

To prove: that

$$A \varphi[(C_k T' J')_a j_k(3), TJ] = 3 \sum_{i=1}^p (C_k T' J'; j_k \parallel \alpha_i TJ) \cdot \psi(\alpha_i, TJ), \quad (\text{A3})$$

where  $(C_k T' J'; j_k \parallel \alpha_i TJ)$  is a fractional parentage coefficient.

*Proof.*—The usual expansion of  $\psi(\alpha_i, TJ)$  is

$$\psi(\alpha_i, TJ) = \sum_k' \sum_{T' J'} \varphi[(C_k T' J')_a j_k(3), TJ] (C_k T' J'; j_k \parallel \alpha_i TJ), \quad (\text{A4})$$

where the first sum is over all distinct  $j_k$ . Since the set of  $\psi(\alpha_i, TJ)$  is complete, the wave function (A3) can be expanded as

$$A \varphi[(C_k T' J')_a j_k(3), TJ] = \sum_{i=1}^p b(\alpha_i) \cdot \psi(\alpha_i, TJ), \quad (\text{A5})$$

where

$$b(\alpha_i) = \int \psi(\alpha_i, TJ)^* \cdot A \varphi[(C_k T' J')_a j_k(3), TJ] d\tau_1 d\tau_2 d\tau_3. \quad (\text{A6})$$

The integration in (A6) is over the coordinates and spins of the three particles. Introducing the symmetrization operator  $S$ , analogous to  $A$  of (A1), and recalling that  $\psi(\alpha_i, TJ)$  is antisymmetric, we obtain

$$b(\alpha_i) = \int S \{ \psi(\alpha_i, TJ)^* \varphi[(C_k T' J')_a j_k(3), TJ] \} d\tau_1 d\tau_2 d\tau_3, \quad (\text{A7})$$

which, from (A4), equals just three times the same integral without  $S$ , and hence

$$b(\alpha_i) = 3 (C_k T' J'; j_k \parallel \alpha_i TJ), \quad (\text{A8})$$

which proves the theorem.

## Prompt Neutron Emission from Spontaneous-Fission Modes of $\text{Cf}^{252}\dagger$

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The number of prompt neutrons emitted and the velocities of the fragment pairs have been measured for individual spontaneous fissions of  $\text{Cf}^{252}$ . The time-of-flight measurements of the fragment velocities have sufficient resolution to provide a good determination of the mode of fission as characterized by the total kinetic energy  $E_K$  and the mass ratio  $R_A$  of the fragments. The neutrons are detected with high efficiency in a large cadmium-loaded liquid scintillator. It is found that the dependence of the average number of neutrons per fission,  $\bar{\nu}$ , on the parameters  $E_K$  and  $R_A$  may be approximated by a plane  $\bar{\nu}(E_K, R_A)$  over the region that includes the majority of the fission events. The slopes that specify the orientation of this plane are determined to be  $\partial \bar{\nu}(E_K, R_A) / \partial E_K = -0.143 \pm 0.020$  (neutrons/fission)/Mev and  $\partial \bar{\nu}(E_K, R_A) / \partial R_A = -6.3 \pm 1.1$  (neutrons/fission)/unit mass ratio. The value of the first slope indicates that the average total excitation energy of the fragments, required for the emission of one more neutron on the average, is  $7.0 \pm 1.0$  Mev. From this number and the measured dependence of  $\bar{\nu}$  on mass ratio, the average excitation energy  $\bar{E}_X$  of the fragments is determined as a function of the mass ratio. This function  $\bar{E}_X(R_A)$  and the measured dependence  $\bar{E}_K(R_A)$  determine the average prompt energy of fission as a function of mass ratio. The widths of the neutron-number distributions have been obtained as functions of  $E_K$  and  $R_A$ . The data do not support the statistical theory of fission proposed by Fong.

### I. INTRODUCTION

THE fission process, even for but one species of a spontaneously fissioning nucleus, yields a large variety of fragment nuclei and associated neutron and gamma radiations. Experimental technique has developed to the point where it has become feasible to

investigate correlations that may exist between modes of fission and the associated radiations. Measurements of the velocity distribution of fission-fragment pairs from  $\text{Cf}^{252}$  and the coincident gamma-ray spectrum have recently been reported.<sup>1</sup> Studies of the neutron-emission probability as a function of the mode of fission

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

<sup>1</sup> J. C. D. Milton and J. S. Fraser, Bull. Am. Phys. Soc. Ser. II, **2**, 197 (1957).

have been made, with uncertain resolution, for neutron-induced fission of  $U^{235}$  and for spontaneous fission of  $Cf^{252}$  (Fraser and Milton<sup>2</sup> and Hicks *et al.*,<sup>3</sup> respectively).

In our experiment<sup>4</sup> advantage is taken of both the high resolution<sup>5</sup> provided by the time-of-flight method<sup>6</sup> in determining the fission mode, and the high detection efficiency of the large cadmium-loaded liquid scintillator<sup>7</sup> in counting the fission neutrons,<sup>3,8,9</sup> to determine how the total number of prompt neutrons emitted in the spontaneous fission of  $Cf^{252}$  is affected by the division of the mass between the fragments and by the amount of the energy going into kinetic energy of these fragments. The low intensity of the fission source has imposed such a restriction on the flight distances of the fragments that the full resolution capability of the time-of-flight method has not been realized. An energy dispersion has been attained which is perhaps two thirds the size of that believed, from a comparison of the measured energy distributions, to be present in the  $Cf^{252}$  ionization-chamber measurements,<sup>3</sup> and a mass-ratio dispersion, therefore,<sup>5</sup> perhaps one-third as large. Dispersions of this order of magnitude have appreciable effects on the distributions of the average number of neutrons per fission as functions of the total kinetic energy and the mass ratio.

The measurement of the neutron-emission probabilities is of particular interest, since it provides the best measure at present of the excitation energies of the fission fragments at the time of scission, and since a knowledge of the magnitude and dependence on fission mode of these energies is basic to a description of the fission process. The energetics of a spontaneous-fission process is described by the following equations:

$$E_T = M - (m_H + m_L) = E_K + E_X, \quad (1)$$

where  $E_T$  is the energy made available when the mass  $M$  is split into the primary fragments of ground-state masses  $m_H$  and  $m_L$ . This energy is shared between the total kinetic energy  $E_K$  of the fragments and their total

excitation energy  $E_X$ . It is evident that  $E_X$  may be determined from measurements of  $E_K$  and the masses involved. Measurements of sufficient accuracy of the ground-state masses of the fragments are not available, however, and estimates obtained by extrapolation from measured masses of more-stable nuclides are necessarily uncertain. The total excitation energy  $E_X$  for a given total kinetic energy and mass-ratio mode, averaged over the nuclear-charge distributions of the two fragments, can be expressed in terms of an average prompt gamma-ray energy per fission  $\bar{E}_\gamma$ , derived from the residual excitation energy after the neutron emission,<sup>10</sup> the average number of neutrons emitted per fission  $\bar{\nu}$ , the average separation energy  $\bar{S}_\nu$  of a neutron, the average kinetic energy of a neutron in the rest-frame of a fragment  $\bar{E}_\nu$ , and an additional average prompt gamma-ray energy per neutron  $\bar{E}_{\nu\gamma}$ , to allow for the possibility of gamma-ray competition with neutron emission:

$$\bar{E}_X = \bar{E}_\gamma + \bar{\nu}(\bar{S}_\nu + \bar{E}_\nu + \bar{E}_{\nu\gamma}). \quad (2)$$

Equations (1) imply that  $\partial E_X / \partial \bar{\nu} = -(\partial \bar{\nu} / \partial E_K)^{-1}$ , which is found in the present experiment to be approximately constant over an extended region of the total kinetic energy  $E_K$  and the mass ratio  $R_A$  that includes the most probable modes. Therefore, in the region of the most probable modes, the dependence of the average total excitation energy on the fission mode can be determined from the measurements of  $\bar{\nu}(E_K, R_A)$ , the measured value of  $-(\partial \bar{\nu} / \partial \bar{E}_K)^{-1} = \partial \bar{E}_X / \partial \bar{\nu} = \bar{S}_\nu + \bar{E}_\nu + \bar{E}_{\nu\gamma}$ , and an estimate of the relatively small contribution from  $\bar{E}_\gamma$ .<sup>10</sup>

A statistical theory of fission, proposed by Fong,<sup>11</sup> explains the observed mass asymmetry of fission as resulting from larger excitation energies in the asymmetric modes. According to this view, one should expect to find in the experiment reported here a maximum in the neutron emission probability in the region of the most probable mass ratio.

## II. EXPERIMENTAL APPARATUS AND PROCEDURE

A schematic diagram of the apparatus and the electronic recording system is shown in Fig. 1.

### 1. Source

The source consists of an amount of  $Cf^{252}$  yielding about 2300 spontaneous fissions per minute deposited over an area of about 1 cm<sup>2</sup> on a 0.1-mg/cm<sup>2</sup> nickel foil. To prepare this source, a recently developed "electrostatic-spray" technique<sup>12</sup> was used.

### 2. Time-of-Flight Measurements

A time-of-fission signal<sup>13</sup> is obtained by mounting the source in an electrostatic lens, so that electrons, ejected

<sup>10</sup> R. B. Leachman and C. S. Kazek, Phys. Rev. **105**, 1511 (1957). A negligibly small dependence of  $\bar{E}_\gamma$  on  $\bar{\nu}$  is predicted by these calculations.

<sup>11</sup> P. Fong, Phys. Rev. **102**, 434 (1956).

<sup>12</sup> D. J. Carswell and J. Milsted, J. Nuclear Energy **4**, 51 (1957).

<sup>13</sup> W. E. Stein and R. B. Leachman, Rev. Sci. Instr. **27**, 1049 (1956).

<sup>2</sup> J. S. Fraser and J. C. D. Milton, Phys. Rev. **93**, 818 (1954).

<sup>3</sup> Hicks, Ise, Pyle, Choppin, and Harvey, Phys. Rev. **105**, 1507 (1957).

<sup>4</sup> A preliminary account of this work was presented at the American Physical Society meeting in Boulder, Colorado, on September 6, 1957. Whetstone, Stein, and Smith, Bull. Am. Phys. Soc. Ser. II, **2**, 308 (1957).

<sup>5</sup> The time-of-flight method can achieve, with reasonable fragment flight distances, energy dispersions perhaps half the size of those we estimate to be inherent in the best of the ionization-chamber measurements. (The limitation in the time-of-flight precision being due, in principle, to the effects of the fragment recoil from neutron emission.) Furthermore, since the time-of-flight measurements permit the mass ratio of the fragments to be determined from a velocity ratio, rather than from an energy ratio, for the condition of equal energy dispersion in the two instruments, the dispersion in the measurement of a mass ratio by time-of-flight is slightly less than half the corresponding dispersion obtained by ionization chambers.

<sup>6</sup> W. E. Stein, Phys. Rev. **108**, 94 (1957).

<sup>7</sup> Reines, Cowan, Harrison, and Carter, Rev. Sci. Instr. **25**, 1061 (1954).

<sup>8</sup> Diven, Martin, Taschek, and Terrell, Phys. Rev. **101**, 1012 (1956).

<sup>9</sup> Hicks, Ise, and Pyle, Phys. Rev. **101**, 1016 (1956).

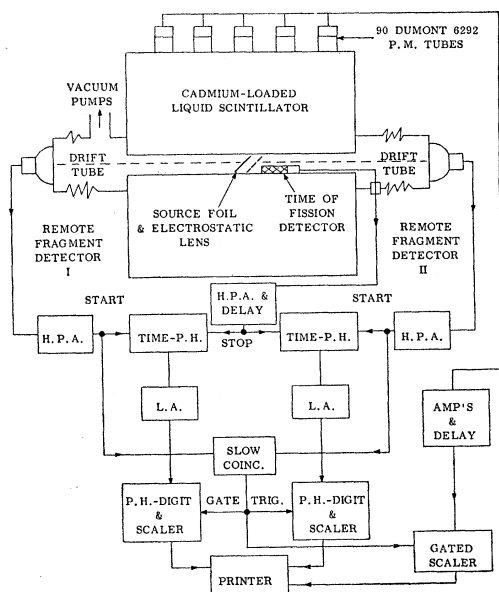


FIG. 1. Schematic diagram of the apparatus. H.P.A. = Hewlett-Packard Model 460B distributed amplifier, H.P.A. and DELAY = Hewlett-Packard Model 460A and 460B distributed amplifiers and 200-ohm cable. TIME-P.H. = time-to-pulse-height converter, L.A. = Modified Los Alamos Model 101A linear amplifier, P.H.-DIGIT = pulse-height-to-digital converter, AMP'S and DELAY = modified Los Alamos Model 503A preamplifier and amplifier, Hewlett-Packard Model 460A amplifier, 800 feet of RG/7U cable. The pulse-height-to-digital converters are gated on by a signal from the output of the SLOW COINC. = slow coincidence unit, which requires that start pulses appear at the inputs to both time converters within a time interval of about  $0.3 \mu\text{sec}$  covering the range of possible relative time differences for the flight times of the fragments from a given fission event. The use of this coincidence arrangement permits the time converters themselves to be operated at a very low input-voltage discrimination level, which reduces the timing jitter caused by the variation in height of the start pulses.

from the foil by one of the fragments, are accelerated and focused onto a 0.002-inch plastic scintillator which is cemented to the face of an RCA 6199 photomultiplier.

Fragments from a fission event that leave the foil within the proper solid angle travel in approximately opposite directions down the 152-cm-long evacuated flight tubes and strike the remote detectors. These detectors, which subtend solid angles of about  $3.5 \times 10^{-3}$  sterad at the source, consist of thin plastic scintillators cemented to the faces of RCA C7170 photomultipliers.

Pulses from the remote detectors are amplified and used to start the two time-to-pulse-height converters.<sup>14</sup> These two converters are stopped by the pulse, suitably delayed, from the time-of-fission detector. The height of the pulse at the output of each converter, which is proportional to the length of time that the converter is on, thus gives a measure of the flight time of the corresponding fragment. Each of these pulses, after

being suitably amplified by a linear amplifier, is fed into a 199-channel pulse-height-to-digital converter.<sup>15</sup> When the desired information appears in both pulse-height analyzers, a printer-control unit, not shown in Fig. 1, generates the proper blocking signals and transfers the scaled channel numbers *via* a Victor printer to paper tape.

The time corresponding to zero fragment flight time, the zero time, is determined for each fragment detector from the times measured when each remote detector is placed close to the source. An extrapolation to zero distance is then easily made. Care is taken that the signal-cable lengths for the remote and time-of-fission detectors are not disturbed. An absolute flight time for a fragment is then given by the difference between the zero time and the time measured with the detector in its remote position.

The timing circuits are calibrated by introducing pulses, separated in time by various known amounts, into the time converters. The various time separations of these pulses have been measured by photographing their oscilloscope traces in conjunction with those of a 50-Mc crystal-controlled oscillator. So that corrections can be made for the small drifts in the calibrations, due primarily to the effects of the diurnal temperature fluctuation, pulses derived from a single pulser are introduced automatically at hourly intervals into the signal outputs of the remote and time-of-fission detectors.

### 3. Neutron Measurements

The large cadmium-loaded liquid scintillator, which effectively surrounds the fission source, has been described elsewhere.<sup>7,8</sup> Briefly, the liquid is confined in a cylindrical tank, 30 inches long and  $28\frac{1}{2}$  inches in diameter, with a  $2\frac{5}{8}$ -inch-diameter tube along the axis. For this experiment a second tube is fitted into this tube so that the source foil and the time-of-fission detector can be introduced into the center of the scintillator in a vacuum. The scintillator solution is composed of triethylbenzene, 2.7 g/l of *p*-terphenyl, 0.1 g/l of POPOP,<sup>16</sup> and an amount of cadmium octoate<sup>17</sup> to give a ratio of cadmium to hydrogen atoms of  $1.3 \times 10^{-3}$ . The mean capture time for neutrons in this solution, as measured, is  $10 \mu\text{sec}$ .

The scintillator solution is viewed by 90 DuMont 6292 and 1177 photomultipliers. Pulses from these photomultipliers are amplified, delayed, clipped to  $0.15 \mu\text{sec}$ , and presented to a gated scaler. This fast flip-flop scaler, with a resolving-time capability of better than  $0.2 \mu\text{sec}$  for very narrow pulses, yields a resolving time of about  $0.3 \mu\text{sec}$  for the pulsewidths achieved by the amplification system and for the discriminator level

<sup>14</sup> The Los Alamos Model 13 converter. A similar instrument has been described, Weber, Johnstone, and Cranberg, *Rev. Sci. Instr.* **27**, 166 (1956). See also, L. Cranberg, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 4, p. 43.

<sup>15</sup> P. W. Byington and C. W. Johnstone, *Inst. Radio Engrs. Convention Record 3*, Part 10, 204 (1955).

<sup>16</sup> Ott, Hayes, Hansbury, and Kerr, *J. Am. Chem. Soc.* **79**, 5448 (1957).

<sup>17</sup> Ronzio, Cowan, and Reines, *Rev. Sci. Instr.* (to be published).

required to bias out the majority of the background pulses.

The scaler-gate generator unit is triggered by the same pulse that triggers the pulse-height-to-digital converters. After a delay time, adjusted to open the scaler gate 0.5  $\mu$ sec after the arrival of the prompt pulse produced by fission gamma-rays and proton recoils in the scintillator, the scaler is gated on for 50  $\mu$ sec to accept the pulses resulting from neutron captures in the scintillator, and then, after a delay of 500  $\mu$ sec, another scaler is gated on for 50  $\mu$ sec to sample the background. The number of pulses occurring in each gate is scaled separately, with a provision for a surplus to be indicated when the number in either scaler exceeds nine. The printer control ensures that the number of fission-neutron counts  $n_f$ , the number of background counts  $n_b$ , and a number indicating a surplus condition are printed on the paper tape with the numbers giving the fragment time-of-flight data for the same event.

#### 4. Data Treatment

The data were processed with the help of an IBM 704 data-processing machine. Calculations are made that determine from the time-of-flight data for each event the velocities  $v_H$  and  $v_L$ , the masses  $m_H$  and  $m_L$ , and the kinetic energies  $E_H$  and  $E_L$  of the heavy and the light fragments. To characterize more succinctly the mode of fission, the parameters  $R_A = m_H/m_L$  (the mass ratio) and  $E_K = E_H + E_L$  (the total kinetic energy of the fragments) are obtained. The calculations of the masses and energies are based on the conservation of mass number and momentum in the fission breakup, and on the usual assumption that the neutron emission from the fragments does not appreciably affect the mean velocities. A small adjustment is made in the course of

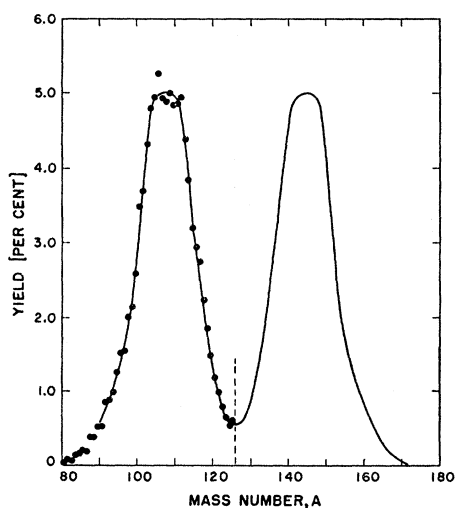


FIG. 2. The primary mass-yield distribution of the fragments. The measured distributions are necessarily symmetric about the mass number  $A = 126$ .

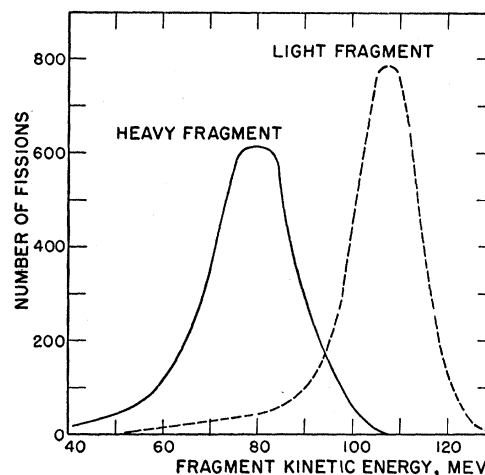


FIG. 3. The kinetic-energy distributions of the fragments. The solid curve represents the heavy fragment, the dashed curve, the light.

the calculation to ensure that the average velocities of the heavy- and light-fragment groups for those fragments passing through the foil match the corresponding velocities for fragments not passing through the foil. The data are then sorted for events falling within specified intervals of  $E_K$  and  $R_A$ , and the neutron numbers, associated with the selected events, treated to obtain the neutron-emission-probability distributions  $P(\nu)$  and the various moments of the distributions  $\bar{\nu}$ ,  $\langle \nu^2 \rangle_{av}$ ,  $\dots$ . Corrections for the resolution and efficiency of the neutron counter and for the background-count rate are made as described in reference 8.

### III. EXPERIMENTAL RESULTS

A total of 15 333 events have been analyzed. The time-of-flight data are summarized in Figs. 2 and 3, which show the primary mass number and kinetic-energy distributions of the fragments. From the analysis of these events and the measured<sup>8,9,18</sup> value of  $\bar{\nu} = 3.86 \pm 0.07$  averaged over all fission modes, the neutron-detection efficiency is found to have been  $78.1 \pm 2.1\%$ , where the uncertainty is a standard error due primarily to the uncertainty in  $\bar{\nu}$  and in the value of the resolution parameter<sup>8</sup>  $k = 2\tau \int f^2(t) dt = 0.028 \pm 0.014$ . The ratio of the number of background counts to the number of fission-neutron counts is 0.03.

#### 1. Distributions of $\bar{\nu}$

The dependence of the average number of neutrons per fission  $\bar{\nu}$  on the total kinetic energy of the fragments  $E_K$ , when no discrimination is made as to  $R_A$ , is shown in Fig. 4. There is a correlation between  $\bar{\nu}$  and  $E_K$ , particularly in the interval of  $E_K$  containing the majority of the events  $N(E_K)$ . The observed correlation is what one would expect qualitatively if there is a given average amount of available energy to be shared

<sup>18</sup> Obtained from an average of the results of references 8 and 9.

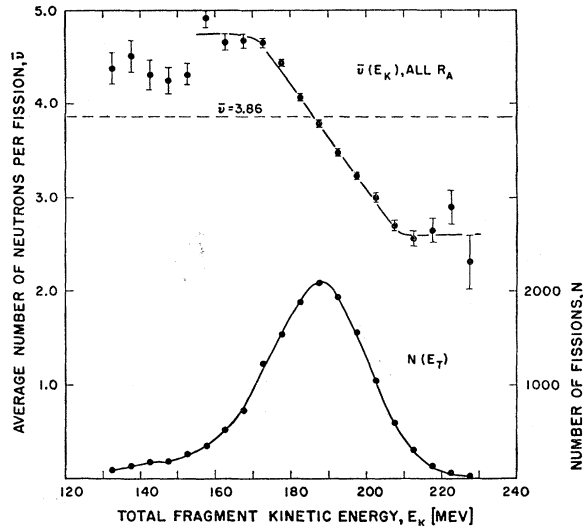


FIG. 4. The average number of neutrons per fission and the number distribution of fission events as functions of the total kinetic energy of the fragments, with no discrimination made as to the mass ratio of the fragments. Uncertainties shown are relative standard errors.

between the kinetic and excitation energies of the fragments.

From the data shown in Fig. 5 it is clear that there exists a distinct variation of  $\bar{\nu}$  with the mass ratio  $R_A$ , with no discrimination made with regard to  $E_K$ . The dependence appears to be more complicated, but again, across the interval containing the majority of the events  $N(R_A)$ , the variation is approximately linear.

In Fig. 6,  $\bar{\nu}$  is shown plotted against  $E_K$ , with the data separated into intervals of  $R_A$ . These curves, in the region containing the majority of the events

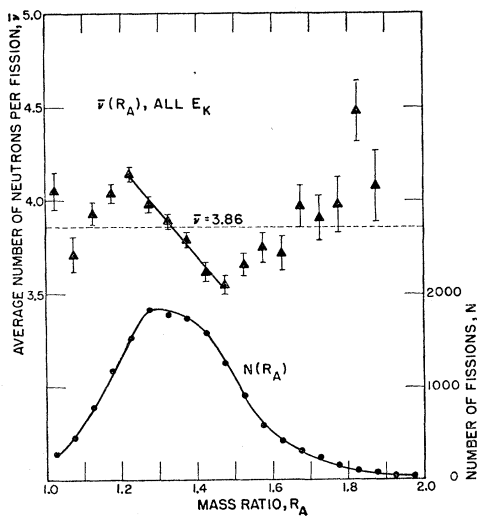


FIG. 5. The average number of neutrons per fission and the number distribution of fission events as functions of the mass ratio of the fragments, with no discrimination made as to the total kinetic energy of the fragments. Uncertainties shown are relative standard errors.

$N_{R_A}(E_K)$ , are approximately linear and steeper<sup>19</sup> than the curve  $\bar{\nu}(E_K)$  shown in Fig. 4. With larger mass ratios there appears to be a slight trend to steeper slopes.

In Fig. 7,  $\bar{\nu}$  is shown plotted against  $R_A$ , with the data separated into intervals of  $E_K$ . These curves, in the region containing the majority of the events  $N_{E_K}(R_A)$ , are also approximately linear and in every case steeper than the curve  $\bar{\nu}(R_A)$  shown in Fig. 5. There may be a very slight trend to steeper slopes with larger kinetic energies.

The results given above must be corrected for the effects of dispersions in the determination of  $E_K$  and  $R_A$ . These dispersions are given, in terms of the dispersions in the velocity determinations of the fragments, by the formulas:

$$\delta E_K/E_K = \delta R_A/R_A = [(\delta v_L/v_L)^2 + (\delta v_H/v_H)^2]^{1/2}.$$

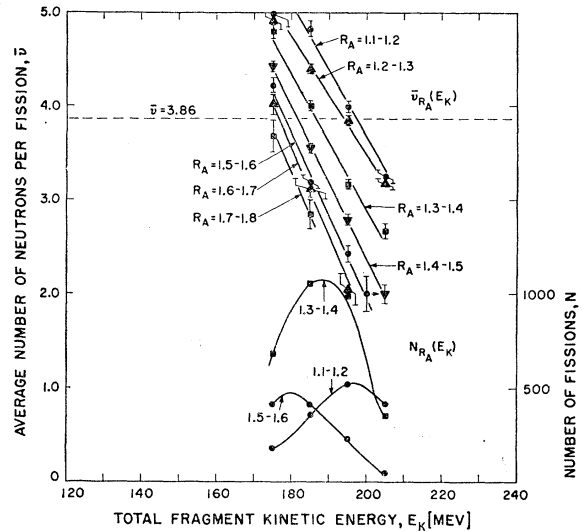


FIG. 6. The average number of neutrons per fission and the number distributions of fission events as functions of the total kinetic energy of the fragments, with the data separated into intervals of the mass ratio of the fragments. Uncertainties shown are relative standard errors.

(All dispersions are given in terms of the full-width at half-maximum of the dispersion functions assumed to be approximately Gaussian in shape.) The contribution to the velocity dispersion due to the instrumental limitations of the time-of-flight measurements is given by:

$$|\delta v/v| = v(\delta T/D),$$

where  $\delta T$  is the time resolution and  $D$  is the length of the flight path of the fragments. The time resolution is

<sup>19</sup> It can be seen qualitatively that, taking into account the correlation (shown in Fig. 10) between the mass ratio and the average total fragment kinetic energy, if the curves for  $\bar{\nu}(E_K)$  are approximately straight lines with equal slopes  $\partial \bar{\nu}(E_K, R_A)/\partial E_K$  for each value of  $R_A$ , then the weighted sum of these curves for all  $R_A$  will give a resultant curve  $\bar{\nu}$  versus  $E_K$  with a slope  $\partial \bar{\nu}(E_K)/\partial E_K$  that is less steep than the individual slopes. The argument also applies to the curves  $\bar{\nu}$  versus  $R_A$ .

determined from the width of the measured zero-time distributions, with small corrections made to allow for the velocity distribution of the fragments and for the uncompensated part of the effect on the electronics produced by the day-to-night temperature fluctuation. The result

$$\delta T = (6.4 \pm 0.6) \times 10^{-9} \text{ sec}$$

is obtained. The contribution to the dispersion due to the fragment recoil from neutron emission increases the dispersion by only about 10%. This is determined from the formula<sup>20</sup>:

$$\delta v/v = Cn^{1/2}/P,$$

where  $C = 2.33 \times 10^9$  cm-amu/sec,  $n$  = the number of neutrons emitted per fragment, and  $P = m_L v_L = m_H v_H$ . Combining the two dispersions with  $n = 2$ , one obtains,

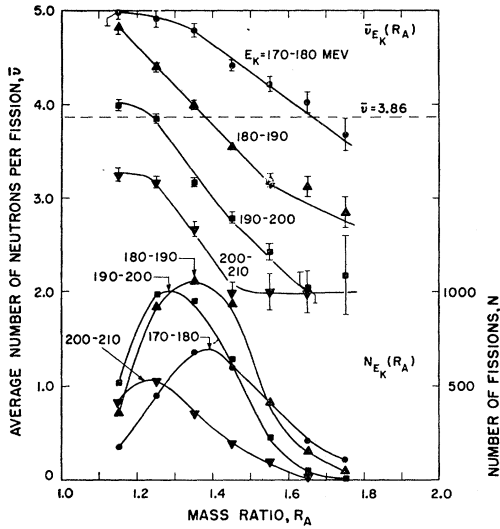


FIG. 7. The average number of neutrons per fission and the number distributions of fission events as functions of the mass ratio of the fragments, with the data separated into intervals of the total kinetic energy of the fragments. Uncertainties shown are relative standard errors.

for the most probable mass ratio:

$$\delta v_L/v_L = (6.2 \pm 0.6)\%, \quad \delta v_H/v_H = (5.0 \pm 0.5)\%.$$

Thus,

$$\delta E_K/E_K = \delta R_A/R_A = (8.0 \pm 0.8)\% \text{ (lower limit).}$$

Since the magnitude of the dispersive effect due to the source foil is difficult to determine, the value given

above is considered a lower limit to the dispersions. To obtain an estimate of an upper limit, a comparison is made between our time-of-flight data and those obtained by Milton and Fraser.<sup>1</sup> Based on their estimate<sup>21</sup> of  $(9.5 \pm 1.0)\%$  for the true relative width of the total kinetic-energy distribution of the fragments at  $R_A = 1.35$ , we find for our data

$$\delta E_K/E_K = \delta R_A/R_A = (9.25 \pm 0.50)\% \text{ (upper limit).}$$

A reasonable estimate of our dispersions is thus

$$\delta E_K/E_K = \delta R_A/R_A = (8.9 \pm 0.9)\% \text{ (accepted value).}$$

Therefore, near the most probable mode,

$$\delta E_K = 16.3 \pm 1.6 \text{ Mev} \quad \text{and} \quad \delta R_A = 0.12 \pm 0.012.$$

The data suggest that the "true" dependence of  $\bar{v}$  on  $R_A$  and  $E_K$  may be well approximated, at least in the region containing the majority of the events, by a plane  $\bar{v}(E_K, R_A) = \alpha E_K + \beta R_A + \bar{v}_0$ . If the further assumptions are made that the dispersion functions for  $E_K$  and  $R_A$  may each be approximated by normal distributions, and that the population distribution may be adequately represented by a function of the form<sup>23</sup>

$$\exp[-(E_K - \bar{E}_K)^2/(k\Delta E_K)^2 - (R_A - \bar{R}_A)^2/(k\Delta R_A)^2],$$

with the correlation between  $E_K$  and  $R_A$  approximated by  $\bar{E}_K = \rho R_A + \sigma$ , it is possible to perform analytically the convolution integrals necessary to obtain expressions for the corrected values of the slopes  $\alpha$  and  $\beta$  in terms of the observed slopes. These expressions are

$$\alpha = (\mathcal{E}\mathcal{R} + \rho^2\mathcal{B})(\mathcal{E}\alpha' - \rho\mathcal{B}\beta')/[\mathcal{E}(\mathcal{R} + \rho^2\mathcal{B}) - \rho^2\mathcal{A}\mathcal{B}], \quad (3)$$

$$\beta = (\mathcal{E}\mathcal{R} + \rho^2\mathcal{B})[(\mathcal{R} + \rho^2\mathcal{B})\beta' - \rho\mathcal{A}\alpha']/[\mathcal{E}(\mathcal{R} + \rho^2\mathcal{B}) - \rho^2\mathcal{A}\mathcal{B}], \quad (4)$$

where  $\mathcal{E} = 1 + (\delta E_K/\Delta E_K)^2$ ,  $\mathcal{R} = 1 + (\delta R_A/\Delta R_A)^2$ ,  $\mathcal{A} = (\delta E_K/\Delta E_K)^2$ ,  $\mathcal{B} = (\delta R_A/\Delta E_K)^2$ , and  $\rho = \partial \bar{E}_K/\partial R_A$ . Also  $\alpha \equiv \partial \bar{v}(E_K, R_A)/\partial E_K$ ,  $\beta \equiv \partial \bar{v}(E_K, R_A)/\partial R_A$ ; primed symbols indicate observed values and unprimed symbols indicate corrected values.  $\delta E_K$  and  $\delta R_A$  are the full-widths at half-maximum of the dispersion functions in the indicated variable.  $\Delta E_K$  is the corrected full-width at half-maximum of the total kinetic-energy distribution for a given mass ratio, and  $\Delta R_A$ , the corrected full-width at half-maximum of the mass-ratio distribution for all values of the total kinetic energy.

For the conditions of this experiment, these equations reduce to the approximate relations  $\alpha \cong \mathcal{E}\alpha'$  and  $\beta \cong \mathcal{R}\beta' - \rho\mathcal{A}\alpha'$ , which are used to estimate the propagation of errors.

To obtain values for  $\alpha$  and  $\beta$  representative of all of the data, in the spirit of the simple representation of the data assumed, weighted averages of the values of

<sup>21</sup> J. C. D. Milton (private communication).

<sup>22</sup> A 10% uncertainty has been assigned by the present authors to ensure a conservative estimate of the upper limit for the value of the dispersion.

<sup>23</sup> The factor  $k$  is inserted for consistency, so that all widths in these calculations are full-widths at half-maximum.

<sup>20</sup> The derivation of this formula is based on the simple neutron evaporation model that describes the neutron-energy distribution in the center-of-mass system of the fragment by a single Maxwellian distribution of the form  $E^{1/2}e^{-E/Q}$ . This distribution, with the nuclear temperature  $Q$  of 1 Mev assumed, has been shown by B. E. Watt, Phys. Rev. **87**, 1037 (1952), to give an acceptable fit to the observed neutron spectrum for thermal-neutron-induced fission of  $U^{235}$ . Smith, Friedman, and Roberts, Phys. Rev. **108**, 411 (1957), have measured the neutron spectrum from  $Cf^{252}$  and found it essentially identical to that from  $U^{235}$ . Their curve-fitting calculations also favor a nuclear temperature of 1 Mev.

TABLE I. The values of the slopes  $\partial\bar{\nu}(E_K, R_A)/\partial E_K$  [in (neutrons/fission)/Mev] and  $\partial\bar{\nu}(E_K, R_A)/\partial R_A$  [in (neutrons/fission)/unit mass ratio] are as observed in the data of Figs. 6 and 7 and as corrected by Eqs. (3) and (4). The values of the slopes  $\partial\bar{\nu}(E_K)/\partial E_K$  and  $\partial\bar{\nu}(R_A)/\partial R_A$  are as observed in the data of Figs. 4 and 5 and as corrected by Eqs. (5) and (6). Uncertainties are standard errors.

Slope	Observed value	Corrected value
$\partial\bar{\nu}(E_K, R_A)/\partial E_K$	$-0.070 \pm 0.004$	$-0.143 \pm 0.020$
$\partial\bar{\nu}(E_K, R_A)/\partial R_A$	$-3.8 \pm 0.8$	$-6.3 \pm 1.1$
$[\partial\bar{\nu}(E_K)/\partial E_K]_{\text{all } R_A}$	$-0.056 \pm 0.003$	$-0.079 \pm 0.008$
$[\partial\bar{\nu}(R_A)/\partial R_A]_{\text{all } E_K}$	$-2.5 \pm 0.5$	$-2.8 \pm 0.6$

the slopes, obtained from least-square fits to the data points shown in Figs. 6 and 7, are used for the values of the observed slopes  $\alpha'$  and  $\beta'$ . The value  $\Delta E_K = 18 \pm 2$  Mev<sup>21</sup> is used for an average corrected total kinetic-energy width in the region of the most probable modes, and  $\Delta R_A = 0.34 \pm 0.04$  for the corrected mass-ratio width for all modes. The results of the calculations, with  $\alpha' = -0.070 \pm 0.004$ ,  $\beta' = -3.8 \pm 0.8$ ,  $\mathcal{E} = 1.97 \pm 0.27$ ,<sup>24</sup>  $\mathcal{R} = 1.17 \pm 0.05$ ,  $\alpha = 0.97 \pm 0.27$ ,  $\mathcal{B} = (5.9 \pm 2.2) \times 10^{-5}$ , and  $\rho = -25 \pm 5$ , are  $\alpha = -0.143 \pm 0.020$  and  $\beta = -6.3 \pm 1.1$ .

In a similar way, the observed slopes  $\gamma' \equiv \partial\bar{\nu}(E_K)/\partial E_K$  and  $\delta' \equiv \partial\bar{\nu}(R_A)/\partial R_A$  obtained from the data shown in Figs. 4 and 5 are corrected for the dispersions by the relations

$$\gamma = \mathcal{E}\gamma', \quad (5)$$

$$\delta = \mathcal{R}\delta'. \quad (6)$$

The values  $\gamma = -0.079 \pm 0.008$  and  $\delta = -2.8 \pm 0.6$  are obtained.

The results are summarized in Table I.

## 2. Distributions of $\langle \nu^2 \rangle_{\text{Av}} - \bar{\nu}^2$

A measure of the width of a neutron-emission-probability distribution  $P(\nu)$  is provided by the quantity  $D = \langle \nu^2 \rangle_{\text{Av}} - \bar{\nu}^2$ , which is essentially equal to the mean-square deviation from the mean. The most statistically significant portions of the distributions of  $D$  as a function of the mass ratio are shown in Fig. 8.  $D$  remains substantially constant, particularly in the most populated intervals of  $R_A$ . There is, perhaps, a trend to smaller values of  $D$  as the mass ratio increases. A large difference is found between values of  $D$  for the data separated into small intervals of both  $R_A$  and  $E_K$  and the values obtained with no discrimination made as to  $E_K$ . The latter values fall about the value of  $D = 1.53 \pm 0.04$ , found from all of the data, while the former lie in the neighborhood of  $0.75 \pm 0.15$ . No attempt has been made to correct these observed values.

<sup>24</sup>  $\Delta E_K$  has been increased to  $17.7 \pm 1.8$  Mev by the Sheppard adjustment, to correct for the 10-Mev grouping error in the determination of the slope  $\alpha'$  and, similarly,  $\Delta R_A$  has been increased to  $0.138 \pm 0.014$  to correct for the 0.05 grouping error in the determination of the slope  $\beta'$ .

## 3. Effects of Data Bias

The neutron-detection geometry dictated by the requirements of the time-of-flight measurements results in a loss of those neutrons emitted at small angles to the direction of motion of the detected fragments. The average angle for neutron loss is less than 5 degrees and the fraction of the total solid angle into which neutrons can escape the detector completely is approximately 0.4%. Unless there is an extremely sharp forward peaking of the neutron angular distribution that is preferential for particular fission modes, the effect on our data should be negligible.

A more important source of bias results from the use of remote detectors subtending equal solid angles at the source, a geometry chosen to achieve sufficient counting rate with the low-intensity source available. Since an effect of neutron emission is to cause the flight directions of the fragments to deviate further from collinearity, there is a bias against recording events in which a large number of neutrons are emitted. An expression has been derived, consistent with the results of the neutron angular distribution measured by Fraser,<sup>25</sup> for the probability of detecting a fragment in one of the remote detectors when the complementary fragment has been detected in the other as a function of the total number of neutrons emitted. The necessary integrations are performed graphically. The effect of these detection efficiencies when folded into neutron-number distributions characterized by constant widths and various average values, is to increase the average value in each case by  $0.08 \pm 0.02$  neutron. There is, therefore, a negligible effect on the values of the slopes of the  $\bar{\nu}$  distributions. These results are not sensitive

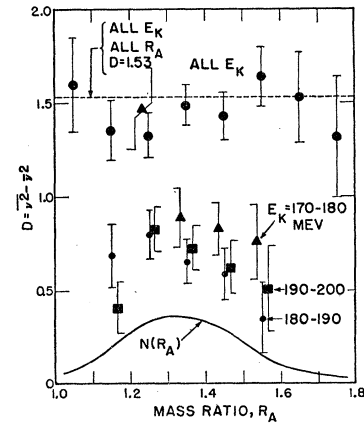


FIG. 8. Distributions of the neutron-number width parameter  $D = \langle \nu^2 \rangle_{\text{Av}} - \bar{\nu}^2$  as functions of the mass ratio of the fragments. The dashed line is drawn at the value of  $D$  determined from all of the data. The large circles show the distribution with no discrimination made as to the total kinetic energy of the fragments. The data, separated into intervals of the total kinetic energy of the fragment, give the distributions shown by the triangles (170–180 Mev), the circles (180–190 Mev), and the squares (190–200 Mev). Uncertainties shown are relative standard errors.

<sup>25</sup> J. S. Fraser, Phys. Rev. 88, 536 (1952).

to the choice of the width of the neutron-number distributions in the region of an rms full-width of two neutrons.

Strong evidence that the time-of-flight geometry does not introduce appreciable bias against the detection of events associated with larger numbers of neutrons is the satisfactory agreement between the neutron-emission-probability distribution  $P(\nu)$  obtained for this and other experiments. This distribution is shown in Fig. 9.

#### IV. DISCUSSION

It is found that the dependence of the average number of neutrons per fission  $\bar{\nu}$  on the total kinetic energy  $E_K$  and the mass ratio  $R_A$  may be approximated by a plane  $\bar{\nu}(E_K, R_A)$  over the region of  $E_K, R_A$  that includes the majority of the fission events. This plane is specified by the two slopes  $\partial\bar{\nu}(E_K, R_A)/\partial E_K = -0.143 \pm 0.020$  (neutron/fission)/Mev and  $\partial\bar{\nu}(E_K, R_A)/\partial R_A = -6.3 \pm 1.1$  (neutron/fission)/unit mass ratio and some intercept that establishes the over-all average value of  $\nu$ . There is a large variation in  $\bar{\nu}$  with  $E_K$  and  $R_A$ . Across the corrected full-width at half-maximum of the total kinetic-energy distribution for all fission modes,  $\bar{\nu}$  varies by almost four neutrons, and across the corrected full-width at half-maximum of the mass ratio distribution for all fission modes, by about two neutrons.

The value determined for the slope  $\partial\bar{\nu}(E_K, R_A)/\partial E_K$  should be compared with the value  $-0.116$  predicted by the calculations of Leachman and Kazek.<sup>10,26</sup> From this quantity it is possible to make an estimate of the average total fragment excitation energy  $E_X$  required for the emission of one more neutron on the average. The relation  $(\partial E_X/\partial \bar{\nu}) = -(\partial \bar{\nu}/\partial E_K)^{-1}$  gives, for the measured value, the result  $7.0 \pm 1.0$  Mev. This average excitation energy is the sum of an average neutron separation energy ( $5.5 \pm 1.0$  Mev) and an average kinetic energy of a fission neutron in the rest-frame of a fragment ( $1.5 \pm 0.5$  Mev), with probably no contribution from competing gamma radiation.

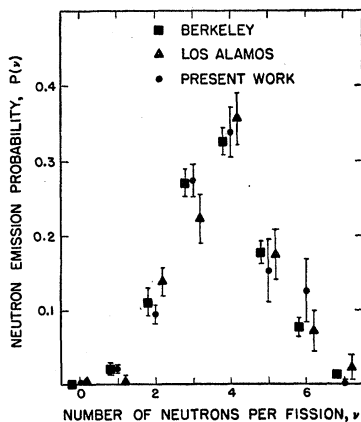


FIG. 9. Neutron-emission probabilities obtained in this experiment (circles) compared with the results obtained in Berkeley (reference 9) and Los Alamos (reference 8). Uncertainties shown are relative standard errors.

<sup>26</sup> The use of a nuclear temperature of 1 Mev in the calculations of reference 10 will improve the agreement of the two values (R. B. Leachman, private communication).

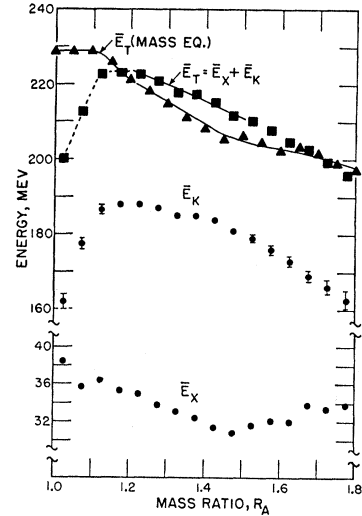


FIG. 10. The average total excitation energy  $\bar{E}_X$  of the fragments as a function of the mass ratio  $R_A$  has been obtained from Eq. (2):

$$\bar{E}_X(R_A) = \bar{E}_\gamma + \bar{\nu}(R_A) \cdot (\bar{S}_\nu + \bar{E}_\nu + \bar{E}_{\nu\gamma}),$$

with  $\bar{E}_\gamma = 6$  Mev,  $\bar{\nu}(R_A)$  as shown in Fig. 5,

$(\bar{S}_\nu + \bar{E}_\nu + \bar{E}_{\nu\gamma}) = -[\partial\bar{\nu}(E_K, R_A)/\partial E_K]^{-1} = 7$  Mev for  $R_A > 1.2$ , with a transition to 8 Mev below  $R_A = 1.2$ . The average total kinetic energy  $\bar{E}_K$  of the fragments as a function of  $R_A$  is determined from the fragment velocity measurements obtained in this experiment. The sum of  $\bar{E}_X + \bar{E}_K$  yields the average total prompt-fission energy  $\bar{E}_T$ , as a function of  $R_A$ , obtained from this experiment (squares). The curve  $\bar{E}_T$  (mass eq.) has been calculated from the mass equation  $E_T = M - (m_H + m_L)$ , where  $M$ ,  $m_H$ , and  $m_L$  are the masses of  $\text{Cf}^{252}$ , the heavy, and the light fragments, respectively, as given by the semiempirical mass table of Cameron (triangles).

This value of  $\partial\bar{E}_X/\partial\bar{\nu} = (\bar{S}_\nu + \bar{E}_\nu + \bar{E}_{\nu\gamma})$  and the measured dependence of  $\bar{\nu}$  on the mass ratio (shown in Fig. 5) is used in Eq. (2) to determine the variation of the average total excitation energy  $\bar{E}_X$  with mass ratio in the range of  $R_A = 1.2$  to 1.5, within which range the assumptions concerning the simple behavior of  $\partial\bar{E}_X/\partial\bar{\nu}$  seem justified. It is assumed<sup>1</sup> that the average gamma-ray energy per fission  $\bar{E}_\gamma$  is approximately constant<sup>27</sup> and equal to  $6 \pm 3$  Mev.<sup>28</sup> If, in addition, the measured dependence of the average total kinetic energy  $\bar{E}_K$  on the mass ratio is included, the variation of the average total prompt-fission energy  $\bar{E}_T$  with mass ratio is obtained. This is shown in Fig. 10. For comparison, the dependence of  $\bar{E}_T$  on  $R_A$  as calculated from the mass equation [Eq. (1)] by using the semi-

<sup>27</sup> The measurements of Milton and Fraser, reference 1, indicate that the variation of the average prompt-gamma-ray energy per fission of  $\text{Cf}^{252}$  across the full-width of the mass-ratio distribution for all total kinetic energies is less than 10 to 20%.

<sup>28</sup> The average prompt-gamma-ray energy per fission of  $\text{Cf}^{252}$  is 8 to 9 Mev according to the measurements of Smith, Fields, and Friedman, Phys. Rev. **104**, 699 (1956), and H. R. Bowman and L. G. Mann, Phys. Rev. **98**, 277 (1955). See also reference 8 in R. B. Leachman and C. S. Kazek, Jr., Phys. Rev. **105**, 1511 (1957). The calculations of Leachman and Kazek give  $\bar{E}_\gamma = 4.0$  Mev. It is not certain whether the discrepancy can be attributed to competing gamma-ray energy.



empirical mass tables of Cameron<sup>29</sup> is also shown. In the calculation of this dependence, the modified postulate of equal-charge displacement<sup>30</sup> and the stability curves of Coryell, Brightsen, and Pappas<sup>31</sup> are used to determine the most probable integral fragment charges  $Z_p$ . To average over the charge distributions, only the neighboring isobars  $A$ ,  $Z_p \pm 1$  are considered and it is assumed that these occur half as frequently as the most probable nuclide  $A$ ,  $Z_p$ . This tends to average out the effects of the pairing term in the semiempirical mass formula. From these calculations we also find the average neutron-separation energy,  $\bar{S}_v(R_A) = 5.0 \pm 0.2$  Mev in the range  $R_A = 1.2$  to  $1.7$ , which is in agreement with the results found for  $\partial \bar{E}_X / \partial \bar{v}$ . Below  $R_A = 1.2$ , we find that  $\bar{S}_v(R_A)$  rises rapidly to a nearly constant value of 6.0 Mev as the 82-neutron shell is crossed. The correction of  $\partial \bar{E}_X / \partial \bar{v}$  for this increase in  $\bar{S}_v$ , which has been included in Fig. 10, is not sufficient to bring the curves for  $\bar{E}_T$  into agreement below  $R_A = 1.1$ . This is probably attributable to the measured behavior of  $\bar{E}_K$  near the symmetric mode. Since the number of events obtained in this region is small, the data is here more vulnerable to possible systematic error.<sup>32</sup> The agreement between the two curves for  $R_A > 1.1$  is remarkably good. It should be noted that the *measured* dependences of  $\bar{E}_K$  and  $\bar{v}$  have been used in the construction of Fig. 10. If corrections were made for the effects of the dispersion in the determination of  $R_A$ , the curve  $\bar{E}_T = \bar{E}_K + \bar{E}_X$  would become more nearly parallel to the curve  $\bar{E}_T$  (mass equation) in the region of the most probable mode. The vertical displacement of the curves is no larger than the uncertainty in the measurements of the absolute values of  $\bar{E}_K$ .

The distributions of  $\bar{v}$  show no maxima in the regions of either the most probable mass ratio or the most probable total fragment kinetic energy. (See Figs. 6 and 7.) Nor does the average total excitation energy  $\bar{E}_X(R_A)$  have a maximum near  $R_A = 1.35$  (see Fig. 10). The absence of a maximum in the neutron-emission probabilities or in the average excitation energy at the most probable mode of fission does not support an argument of Fong's statistical theory<sup>11</sup> that asymmetric fission modes are preferred because of higher excitation energies of the fragments in these modes.

<sup>29</sup> A. G. W. Cameron, Chalk River Report CRP-690, 1957 (unpublished).

<sup>30</sup> A. C. Pappas, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 7, p. 21.

<sup>31</sup> Coryell, Brightsen, and Pappas, *Phys. Rev.* **85**, 732 (1952). See C. D. Coryell, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. 2, p. 325.

<sup>32</sup> The data of reference 1 do not show such a pronounced decrease of  $\bar{E}_K$  near the symmetric mode.

The values obtained for the widths of the neutron-number distributions (see Fig. 8) may be interpreted in terms of the widths of equivalent distributions of the total excitation energy of the fragments. These widths do not show a strong dependence on the fission-mode parameters  $E_K$  and  $R_A$ , although it must be understood that the limited statistics and resolution of this experiment affect the width measurements more severely than the measurements of  $\bar{v}$ . An average excitation energy of  $7.0 \pm 1.0$  Mev per neutron indicates that the full-widths at half-maximum of the distributions of total excitation energy for individual mass-ratio modes all lie near the value  $20 \pm 3$  Mev obtained for the corresponding width of the distribution for all modes of fission.<sup>33</sup> There is an indication of a minimum width equal to  $19 \pm 2$  Mev in the neighborhood of the most probable mass-ratio mode, which is comparable to the 17.5 Mev width of the distribution of the total kinetic energy of the fragments for  $R_A = 1.35$  determined by Milton and Fraser.<sup>1,21</sup> Such an agreement is implied by the relation  $E_T = E_K + E_X$ . That the excitation-energy widths are correlated with the kinetic-energy widths is evident from the much smaller widths measured for modes specified *both* as to mass ratio and total kinetic energy. These full-widths at half-maximum, as defined by our grouping and dispersions are about  $14 \pm 2$  Mev, indicating that the true total excitation-energy widths for individual  $E_K$ ,  $R_A$  modes are certainly less than about 7 Mev. A finite width is expected because of the nuclear charge distribution at a given mass ratio.

## V. ACKNOWLEDGMENTS

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<sup>33</sup> This value is in agreement with the value  $19 \pm 2$  Mev determined by J. Terrell, *Phys. Rev.* **108**, 783 (1957), who assumed an average excitation energy per neutron of  $6.7 \pm 0.7$  Mev and the value  $D = 1.54 \pm 0.04$  obtained from a treatment of the data of references 8 and 9.