# Velocities of Fragment Pairs from U<sup>233</sup>, U<sup>235</sup>, and Pu<sup>239</sup> Fission\*

WILLIAM E. STEIN

Los Alamos Scientific Laboratory of the University of California, Los Alamos, New Mexico (Received June 17, 1957)

Velocities of the fragment pairs from thermal-neutron-induced fission of U233, U235, and Pu239 have been measured by a time-of-flight method. The primary masses and energies of the fragments have been determined from the conservation of mass number and momentum. These results are compared with previous fission-fragment-mass and -energy measurements. A decrease of the total kinetic energy near the symmetric mode is observed, which is in agreement with previous double-ionization-chamber measurements. The intrinsic energy spread for mass-97 fragments was found to be  $8.1 \pm 1.6\%$  with a 95% confidence interval.

## I. INTRODUCTION

ASS yields from the fission of U<sup>233</sup>, U<sup>235</sup>, and Pu<sup>239</sup> have been investigated by radiochemical and mass spectrometric methods and have recently been summarized by Katcoff.1 The results of Thode and Graham<sup>2</sup> on the xenon abundances in U<sup>235</sup> fission indicate an abnormally high yield at masses 133 and 134, and the results of Glendenin et al.3 indicate a complementary fine structure at mass 100. These anomalies have been attributed to a preference for an 82-neutron configuration of the heavy fragment in the fission process,<sup>4</sup> to neutron boil-off following fission,<sup>5</sup> or to a combination of these effects. Since these yields represent the combined effects of fission and promptneutron emission, primary (before neutron emission) mass yield data would be useful in testing these hypotheses.

Kinetic energies of fission fragments have previously been deduced from the ionization produced by stopping the fragments in various gases.<sup>6</sup> Results of recent measurements have shown that the ionization thus produced is not simply proportional to the fragment energy.<sup>7</sup> These results are consistent with the nuclear recoil effect previously discussed by Knipp and Ling.8 From velocity measurements of single fragments, Leachman<sup>9</sup> concluded that the kinetic energies of the fragments exceeded by approximately 6 Mev those obtained by Brunton and Hanna<sup>10</sup> and Brunton and Thompson.<sup>11</sup> Furthermore, the width (full width at

- <sup>5</sup> L. E. Glendenin, Phys. Rev. 75, 337 (1949).
- <sup>6</sup> W. J. Whitehouse, *Progress in Nuclear Physics* (Pergamon Press Ltd., London, 1952), Vol. 2, p. 120. <sup>7</sup> H. W. Schmitt and R. B. Leachman, Phys. Rev. 102, 183
- (1956).
- <sup>8</sup> J. K. Knipp and R. C. Ling, Phys. Rev. 82, 30 (1951).
   <sup>9</sup> R. B. Leachman, Phys. Rev. 87, 444 (1952).
   <sup>10</sup> D. C. Brunton and G. C. Hanna, Can J. Research A28, 190 (1950).
- <sup>11</sup> D. C. Brunton and W. B. Thompson, Can. J. Research A28, 498 (1950).

half-maximum) of the dispersion in these ionization measurements was estimated to be about 9 Mev. When corrected for the above energy difference, the double-ionization-chamber measurement of the average total kinetic energy for fission of U<sup>235</sup> is in agreement with a recent calorimetric measurement.<sup>12</sup>

The variation of the kinetic energy with mass ratio has been considered previously.<sup>13,14</sup> Assuming that the charge of a fragment is proportional to its mass and that the kinetic energy is derived from the Coulomb repulsion of the two fragments, the total kinetic energy is expected to decrease monotonically as a function of mass ratio. However, the results obtained from the double-ionization-chamber measurements of Brunton et al.<sup>10,11</sup> have indicated that the total kinetic energy decreases near the symmetric mode. This decrease has been considered by Fong<sup>14</sup> to be the result of the relatively large dispersions associated with the ionization-chamber measurements.

A determination of the energy distribution of Zr<sup>97</sup> fragments from thermal-neutron-induced fission of U<sup>235</sup> has been reported by Cohen et al.<sup>15</sup> In this measurement, magnetic deflection and radiochemical methods were used. The full width at half-maximum of the total kinetic energy distribution for this mass ratio was deduced to be  $11.4 \pm 0.8\%$  with a most probable energy of  $174.7 \pm 2$  or  $158.5 \pm 2$  MeV, depending on the charge assignment. An energy spread of 5 to 8% for specific fission fragments has been reported by Good and Wollan.<sup>16</sup> Fong<sup>14</sup> predicts an intrinsic energy spread of 6 or 7% for the most probable modes of fission.

The present investigation was undertaken to provide additional data concerning the absolute fragment energies, primary mass distributions, and the intrinsic energy spreads. Time-of-flight techniques were used to determine the velocities of the fragment pairs. From these measured velocities, the primary masses and energies of the fragments were obtained from the principles of conservation of momentum and mass

<sup>\*</sup> Work performed under auspices of the U. S. Atomic Energy Commission.

S. Katcoff (private communication). <sup>2</sup> H. G. Thode and R. L. Graham, Can. J. Research A25, 1 (1947).

<sup>&</sup>lt;sup>3</sup> Glendenin, Steinberg, Inghram, and Hess, Phys. Rev. 84, 860 (1951).

<sup>&</sup>lt;sup>4</sup> Wiles, Smith, Horsley, and Thode, Can. J. Phys. 31, 419 (1953).

<sup>&</sup>lt;sup>12</sup> R. B. Leachman and W. D. Schafer, Can J. Phys. 33, 357

<sup>&</sup>lt;sup>16</sup> K. D. Leaenman and M. Ramsey, Los Alamos Scientific Laboratory Report LA-510 (MDDC-945), 1946 (unpublished).
<sup>14</sup> P. Fong, Phys. Rev. 102, 434 (1956).
<sup>15</sup> Cohen, Cohen, and Coley, Phys. Rev. 104, 1046 (1956).
<sup>16</sup> W. M. Good and E. O. Wollan, Phys. Rev. 101, 249 (1956).

TABLE I. Estimated full width at half-maximum dispersions for one and two neutrons emitted from each fragment. These values are for 269-cm drift distances and measured time resolution  $\Delta T = 5.5 \times 10^{-9}$  sec.

	One neutron from each fragment	Two neutrons from each fragment	
$\Delta v_L (10^7 \text{ cm/sec})$	4.6	5.1	
$\Delta v_H$ (10 <sup>7</sup> cm/sec)	2.4	2.9	
$\Delta M_L$ (mass numbers)	2.4	2.7	
$\Delta M_{H}$ (mass numbers)	2.4	2.7	
$\Delta E_L$ (Mev)	4.8	5.3	
$\Delta E_H$ (Mev)	3.0	3.4	

number. This method not only provides a better and more accurately determined energy resolution than the ionization method, but also allows an absolute determination of the fragment kinetic energies. Preliminary data obtained by the double-velocity method have been reported earlier.<sup>17</sup>

# **II. EQUIPMENT**

As shown schematically in Fig. 1, the velocities of the two fragments from a fission event were measured by their time-of-flight through two 269-cm evacuated drift tubes. Fission was induced in the fissile foil by a beam of thermal neutrons from a reactor. The time of each fission event was provided by the pulse  $P_0$ , which was obtained by detecting the electrons ejected as the fragment emerged from the source backing.<sup>18</sup> This time-of-fission detector consisted of the backing of the fissile deposit, an electron lens, and an electron detector. The methods used for recording the fragment drift times, analyzing the data, and determining the resolution of these velocity measurements were essentially the same as those previously described.<sup>9</sup> The time resolution associated with the present measurements was found to be Gaussian with a  $5.5 \times 10^{-9}$ -sec full width at half-maximum.

The electron detector and the smaller remote detector each consisted of a 2-inch-diameter, 0.001-inch-thick disk of plastic scintillator cemented to the face of a 6342 photomultiplier. The larger remote detector consisted of an 8-inch-diameter, 0.02-inch-thick disk of plastic phosphor cemented to a 2-inch-thick Lucite light pipe which was optically coupled to a 6364 photomultiplier. This large detector was required to insure an adequate efficiency of detecting both fragments from a particular fission event. The diameter of this detector was determined not only by the dimensions of the source and the other fragment detector but also from considerations of neutron emission from the moving fragments and the scattering of the fragments

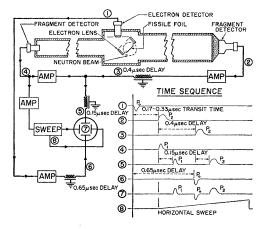


FIG. 1. Schematic diagram of the time-of-flight equipment. Pulses were amplified by Hewlett-Packard 460A and amplifiers and delayed by appropriate lengths of RG 7/U cable. The fragment time of flight is the time between the occurrence of  $P_0$  and  $P_1$  and that of the complementary fragment is the time between the occurrence of  $P_0$  and  $P_2$ . The  $P_1$  pulses were used to initiate the oscilloscope displays of the pulses. Photographs of these sweeps were analyzed for the times between pulses. The time scale was provided at frequent intervals by photographs of a 50-Mc/sec signal from a crystal-controlled oscillator.

in the source. The latter are effects that would cause the fragments to be noncollinear.

The sources were prepared by vacuum evaporation of the fissile material onto 0.1-mg/cm<sup>2</sup> nickel foils. The deposits of  $UO_2$  (enriched in  $U^{235}$ ),  $UF_4$  (enriched in U<sup>233</sup>), and PuF<sub>4</sub> were 38  $\mu$ g/cm<sup>2</sup>, 66  $\mu$ g/cm<sup>2</sup>, and 38  $\mu$ g/cm<sup>2</sup>, respectively. The average velocity loss of the fragments in the fissile material of these sources was estimated from previous measurements<sup>19</sup> to be less than 0.5%. Analysis of the present data showed the fragment velocity loss in the nickel to be less than 1%. These sources were mounted in the electron lens with the nickel side facing the electron detector.

#### **III. RESULTS**

The data obtained in this experiment were the velocities of 3050, 2070, and 680 fragment pairs from the thermal-neutron fission of U<sup>235</sup>, U<sup>233</sup>, and Pu<sup>239</sup>, respectively. Since the neutron emission time is small compared with the flight time of the fragments,<sup>20</sup> these measured quantities are the velocities of the fission fragments after prompt-neutron emission. However, with the assumption of isotropic emission of neutrons from the moving fragments, the most probable velocity after neutron emission is essentially equal to the fragment velocity before neutron emission. Thus, the measured velocities were considered to be the fragment velocities before prompt-neutron emission, with an increased velocity dispersion for each prompt neutron emitted in addition to the measured instrumental dispersion. The masses and energies were then

<sup>&</sup>lt;sup>17</sup> W. E. Stein, Bull. Am. Phys. Soc. Ser. II, 1, 96 (1956) and Atomic Energy Commission Report AECD-3729 (unpublished). (Office of Technical Services, U. S. Department of Commerce, Washington, D. C., 1955.) <sup>18</sup>W. E. Stein and R. B. Leachman, Rev. Sci. Instr. 27, 1049

<sup>(1956).</sup> 

<sup>&</sup>lt;sup>19</sup> J. A. Northrop and J. E. Brolley, Phys. Rev. 92, 1091(A) (1952).

<sup>&</sup>lt;sup>20</sup> J. S. Fraser, Phys. Rev. 88, 536 (1952).

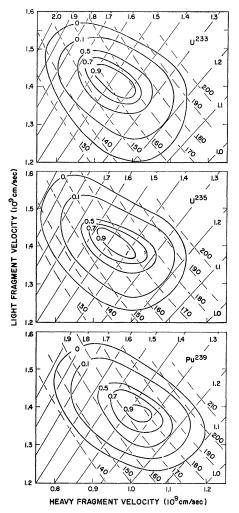


FIG. 2. Relative probability of fission modes for  $U^{233}$ ,  $U^{235}$ , and  $Pu^{239}$ . Contour lines are of relative probability, solid light lines are of constant mass ratio, and dashed lines are of constant kinetic energy in Mev.

computed by using the principles of conservation of momentum and mass number. Table I gives the estimated uncertainties of the velocity v, mass M, and

TABLE II. Comparison of kinetic energies. Values given for references 10 and 11 are most probable energies, whereas those listed for the present data are average energies. Probable errors assigned to the present data are based on estimates of possible systematic errors. The statistical standard deviations are  $\pm 1$  Mev or less.

	Previous data (Mev)			Present data (Mev)			
	$U^{233}$	$U_{235}$	Pu <sup>239</sup> F	Reference	$U^{233}$	$U^{235}$	Pu <sup>239</sup>
Light-fragment energy	93.0	94.5	94.6	10.11	97	98	100
Heavy-fragment energy	56.6	60.2	65.2	10,11	66	67	72
Light-fragment ionization defect Heavy-fragment ionization	6.1	5.7	5.2	9			
defect Total energy	7.3 163.0	6.5 166.9	6.4 171.4	9	$163 \pm 2$	$165\pm 2$	$172 \pm$

energy E for one and two neutrons emitted from each fragment. The subscripts L and H refer to the light and heavy fragments.

The relative probability of the various fission modes for the three isotopes investigated is shown in Fig. 2. Ionization data obtained by Brunton and Hanna<sup>10</sup> for  $U^{233}$  and  $U^{235}$  and by Brunton and Thompson<sup>11</sup> for  $Pu^{239}$  are compared with the energy data from this experiment in Fig. 3. Table II contains a summary of the fragment energies obtained by the ionization and double-velocity methods.

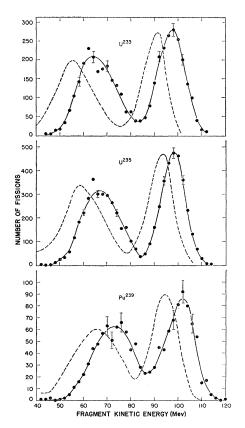


FIG. 3. Energy distributions of single fragments from U<sup>233</sup>, U<sup>235</sup>, and Pu<sup>239</sup>. The solid curves represent the data obtained from this experiment and the dashed curves are renormalized data from double-ionization-chamber measurements of references 10 and 11.

Shown in Fig. 4 are comparisons of the mass yields obtained by radiochemical and mass spectrometric methods<sup>1</sup> and the primary mass distributions which were obtained from the velocities of this experiment and the relation  $M_H/M_L = v_L/v_H$ , with  $M_L + M_H$ equaling the mass number of the compound nucleus undergoing fission. As expected, the primary mass yield curves are displaced toward larger mass numbers by an amount consistent with the average number of prompt neutrons emitted.<sup>21</sup> The fine structure in the

<sup>&</sup>lt;sup>21</sup> D. J. Hughes and J. A. Harvey, Neutron Cross Sections, Brookhaven National Laboratory Report BNL-325 (Superintend-

U<sup>235</sup> radiochemical and mass spectrometric data at mass 134 is not evident in the present data. In Fig. 5 the primary mass distribution in the heavy-fragment region is compared with the radiochemical and mass spectrometric yields dispersed by a Gaussian function with a full width at half-maximum of three mass numbers, corresponding to the estimated mass resolution of the present measurements. In terms of resolution, the distributions of Fig. 5 are then comparable, differing only by the effects of neutron emission. Within the mass resolution and statistical accuracy, the present data are inconsistent with a fine structure in the primary mass distribution at mass 134. They are,

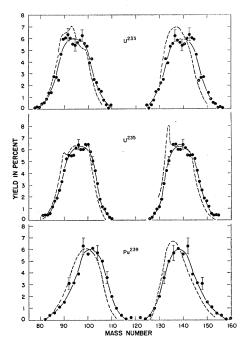


FIG. 4. Primary mass yields of fission fragments from U<sup>233</sup>, U<sup>235</sup>, and Pu239. The solid curves represent the data from this experiment and the dashed curves represent the radiochemical and mass spectrometric data of reference 1.

however, consistent with a possible fine structure in the 135 or 136 mass-number region.

In Fig. 6 the average values of the total kinetic energies are plotted against the mass ratio. Since the energy distributions for various mass ratios were found to be symmetrical, these data can be compared with the curves for the most probable total kinetic energy vs mass ratio given by the double-ionization-chamber measurements.<sup>10,11</sup> A decrease in total kinetic energy near the symmetric mode is apparent and is in agreement with range measurements of Katcoff et al.22

The velocity distributions for all masses were also obtained. As an example, the velocity distribution of

ent of Documents, U. S. Government Printing Office, Washington, D. C., 1955). <sup>22</sup> Katcoff, Miskel, and Stanley, Phys. Rev. 74, 631 (1948).



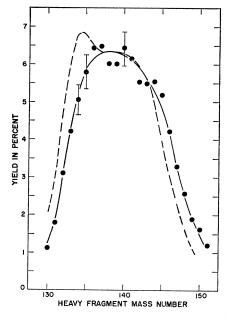
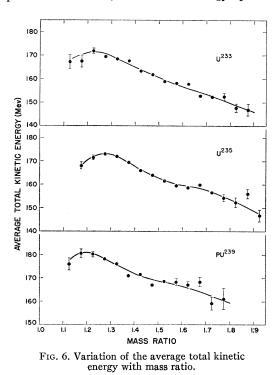


FIG. 5. U<sup>235</sup> heavy-fragment mass distributions. The solid curve represents the data from this experiment and the dashed curve represents the radiochemical and mass spectrometric data dispersed by a Gaussian function with a full width at half-maximum of three mass numbers.

U<sup>235</sup> fragments with mass 97 is given in Fig. 7. This velocity distribution has an average of  $1.39 \times 10^9$ cm/sec and a relative width (full-width at halfmaximum) of  $5.4 \pm 0.6\%$ . When the estimated velocity dispersion is removed, the intrinsic energy spread for



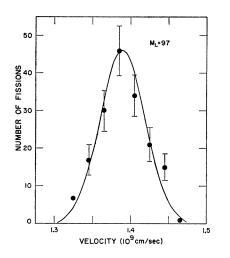


FIG. 7. Velocity distribution of U<sup>235</sup> fragments with mass 97.

mass 97 is found to be  $8.1\pm1.6\%$  with a 95% confidence interval. Since the momentum condition imposes a correlation of this spread with the energy spread of the complementary mass 139, an identical percentage energy spread is obtained for the full width at halfmaximum of the total energy distribution for this mass ratio.

### IV. DISCUSSION

The kinetic energy data from this experiment are in agreement with the corrected ionization data and the recent calorimetric measurement of the average kinetic energy of U<sup>235</sup> fragments. The decrease of the average total kinetic energy near the symmetric mode has been observed with these improved energy resolution measurements.

The mass yields from this experiment are consistent with mass data obtained by other means. They are, however, inadequate to test fully the fine structure hypotheses mentioned above. These data are inconsistent with a primary fine structure at mass 134, but are consistent with a possible fine structure at mass 135 or 136 in the primary mass distribution.

The  $8.1\pm1.6\%$  intrinsic energy spread for mass-97 fragments obtained from these U<sup>235</sup> data is lower than the  $11.4\pm0.8\%$  spread reported by Cohen<sup>15</sup> and is in better agreement with the 6 to 7% predicted by Fong.<sup>14</sup> The present energy spread is smaller than the 10.8% which was used by Leachman<sup>23</sup> in calculations of the emission of prompt neutrons from fission. It is, however, in better agreement with the minimum value of 9.8% which was also used in these calculations.

# V. ACKNOWLEDGMENTS

The author is indebted to Miss Elsie Pierce for reading the velocity data from the film, to Chester Kazek, Jr., for processing these data, to J. M. Peterson of the University of California Radiation Laboratory, Livermore, California, for supplying the thin plastic scintillators, and especially to Robert B. Leachman, who suggested the experiment, for his continued interest and advice.

<sup>23</sup> R. B. Leachman, Phys. Rev. 101, 1005 (1956).