

for the isodiaspheric system  $^{135}\text{I}$ - $^{135}\text{Xe}$ . In that system the strongest  $\beta$  decay is observed to five levels near 2.3 MeV.

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## Subthreshold Neutron-Induced Fission in $^{238}\text{U}^\dagger$

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In an attempt to observe subthreshold fission in  $^{238}\text{U}$ , a sample with very low  $^{235}\text{U}$  content was exposed to the intense neutron beam of the Physics-8 nuclear explosion. A high-sensitivity oscilloscope setting allowed individual fission-fragment pulses from a semiconductor detector to be counted. In the region from 37 to 327 eV, the average fission width for eight resonances was determined to be  $(0.03 \pm 0.05) \times 10^{-6}$  eV. In the keV region, the average fission cross section was measured to be  $61 \pm 24 \mu\text{b}$  from  $10^4$  to  $3 \times 10^4$  eV and  $36 \pm 14 \mu\text{b}$  from  $3 \times 10^4$  to  $10^5$  eV.

### I. INTRODUCTION

In exploring the systematics of neutron-induced fission and in characterizing the double-peaked

fission barrier now believed to prevail in the U-Pu region, it is of interest to examine the fission behavior of as many diverse isotopes as possible. Although  $^{238}\text{U}$  is readily available, its cross sec-

tion for neutron-induced fission below the fission threshold at about 1.5-MeV neutron energy is so small that knowledge of the subthreshold behavior is sparse. Measurements are made more difficult by the large fission cross section of the  $^{235}\text{U}$  which is present in  $^{238}\text{U}$  samples. Leonard and Odegaard<sup>1</sup> were able to set an upper limit of  $0.2 \times 10^{-6}$  eV for the fission width of the lowest-energy neutron resonance at 6.7 eV. The thermal-neutron fission cross section is known to be small.

One of the experiments included in the Physics-8 nuclear-explosion measurements was an attempt to observe subthreshold fission in  $^{238}\text{U}$ . The experimental details of these single-pulse neutron time-of-flight experiments have been described previously.<sup>2</sup> In the present measurement, a sample of  $^{238}\text{U}$  was exposed to the intense neutron beam at a flight path of 244 m. The sample was  $0.9 \text{ mg/cm}^2$   $^{238}\text{U}$  vacuum evaporated on a  $0.00036\text{-cm}$  stainless-steel backing with a total of 3.8 mg of  $^{238}\text{U}$  in the neutron beam. The source material was procured from the Oak Ridge National Laboratory from batch No. U-238 Q-505, stated to be 99.9999%  $^{238}\text{U}$  with 0.00002%  $^{234}\text{U}$  and less than 0.00005%  $^{235}\text{U}$  (0.5 ppm). The sample was viewed by two semiconductor detectors outside the neutron beam. Detector signals were amplified logarithmically and recorded photographically by a combination of moving-film cameras and oscilloscopes.<sup>2</sup> In the case of  $^{238}\text{U}$ , one signal was recorded with an oscilloscope sensitivity five times that normally used, so it was possible to distinguish clearly on the film record individual fission-fragment pulses from the detector.

The experiment reported here establishes a rather firm upper limit of  $0.13 \times 10^{-6}$  eV (2 standard deviations above the most probable value) for the average fission width of eight resonances between 37 and 237 eV, and yields an average fission cross section of  $40 \pm 13 \mu\text{b}$  between  $10^4$  and  $10^5$  eV.

## II. RESONANCE REGION

Many smooth resonances were observed in  $^{238}\text{U}$ , corresponding to known<sup>3</sup> resonance energies, because the fission-fragment detectors also responded to radiative capture in the sample through the conversion electrons produced by the  $\gamma$ -ray cascade. For each capture event in the sample about 1/1000 as much charge was generated in the detectors as for each fission event. The smooth resonances then corresponded to the detection of many conversion electrons, with correspondingly excellent statistical accuracy. In addition to the smooth resonances arising from capture, several dozen fission-fragment pulses were observed scattered throughout the resonance region. The fragment pulses were distinct spikes with a height (at the 50- $\Omega$  amplifier input) of  $\sim 0.8$  mV and a width at the base of  $\sim 0.3 \mu\text{sec}$ . While most of them were attributed to the minute  $^{235}\text{U}$  impurity in the sample, some may have been due to subthreshold fission in  $^{238}\text{U}$ .

Eight resonances in  $^{238}\text{U}$  were selected for analysis between 37 and 237 eV, the region of highest cross section times neutron flux, i.e., detector signal. By virtue of our linear-logarithmic amplifiers, individual fragment pulses on a low baseline were readily observed, but those near resonance peaks were not visible. Each resonance was then analyzed by the following procedure. A level of 4 mV was established as the point above which fragments could not be reliably counted. Each resonance was divided into that part below 4 mV (the resonance wings) and that above 4 mV (the central region). The ratio of wing area to total area was determined by integration of the smoothly varying electron signal. The number of fragments in the wings (from 0 to 3 per resonance in our sample of eight resonances) was counted, multiplied by the ratio of the total resonance area to wing area, and converted to a fission area in

TABLE I. Summary of experimental observations. Net  $^{238}\text{U}$  pulses =  $2.1 \pm 3.9$ . Corresponding average fission width =  $(0.03 \pm 0.05) \times 10^{-6}$  eV.

Resonance energy (eV)	Number of observed fragment pulses in wings	Expected fragment pulses from $^{235}\text{U}$	Fraction of resonance area in wings (%)
37	0	1.1	2.5
66	0	0.6	2.2
103	2	2.1	3.1
117	2	0.9	3.6
165	0	0.4	35.9
190	3	0.3	12.5
209	1	0.3	21.2
237	0	0.3	36.7
Total	$8 \pm 3.4$	$5.9 \pm 1.9$	

b eV. Then, since the experimental fission area is equal to  $C\Gamma_n\Gamma_f/E_n\Gamma$ , and  $C$ ,  $\Gamma_n$ ,  $\Gamma$ , and  $E_n$  are known,<sup>3</sup> it was possible to calculate a fission width for each resonance. Because the number of fragments observed in the wings averaged only one per resonance, they were summed and an average fission width derived.

The final step was subtraction of the expected contribution from the 1.9 mg of <sup>235</sup>U in the 3.8 mg of <sup>238</sup>U in the beam. Sample purity was stated to be <0.5 ppm <sup>235</sup>U. By counting fragments in the <sup>238</sup>U signal in two regions (50–60 eV and 68–92 eV) between <sup>238</sup>U resonances but with a high <sup>235</sup>U fission cross section, and comparing directly with a signal from one of the <sup>235</sup>U samples, this level of impurity was confirmed to  $\pm 32\%$  ( $26 \pm 8$  fragments observed). The same signal from a <sup>235</sup>U sample then allowed a prediction of the fragments due to <sup>235</sup>U in the specific resonance wing regions used to test for <sup>238</sup>U fission. The results of this analysis are presented in Table I.

Uncertainties (standard deviations) are based on the statistical uncertainty and an estimate of the reliability ( $\pm 25\%$ ) of fragment pulse counting from the film record. The measured fission width of  $(0.03 \pm 0.05) \times 10^{-6}$  eV is consistent with a vanishingly small value, or may alternatively be considered to establish an upper value somewhat below the measurement of Ref. 1. It has the additional advantage of being an average over a number of resonances. If it is assumed that this average value also holds at 0.025 eV, the relation  $\sigma_f = \sigma_c\Gamma_f/\Gamma_c$  can be used to calculate the thermal-neutron fission cross section. With  $\sigma_c(\text{thermal}) = 2.7$  b,  $\Gamma_c = 0.025$  eV, and  $\Gamma_f = (0.03 \pm 0.05) \times 10^{-6}$  eV, the value for  $\sigma_f(\text{thermal})$  is  $(3 \pm 5) \times 10^{-6}$  b.

### III. keV REGION

Between  $10^4$  and  $10^5$  eV, the <sup>238</sup>U signal was also amenable to analysis because the baseline level was low enough to allow observation of individual fragment pulses. In addition, individual <sup>238</sup>U capture resonances essentially had disappeared, in contrast to the lower-energy region. Between  $10^4$  and  $3 \times 10^4$  eV, 12 fragments were counted, while between  $3 \times 10^4$  and  $10^5$  eV, 13 fragments were counted. Here the <sup>235</sup>U contribution was negligible, 0.4 fragments being expected in each region. These count rates correspond to  $61 \mu\text{b} \pm 40\%$  and  $36 \mu\text{b} \pm 40\%$  for the two regions, respectively, the uncertainty being a combination of statistical un-

certainty ( $\pm 30\%$ ) and an estimate of the reliability ( $\pm 25\%$ ) of fragment pulse counting from the film record.

### IV. DISCUSSION

Comparison can be made between the value for  $\bar{\Gamma}_f$  determined around 100 eV and the average fission cross section measured at 30 keV. Assuming that the *s*-wave contribution at 30 keV is  $\frac{1}{4}$  of the observed cross section, or  $10 \times 10^{-6}$  b, the average fission width can be derived from a sum of *s*-wave Breit-Wigner resonances,  $\bar{\sigma}_f \approx C\bar{\Gamma}_n\bar{\Gamma}_f/\bar{\Gamma}E_nD$ . For  $\bar{\sigma}_f = 10 \times 10^{-6}$  b at  $E_n = 3 \times 10^4$  eV, with the average level spacing  $D = 18$  eV, and  $\bar{\Gamma}_n/\bar{\Gamma} \approx 1$ , the calculated value for  $\bar{\Gamma}_f$  is  $1.3 \times 10^{-6}$  eV. This value of  $1.3 \times 10^{-6}$  eV at 30 keV can be compared with the upper limit of  $0.13 \times 10^{-6}$  eV for the 100-eV region.

This general problem has been discussed by Lynn,<sup>4</sup> who calculated *p*-wave fission cross sections for even-target nuclei at 30 keV. Using Fig. 8.28 of Ref. 4, the experimental value of  $30 \times 10^{-6}$  b can be reproduced with a fission-barrier height  $E_f = 1.5$  MeV and curvature  $\hbar\omega_f = 0.67$  MeV.

In terms of a doubled-peaked fission barrier, the penetrability *P* can be calculated by the method of Cramer and Nix<sup>5</sup> for reasonable values of the barrier parameters. Since little is known directly about the parameters of the compound nucleus <sup>239</sup>U, values were extrapolated from neighboring nuclei. For barrier heights relative to the ground state of  $E_A = 5.7$  MeV,  $E_{II} = 2.6$  MeV, and  $E_B = 6.3$  MeV for the first peak, second minimum, and second peak, respectively, and curvatures  $\hbar\omega_A = 1.2$  MeV,  $\hbar\omega_{II} = 0.7$  MeV, and  $\hbar\omega_B = 0.7$  MeV, the value for  $\bar{\Gamma}_f = DP/2\pi$  is within a factor of 10 of the experimental value of  $0.03 \times 10^{-6}$  eV at the neutron binding energy of 4.8 MeV.

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## Energy and Angular Distribution of Alpha Particles Emitted in Thermal-Neutron Fission of $^{235}\text{U}$

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The energy and angular distributions of  $\alpha$  particles emitted in thermal-neutron fission of  $^{235}\text{U}$  have been studied in a three-parameter correlation experiment for  $\alpha$  angles between 50 and 130°. These distributions are compared with the results of trajectory calculations to get a "best fit" set of initial scission conditions. The derived initial fragment separation of 24 fm and initial average kinetic energy of 31 MeV are in good agreement with liquid-drop calculations, while the initial most probable  $\alpha$  energy was found to be 3.4 MeV. The results indicate a shift in the scission point of 1–2 fm towards the light fragment, for a variation in mass ratio between 1.1 and 2. The effect of  $\alpha$  recoil on the experimental distributions is discussed in detail.

### I. INTRODUCTION

The fission process is normally a binary process in which the fissioning nucleus splits into two fragments of comparable mass. However, once in every few hundred fission events, a third light-mass charged particle is emitted, the great majority of these being  $\alpha$  particles. Despite the relatively low yield, great interest has been shown in recent years in the long-range alpha (LRA) fission mode as a tool for studying the configuration of the nucleus at the moment of scission.<sup>1,2</sup>

The early experiments reported on LRA fission<sup>3–5</sup> were carried out using the nuclear emulsion technique, and showed the preferential emission of the  $\alpha$  particles in a direction normal to the fission axis. The development of solid-state detectors permitted detailed measurements of the  $\alpha$  and fragment energy distributions, both in spontaneous fission of  $^{252}\text{Cf}$ <sup>2,6,7</sup> and in thermal-neutron-induced fission in  $^{235}\text{U}$ .<sup>8,9</sup> These showed the general similarity between binary and LRA fission with regard to fragment mass and energy distributions. In the case of  $^{252}\text{Cf}$ <sup>2,6</sup> the angular distributions were measured for  $\alpha$  angles between 60 and 120°. A similar experiment reported for  $^{235}\text{U}$  thermal fission<sup>10,11</sup> utilized the emulsion technique.

The pronounced peaked angular distribution of the  $\alpha$  particles perpendicular to the direction of

the fission fragments suggests that the  $\alpha$  particles are emitted during the scission process, and are probably emitted close to the actual point of scission.<sup>4</sup> Detailed trajectory calculations have been carried out by Halpern,<sup>1</sup> Geilikman and Khlebnikov,<sup>12</sup> Boneh, Fraenkel, and Nebenzahl,<sup>13</sup> Katase,<sup>14</sup> and Fong<sup>15</sup> based on the Coulomb repulsion between the three particles, and a "best fit" of initial scission conditions was obtained from the experimental results. The calculations of Boneh *et al.*<sup>13</sup> for  $^{252}\text{Cf}$  spontaneous fission gave initial average values of  $\alpha$  energy of 3 MeV, a fragment separation of 26 fm, and total fragment kinetic energy of about 40 MeV at scission. These values are in contradiction to those obtained by Fong,<sup>15</sup> which give an initial fragment energy and  $\alpha$  energy of the order of 0.5 MeV. However, the higher value of initial fragment kinetic energy obtained by Boneh *et al.* fits quite well with the predictions of the liquid-drop calculations of Nix.<sup>16</sup> In this type of study it is important to carry out measurements at well-defined  $\alpha$  angles, which permits comparison of the experimental angular distributions with calculation. This also permits applying corrections due to  $\alpha$  recoil, which are angular dependent, to the experimental distributions.

The experimental arrangement is discussed in Sec. II, while the experimental results and discussion are presented in Sec. III. The Appendix discusses in detail the effect of  $\alpha$  recoil on geo-