, and checked the stability of the equipment during the ternary fission runs. Once the calibration was obtained and proper corrections made, the ratio of counts, normalized to time, with and without the aluminum foil, between the source and de-

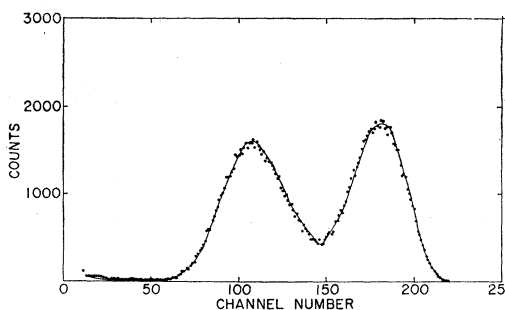


FIG. 2. The fission-fragment distribution obtained from an 0.5-mg/cm<sup>2</sup>-thick U<sup>235</sup> foil.

tor, yielded the ratio of binary to ternary fission for the energy distribution of the neutron beam.

## B. Results

A typical spectrum of a fission distribution obtained is shown in Fig. 2. As can be seen from this figure, the high resolution of the detector for fission fragments permits an accurate determination of the number of fission fragments incident upon the counter. In a binary fission event either of the two heavy fragments may be detected, whereas in ternary fission only the ternary alpha

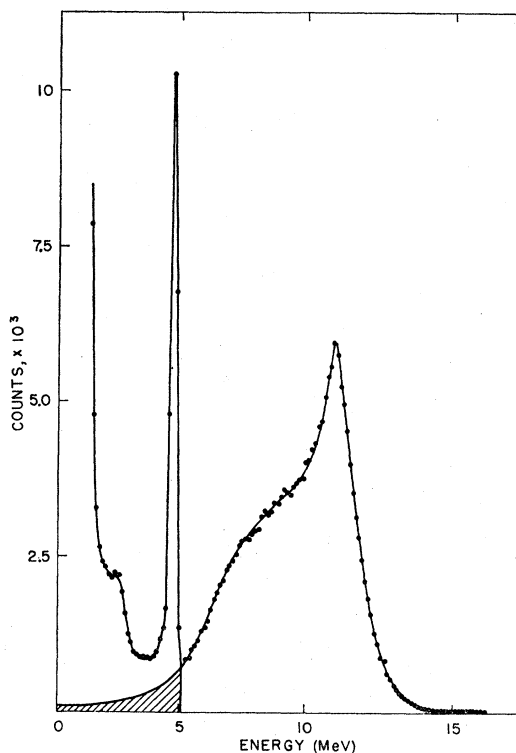


FIG. 3. The alpha particle spectrum from ternary fission using an 0.696-mil aluminum absorber to filter out the fission fragments and the natural alpha particles in a detector whose depletion layer is too thin to stop the most energetic alpha particles.

particle can pass through the absorber foil. Thus, the observed binary fission rate has to be corrected by this factor in order to obtain the desired binary-to-ternary fission ratio.

Figure 3 shows the distribution (with a U<sup>234</sup> calibration line) obtained in a typical ternary alpha run. As can be seen from this figure, this distribution is obscured below 5 MeV by background which arises from the interaction with the aluminum foil and the counter material of both the neutrons and gamma rays associated with the fission process. Furthermore, because of the shallow depletion layer of the counter, alpha particles of energy greater than 11 MeV "pile up" at this energy. The distortion in the distribution due to this effect is

<sup>15</sup> J. Hahn and R. O. Mayer, IRE Trans. Nucl. Sci. NS-9, 20 (1962).

TABLE I. Ratio of binary to ternary fission for two reactor beams of different cadmium ratios.

Cadmium ratio	Binary-to-ternary ratio
30	$518 \pm 13$
200	$512 \pm 14$

unimportant as long as there is a clear boundary between the ternary alpha particles and the background below 5 MeV.

The unresolved tail in the ternary alpha distribution below 5 MeV (shaded area in Fig. 3) accounts for approximately 2% of the total distribution and contributes the main error in the determination of the total number of long-range alpha particles detected.

The ratio of binary to ternary fission obtained from the measurements performed with both reactor beams are shown in Table I. The errors quoted result from both the unresolved low-energy portion of the spectrum and from the statistical error.

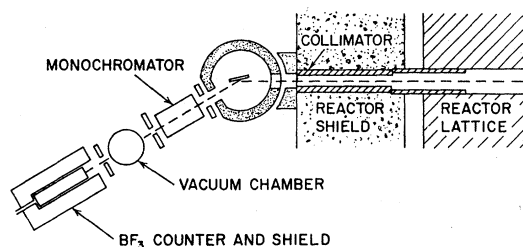


FIG. 4. Schematic diagram of the experimental arrangement used on the Columbia University crystal spectrometer.

### III. RATIO OF BINARY TO TERNARY FISSION IN THE NEUTRON ENERGY REGION BETWEEN 0.003 AND 0.3 eV

The dependence of the probability of emission of long-range alpha particles on the energy of the incident neutrons in the thermal region was investigated by making a relative measurement of the binary-to-ternary fission ratio for incident neutron energies of 0.003, 0.0253, 0.08, and 0.3 eV, respectively.

#### A. Apparatus and Experimental Procedure

The Columbia University crystal spectrometer<sup>16</sup> at the Brookhaven National Laboratory graphite reactor was used to measure the binary-to-ternary fission ratio at incident neutron energies of 0.0253, 0.08, and 0.3 eV. These measurements were performed with the same detector and source arrangement used in the measurements of Sec. II which were placed on the spectrometer arm. The counting procedure employed at the selected neutron energies was the same except that a  $\text{BF}_3$  counter was used, in addition, to continuously monitor the neu-

<sup>16</sup> F. T. Gould, T. I. Taylor, W. W. Havens, Jr., B. M. Rustad, and E. Melkonian, Nucl. Sci. Eng. 8, 453 (1960).

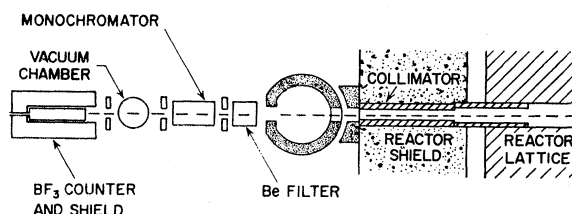


FIG. 5. Schematic diagram of the experimental arrangement used to obtain the ratio of binary to ternary fission at 0.003 eV.

tron beam so that corrections could be made for reactor fluctuations. A mechanical monochromator<sup>17,18</sup> was employed to eliminate order contamination in the neutron beam (Fig. 4).

The measurement of the binary-to-ternary fission ratio at 0.003 eV was also performed on the spectrometer arm so as to preserve the same beam geometry (slit arrangement), except that the direct beam was used in conjunction with a beryllium filter<sup>19</sup> at liquid-nitrogen temperature and a mechanical monochromator<sup>17,18</sup> to select the desired neutron energy. This experimental arrangement is shown in Fig. 5. The measurement was performed with the same detector and source arrangement as on the spectrometer runs, and the same counting procedure was employed.

#### B. Results

The fission and long-range alpha particle distributions obtained were similar to those shown in Figs. 2 and 3, respectively. The relative values of the binary-to-ternary fission ratio were obtained by taking, at each neutron energy, the ratio of the total number of fission events to the number of counts above 7 MeV in the corresponding ternary alpha spectrum. This restriction

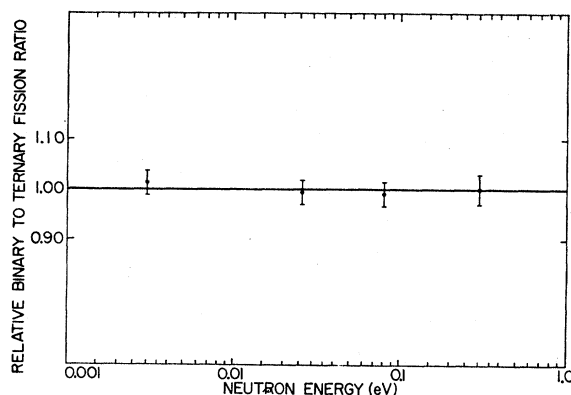


FIG. 6. Deviation of the binary-to-ternary fission ratio from its mean value in the neutron energy region between 0.003 and 0.3 eV.

<sup>17</sup> N. Holt, Rev. Sci. Instr. 28, 1 (1957).

<sup>18</sup> J. A. Moore, J. J. Rush, and B. M. Rustad, Bull. Am. Phys. Soc. 4, 245 (1959).

<sup>19</sup> B. M. Rustad, E. Melkonian, W. W. Havens, Jr., T. I. Taylor, F. T. Gould, and J. A. Moore (to be published).

on the ternary spectrum was imposed so that no correction would be necessary for the background present below 5 MeV. The only corrections made were, therefore, for the small fluctuations in the reactor beam intensity.

The results obtained are shown in Fig. 6. This figure illustrates the deviation of the binary-to-ternary fission ratio from its mean value in the neutron energy region between 0.003 and 0.3 eV. The errors indicated result entirely from the statistical error.

#### IV. MEASUREMENT OF THE RATIO OF BINARY TO TERNARY FISSION FOR REACTOR NEUTRONS USING AN ANTICOINCIDENCE METHOD

The excellent geometry provided by solid-state detectors has led to a new method of measuring the binary-to-ternary fission ratio.

##### A. Apparatus and Experimental Procedure

A thin VVNS foil of effective thickness  $8 \mu\text{g}/\text{cm}^2$  coated by evaporation with  $^{13}80 \mu\text{g}/\text{cm}^2$   $\text{U}^{235}$  was mounted between two surface-barrier gold-silicon detectors in a vacuum chamber. A neutron beam from the Brookhaven National Laboratory graphite reactor, with a cadmium ratio of 30 to 1, was incident upon the foil. This counter and foil arrangement, together with a block diagram of the associated electronics, are shown in Fig. 7. Of the two detectors used, one (the ternary alpha detector) had a resistivity of  $3000 \Omega\text{-cm}$  and was capable of detecting alpha particles of energy greater than 30 MeV; the other (the fission detector) had a  $300 \Omega\text{-cm}$  resistivity. These two detectors were so placed with respect to the foil that all fission fragments inci-

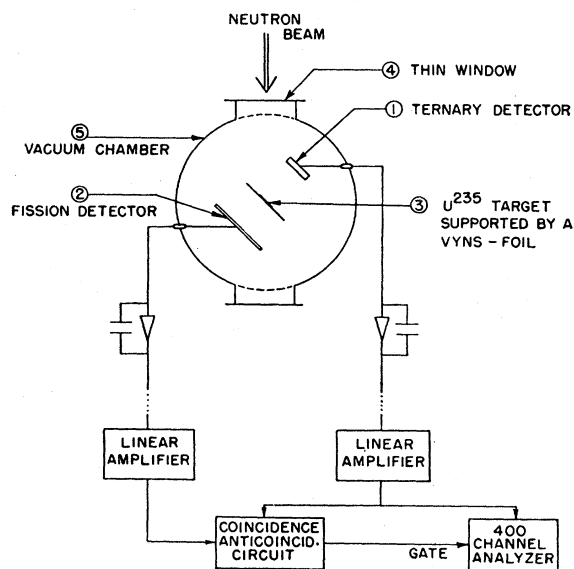


FIG. 7. Experimental arrangement and block diagram of the associated electronics used in the coincidence-anticoincidence method.

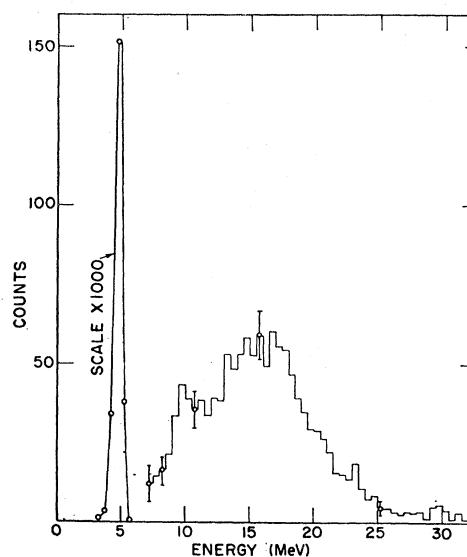


FIG. 8. Distribution of ternary alpha particles obtained with the anticoincidence technique.

dent upon the alpha detector had their corresponding fragments incident upon the fission detector, i.e., a shadow detector. Furthermore, since long-range alpha particles are emitted within the range of angles  $90^\circ \pm 60^\circ$  with respect to the line of emission of the fission fragments,<sup>7</sup> the two detectors were arranged so that for any long-range alpha particle incident on the ternary alpha detector the associated fission fragments would fall outside both detectors. The pulses from the alpha detector were recorded on a 400-channel pulse-height analyzer. A gate from a coincidence-anticoincidence circuit, which demanded coincidence or anticoincidence between the alpha and fission counters, triggered the analyzer.

The mode of operation was as follows: To prevent the recording of the fission fragments in the ternary alpha detector an anticoincidence condition was demanded between the alpha detector and the fission detector, while, to prevent the recording of the long-range alpha particles in the ternary alpha detector, a coincidence condition was demanded between the alpha detector and the fission detector. The ratio of binary to ternary fission was then determined from measurements of the pulse spectrum of the ternary alpha detector with a coincidence and with an anticoincidence requirement.

##### B. Results

The measurements performed with the anticoincidence arrangement described above is shown in Fig. 8. As can be seen from this figure, the ternary alpha distribution is limited below 6 MeV by the natural alpha particles from the target. This arises from the fact that the natural alpha particles from the source fulfil the anticoincidence condition. To minimize this effect, highly enriched  $\text{U}^{235}$  was used. A typical fission distri-

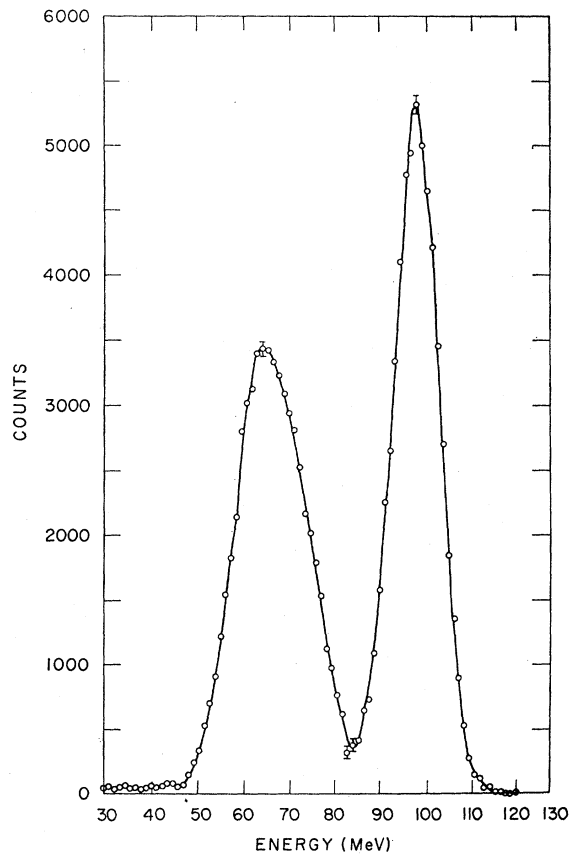


FIG. 9. Fission-fragment distribution obtained with the coincidence technique.

bution obtained with the coincidence requirement is shown in Fig. 9.

The binary-to-ternary fission ratio obtained with this method is  $490 \pm 20$ . The error quoted results from that part of the spectrum hidden by the natural alphas and from the statistical error.

#### V. DISCUSSION OF RESULTS

The results of the present experiment show no variation in the binary-to-ternary fission ratio in  $U^{235}$  for the two neutron beams used, the energies considered, or for the two different methods employed in obtaining the data. These values are in good agreement with the latest value for thermal neutrons obtained by Nobles.<sup>10</sup> Furthermore, the measurements performed at the four neutron energies confirm the results of Auclair<sup>20</sup> and Genin *et al.*<sup>21</sup> for this same region.

It is therefore concluded that, for the energy range considered, the ratio of binary to ternary fission in  $U^{235}$  is a constant within the precision of the present experiment. The considerable spread in the reported results for this ratio in this energy region cannot be explained by assuming that it varies with the energy of the incident neutrons.

#### ACKNOWLEDGMENTS

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<sup>20</sup> J. M. Auclair, *J. Phys. Radium* **19**, 68 (1958).

<sup>21</sup> R. Genin, R. Joly, and M. Ribrag, *J. Phys. Radium* **21**, 473 (1960).