

## Energy Dependence of Prompt $\bar{\nu}$ for Neutron-Induced Fission of $U^{235}\dagger$

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The energy dependence of  $\bar{\nu}_p$ , the average number of prompt neutrons per fission, for  $U^{235}$  has been studied over the incident neutron energy range 0.03 to 1.76 Mev by measuring the ratio of prompt neutrons from neutron-induced fission of  $U^{235}$  to those from spontaneous fission of  $Cf^{252}$ . The extrapolated value at  $E_n=0$  has been normalized to  $\bar{\nu}_p$  for  $U^{235}$  at thermal energies. The results have been fitted to an equation of the type  $\bar{\nu}_p = \bar{\nu}_p(0) + aE_n$ , where  $E_n$  is the incident neutron energy in Mev, yielding  $\bar{\nu}_p(U^{235}) = 2.414 + (0.097 \pm 0.008)E_n$ .  $\bar{\nu}_p$  for  $Cf^{252}$  spontaneous fission is  $3.754 \pm 0.045$ .

### INTRODUCTION

STUDIES of the rate of change of  $\bar{\nu}_p$ , the average number of prompt fission neutrons per fission, with the energy of the incident particle,  $E_n$ , can give information as to the distribution of excitation energy during the fission process. Several authors<sup>1-3</sup> have predicted a linear dependence with energy. They assumed that  $E_n$  goes entirely into the excitation energy of the fragments. Terrell<sup>2</sup> has suggested that

$$d\bar{\nu}_p/dE_n \simeq 1/E_0, \quad (1)$$

where  $E_0$  is the average loss in excitation energy per fission neutron. Bondarenko *et al.*<sup>3</sup> found a similar relation for  $d\bar{\nu}_p/dE_n$  but included the effects of the hardening of the neutron spectrum with increasing excitation energy and the increase in the average neutron separation energy with the increasing number of neutrons emitted. Their expression for  $d\bar{\nu}_p/dE_n$  will have the same form as Eq. (1), providing  $E_0$  is redefined as the average loss in excitation energy by the fission fragment on emitting the next neutron.

They also argue that the linear dependence should persist past the  $(n, nf)$  threshold since  $E_0$  should be roughly the same for neutrons emitted before or after fission. Furthermore,  $d\bar{\nu}_p/dE_n$  should be nearly the same for fissionable nuclides which do not differ greatly in charge and mass since there will then be little difference in the average energy required to remove a neutron from the fragments and a correspondingly small difference in  $E_0$ . However, Smith<sup>4</sup> has critically reviewed the experimental values of  $\bar{\nu}$  for neutron-induced fission up to 15 Mev and obtains values of  $d\bar{\nu}/dE_n$  ranging from 0.11 Mev<sup>-1</sup> for Pu<sup>239</sup> to 0.15 Mev<sup>-1</sup> for Th<sup>232</sup>. These results are derived from total  $\bar{\nu}$  values but  $d\bar{\nu}/dE_n$  should be about the same for both prompt and total  $\bar{\nu}$  since the

delayed neutron fraction is not sensitive to the incident neutron energy.<sup>5</sup> Close examination of the data<sup>6</sup> for  $U^{235}$  shows that below 2 Mev  $d\bar{\nu}/dE_n$  may be less than the 0.135 Mev<sup>-1</sup> obtained from all measurements up to 15 Mev, but the data in this region are not of sufficient accuracy to tell if this difference is significant. In the present experiment a precise determination of the energy dependence of  $\bar{\nu}_p$  for  $U^{235}$  has been made over the incident neutron energy range 0.03 to 1.76 Mev by measuring the ratio of prompt neutrons from neutron-induced fission of  $U^{235}$  to those from spontaneous fission of  $Cf^{252}$ .

### EXPERIMENTAL APPARATUS AND PROCEDURE

The method used involved the measurement of coincidences between a fission counter and a neutron detector. In order to avoid the possibility of a change in the accidental rate or in the efficiency of the neutron detector, measurements of  $U^{235}$ ,  $Cf^{252}$  and the accidentals were made simultaneously.

The experimental arrangement is illustrated in Fig. 1. Two fission pulse ionization chambers were placed in a common container. In each of the chambers the fissionable material was spread over nine foils having a diameter of 6.5 cm. The  $U^{235}$  chamber contained  $\sim 80$  mg of material while the  $Cf^{252}$  chamber contained enough material to give  $\sim 300$  fissions per minute. The two chambers were separated by a bare foil and 2 cm of the counter gas. They were operated as flow counters using a 95% argon-5% carbon dioxide mixture. The fission counters were placed in a large water-filled shield tank and a beam of neutrons from a  $Li^7(p, n)Be^7$  source on an electrostatic accelerator was passed through the collimator into the counters. A hollow cylinder of polyethylene, 40.6 cm long with an inside diameter of 8.6 cm and an outside diameter of 20.3 cm, was placed around the fission chambers. Paraffin was added outside the tube containing the collimator and counter assembly bringing the maximum diameter to 29.5 cm (Fig. 1). The entire assembly was surrounded by a layer of cadmium. The fission neutrons were detected in twelve

<sup>†</sup> This work supported by the U. S. Atomic Energy Commission.

<sup>1</sup> R. B. Leachman, Phys. Rev. **101**, 1005 (1956).

<sup>2</sup> James Terrell, Phys. Rev. **108**, 783 (1957).

<sup>3</sup> I. I. Bondarenko, B. D. Kuzminov, L. S. Kutsayeva, L. I. Proklorova, and G. N. Smirenkin, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 353.

<sup>4</sup> A. B. Smith (private communication); Argonne National Laboratory Report ANL-5800, 2nd ed. (to be published). This survey of  $\bar{\nu}$  values includes data available prior to May, 1961.

<sup>5</sup> G. R. Keepin, T. F. Wimett, and R. K. Zeigler, Phys. Rev. **107**, 1044 (1957).

<sup>6</sup> A compilation of experimental values of  $\bar{\nu}$  has been prepared by R. B. Leachman, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 229.

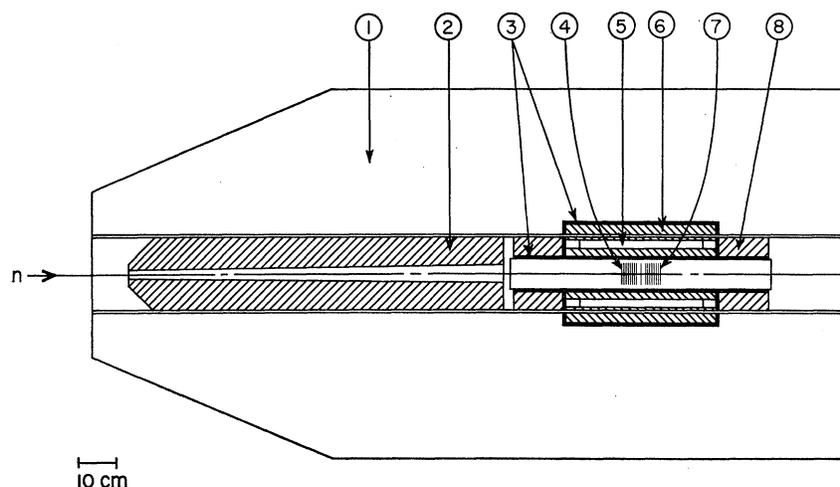


FIG. 1. Schematic diagram of the fission counter, neutron detector, and shield. (1) H<sub>2</sub>O shield. (2) Collimator. (3) Cd shield. (4) Cf<sup>252</sup> fission counter. (5) B<sup>10</sup>F<sub>3</sub> counter. (6) CH<sub>2</sub>. (7) U<sup>235</sup> fission counter. (8) Li-CH<sub>2</sub> shield.

B<sup>10</sup>F<sub>3</sub> proportional counters embedded in the polyethylene on a 16.7-cm diam circle. The efficiency for fission neutrons was 4.9% based on a  $\bar{\nu}_p$  of 2.41 for thermal neutron fission of U<sup>235</sup>.

Figure 2 shows a block diagram of the electronic equipment. A fission pulse passed through the gate, was registered in the corresponding scaler, triggered the time analyzer, and selected the appropriate subgroup of the 400-channel memory where the time distribution of neutrons detected in the 800- $\mu$ sec interval after the fission was recorded. The gates in both fission channels were closed until the time analyzer had completed its sweep. Any neutrons associated with fissions occurring during this interval entered into the random background. Typical time distributions are illustrated in Fig. 3.

The fission neutrons were on top of a constant background caused primarily by scattering of neutrons from the materials in the fission counter. The time analyzer was used to insure that the fission neutrons could be successfully separated from this background. Care was taken to minimize the background by mounting the fissionable material on very thin gold foil, partially shielding the aluminum end windows from the neutron detector, and carefully aligning the counter so the incident beam did not strike the foil supports.

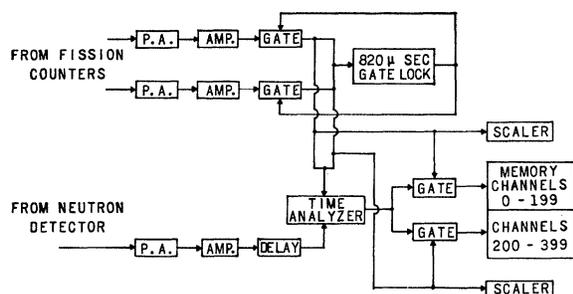


FIG. 2. Block diagram of the electronic equipment.

The discriminators on the fission counter amplifiers were adjusted to accept  $\sim 90\%$  of the fission pulses. Background in these counters was caused primarily by external electrical noise and alpha pile-up. In the U<sup>235</sup> counter the background was  $\sim 0.1\%$ , but was less in the Cf<sup>252</sup> counter since the fission rate was higher and the alpha pile-up less.

#### ANALYSIS AND RESULTS

The experimental data gave the ratio of the number of prompt neutrons detected per U<sup>235</sup> fission to the number detected per Cf<sup>252</sup> fission. To obtain  $\bar{\nu}_p(\text{U}^{235})/\bar{\nu}_p(\text{Cf}^{252})$ , a series of six corrections listed below were made.

1. *Second neutron group.* This correction was made using the data of Bevington *et al.*<sup>7</sup> for the relative yield of the second neutron group from the Li<sup>7</sup>(*p,n*)Be<sup>7</sup> reaction.

2. *Thermal neutron-induced fission.* A number of the U<sup>235</sup> fissions were caused by thermal neutrons produced in the water and other materials, including air, surrounding the equipment. This number was measured experimentally by pulsing the beam on for 50  $\mu$ sec and then observing the time distribution of the U<sup>235</sup> fissions for 1600  $\mu$ sec. It was found that  $(9 \pm 1.5)\%$  of the fissions occurred after the beam was pulsed off with a most probable time of 300  $\mu$ sec. There was no significant variation with neutron energy. Experiments with additional shielding showed that  $\sim 70\%$  of these fissions were caused by thermal neutrons entering the counter through the rear of the shield tank.

3. *Analyzer dead time.* A small correction was made for the loss of neutron counts during the analyzer dead time because of the high instantaneous counting rates occurring immediately after a fission. Since this correction depended primarily on the difference in  $\bar{\nu}_p$  of U<sup>235</sup> and Cf<sup>252</sup>, it was also energy dependent. For a dead time

<sup>7</sup> P. R. Bevington, W. W. Rolland, and H. W. Lewis, Phys. Rev. **121**, 871 (1961).

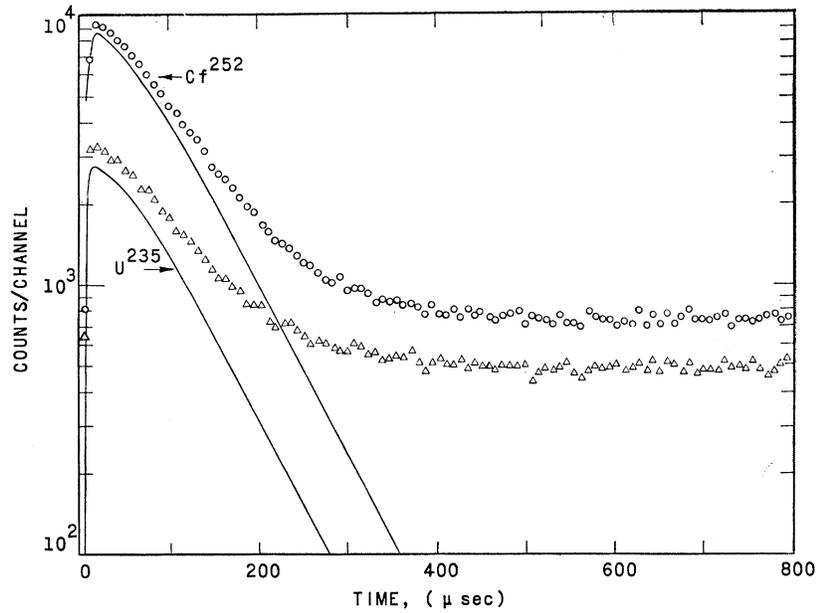


FIG. 3. Time distribution of neutrons detected after a fission pulse. The solid lines are the fission-neutron distributions.

of 40  $\mu\text{sec}$ , a neutron detector efficiency of 4.9%, and assuming  $\bar{\nu}_p$  of  $U^{235}$  and  $Cf^{252}$  to be 2.41 and 3.75, respectively, the value of  $\bar{\nu}_p(U^{235})/\bar{\nu}_p(Cf^{252})$  was decreased by 1.3%. Because of the increase of  $\bar{\nu}_p$  for  $U^{235}$  with energy, this correction resulted in a 2% increase in  $d\bar{\nu}_p/dE_n$ .

4. *Asymmetry of the neutron detector.* The detector asymmetry was measured by making traverses through it with a small Ra- $\alpha$ -Be source, then integrating over the region occupied by the fission chambers. This increased  $\bar{\nu}_p(U^{235})/\bar{\nu}_p(Cf^{252})$  by 1.4%.

5. *Energy dependence of the neutron detector.* Since the response of the neutron detector was not independent of energy, its efficiency was dependent upon the spectrum of the fission neutrons. The energy response was estimated by comparing the neutron detector with a long counter of the Hansen-McKibben type<sup>8,9</sup> using

Sb- $\gamma$ -Be, Na- $\gamma$ -Be, and Ra- $\alpha$ -Be sources. The fission neutron spectrum was represented by<sup>10</sup>

$$N(E) = aE^{\frac{1}{2}} \exp[-3E/2\bar{E}], \quad (2)$$

where  $\bar{E}$ , the average energy, was given by<sup>9</sup>

$$\bar{E} = 0.78 + 0.621(\bar{\nu} + 1)^{\frac{1}{2}}. \quad (3)$$

The resulting correction factor was

$$1 - (0.074 \pm 0.049)\Delta\bar{E}, \quad (4)$$

where  $\Delta\bar{E}$  was the difference in the average energies of the two fission neutron spectra. This correction increased  $\bar{\nu}_p(U^{235})/\bar{\nu}_p(Cf^{252})$  by  $(1.5 \pm 1.0)\%$  at  $E_n = 0$  and increased  $d\bar{\nu}_p/dE_n$  by  $(3 \pm 2)\%$ .

6. *Fission fragment angular distribution.* The neutrons from fissions induced by Mev neutrons do not have an isotropic distribution due to the angular distribution of

TABLE I. Summary of  $\bar{\nu}$  measurements.

Nuclide	$E_n$ (MeV)	$\bar{\nu}_p(U^{235})/\bar{\nu}_p(Cf^{252})$	$\bar{\nu}_p(U^{235})$	Delayed n/fission	Total $\bar{\nu}(U^{235})$	Relative error %	Total error %
$U^{235}$	(0)	(0.6440) <sup>a</sup>	(2.414) <sup>b</sup>	$0.016 \pm 0.0005^c$	(2.430) <sup>d</sup>	...	1.23
	0.03	0.6449	2.421	$0.016 \pm 0.0005$	2.437	1.04	1.61
	0.20	0.6491	2.436	$0.016 \pm 0.0005$	2.452	0.66	1.40
	0.62	0.6580	2.470	$0.016 \pm 0.0005$	2.486	0.78	1.46
	1.11	0.6713	2.520	$0.016 \pm 0.0005$	2.536	0.72	1.42
	1.58	0.6870	2.580	$0.016 \pm 0.0005$	2.596	0.76	1.45
	1.76	0.6860	2.575	$0.016 \pm 0.0005$	2.591	0.80	1.47
$Cf^{252}$	Spon.		3.754	$0.009 \pm 0.001^e$	3.763	0.30	1.27

<sup>a</sup> Calculated by a least-squares fit of a linear equation to the data.

<sup>b</sup> Normalization value.

<sup>c</sup> See reference 5.

<sup>d</sup> World average. See reference 12.

<sup>e</sup> S. Cox, P. Fields, A. Friedman, R. Sjolom, and A. Smith, Phys. Rev. **112**, 960 (1958).

<sup>8</sup> A. O. Hansen and J. L. McKibben, Phys. Rev. **72**, 673 (1947).

<sup>9</sup> W. D. Allen and A. T. G. Ferguson, Proc. Phys. Soc. (London) **A20**, 639 (1957).

<sup>10</sup> James Terrell, Phys. Rev. **113**, 537 (1959).

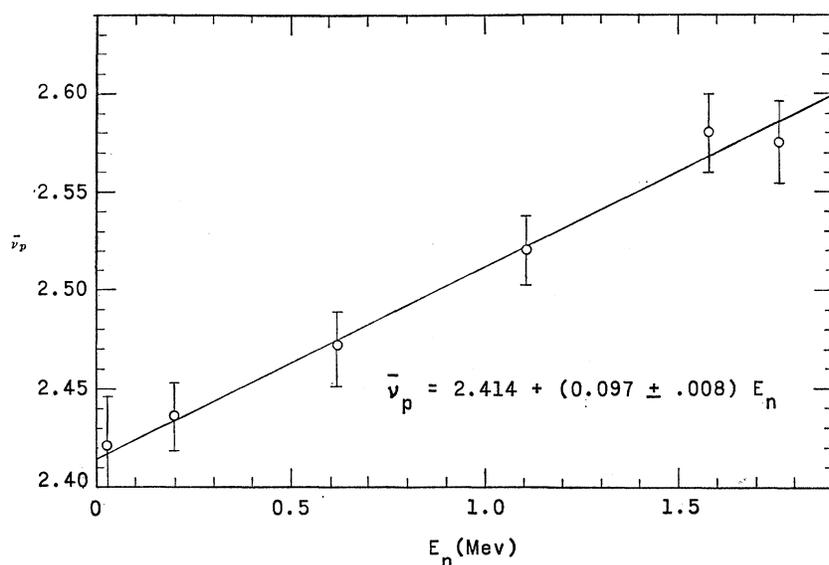


FIG. 4. Energy dependence of  $\bar{\nu}_p$  for  $U^{235}$ . Vertical bars show relative error.

the fragments.<sup>11</sup> Since the neutron detector was not spherically symmetric about the fission source, its overall efficiency depended on the angular distribution of the fission neutrons. For  $U^{235}$  this effect was small, the correction at 1.76 Mev being only  $(-0.4 \pm 0.3)\%$  relative to thermal energies. This reduced  $d\bar{\nu}_p/dE_n$  by  $(5 \pm 4)\%$ .

The corrected values of  $\bar{\nu}_p(U^{235})/\bar{\nu}_p(Cf^{252})$  are listed in column 3 of Table I. A first-order equation in  $E_n$  was fitted to these ratios by the method of least squares and the intercept was normalized to the world average<sup>12</sup> of  $\bar{\nu}$  for thermal-neutron-induced fission of  $U^{235}$  less the delayed neutrons per fission.<sup>5</sup> This normalization was used for all values of  $\bar{\nu}_p$  listed in column 4 of Table I. The energy dependence of  $\bar{\nu}_p$  for  $U^{235}$ , illustrated in Fig. 4, is given by

$$\bar{\nu}_p = 2.414 + (0.097 \pm 0.008)E_n. \quad (5)$$

#### DISCUSSION

The value of  $d\bar{\nu}_p/dE_n$  obtained here is significantly less than the  $0.136 \text{ Mev}^{-1}$  obtained by Smith<sup>4</sup> on the basis of a critical review of other data below 15 Mev and a linear fit to such data. Equation (1) is based upon the assumption that all the kinetic energy of the incident particle goes into the excitation energy of the fragments. If this is correct, then the value of  $d\bar{\nu}_p/dE_n$  obtained in this experiment requires  $E_0$  to be  $10.3 \pm 0.8$  Mev. Since the average fission neutron has about 1.2 Mev kinetic energy in the center-of-mass system,<sup>10</sup> this places the separation energy for removing the last neutron from a fission fragment at  $9.1 \pm 0.8$  Mev instead of the 6 Mev

calculated from the empirical mass formula.<sup>13</sup> Only about 70% of the kinetic energy of the incident neutron is accounted for by increased neutron emission.

At an incident neutron energy of 2 Mev the missing energy amounts to  $\sim 600$  kev. Moldauer<sup>14</sup> has pointed out that an appreciable amount of energy may be lost by gamma-ray emission before fission. The energy in the prefission gamma rays can be estimated by an approximate calculation. For electric dipole radiation the spectrum of the initial gamma ray is given by<sup>15</sup>

$$N(E_\gamma) = A(E^*)E_\gamma^3 \omega(E^* - E_\gamma), \quad (6)$$

$$\omega(E) = C \exp[2(aE)^\frac{1}{2}],$$

where  $E_\gamma$  is the gamma-ray energy,  $E^*$  is the nuclear excitation energy,  $A(E^*)$  is a normalization constant,  $\omega(E)$  is the energy level density, and  $a$  is equal to  $A/13$ .<sup>16</sup> The integrals  $I$  and  $I'$ , the probabilities that the excitation energy after the emission of the initial gamma ray is less than  $E_f$  and  $E_b$ , are given by

$$I = \int_{E^* - E_f}^{E^*} N(E_\gamma) dE_\gamma, \quad (7)$$

$$I' = \int_{E^* - E_b}^{E^*} N(E_\gamma) dE_\gamma,$$

where  $E_b$  is the neutron binding energy and  $E_f$  is the "effective" fission threshold. For the purposes of this calculation,  $E_f$  is the mean of the fission activation energy and the height of the fission barrier as given by

<sup>13</sup> N. Metropolis and G. Reitwiesner, Los Alamos Scientific Laboratory Report NP-1980, 1950 (unpublished).

<sup>14</sup> P. A. Moldauer (private communication).

<sup>15</sup> John M. Blatt and Victor F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952).

<sup>16</sup> R. D. Albert, J. D. Anderson, and C. Wong, *Phys. Rev.* **120**, 2149 (1960).

<sup>11</sup> J. E. Simmons and R. L. Henkel, *Phys. Rev.* **120**, 198 (1960).

<sup>12</sup> *Neutron Cross Sections*, compiled by D. J. Hughes and R. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed.

Vandenbosch and Seaborg.<sup>17</sup> Then by making the assumption that the relative widths for fission, gamma-ray emission, and neutron emission do not change over the energy range considered here, one obtains

$$\alpha = \frac{\Gamma_\gamma [I + (1-I')\Gamma_\gamma/\Gamma_{n,\gamma,f} + (I'-I)\Gamma_\gamma/\Gamma_{\gamma,f}]}{\Gamma_f [1 + (1-I')\Gamma_\gamma/\Gamma_{n,\gamma,f} + (I'-I)\Gamma_\gamma/\Gamma_{\gamma,f}]}, \quad (8)$$

where  $\alpha$  is the number of radiative captures per fission. The only processes considered are: (1) immediate fission or neutron emission; (2) immediate gamma-ray emission where  $E_\gamma < E^* - E_f$ ; (3) immediate gamma-ray emission followed by neutron emission or fission where  $E_\gamma < E^* - E_b$ ; (4) immediate gamma-ray emission followed by fission where  $E^* - E_b < E_\gamma < E^* - E_f$ . It can be shown that under the foregoing assumptions and for the energy range considered, the probability of  $\gamma\gamma x$  occurring is small where the sum of the gamma-ray energies is less than  $E^* - E$ .

Examination of the experimental data for fission and total nonelastic processes shows that over most of the energy range of 0 to 2 Mev the total nonelastic cross section is approximately twice the fission cross section.<sup>12</sup> Since the principal nonelastic processes are fission and inelastic neutron scattering, this indicates that the assumption of constant relative widths for fission and neutron emission is a fairly good one and also shows that  $\Gamma_f \approx \Gamma_n$ . The assumption of a constant relative width for gamma-ray emission can be verified in part by calculating the energy dependence of  $\alpha$  and comparing it with experimental measurements. Using 5.8 Mev for  $E_f$ , 6.5 Mev for neutron binding energy,<sup>17</sup> and setting  $\alpha$  equal to 0.19 at  $E_n = 0$ , then 0.13 is obtained for  $\alpha$  at  $E_n = 1$  Mev. This must be compared to the

<sup>17</sup> R. Vandenbosch and G. T. Seaborg, Phys. Rev. **110**, 507 (1958).

experimental value of 0.08 at this energy.<sup>18</sup> The agreement is only qualitative. It is evident that  $\Gamma_\gamma/\Gamma_f$  is decreasing with increasing energy as would be expected.

The average energy carried off by prefission gamma rays is given by

$$\bar{E}_{\gamma,f} = \left[ 1 + (1-I') \frac{\Gamma_\gamma}{\Gamma_{n,\gamma,f}} + (I'-I) \frac{\Gamma_\gamma}{\Gamma_{\gamma,f}} \right]^{-1} \times \left[ \frac{\Gamma_\gamma}{\Gamma_{n,\gamma,f}} \int_0^{E^*-E_b} E_\gamma N(E_\gamma) dE_\gamma + \frac{\Gamma_\gamma}{\Gamma_{\gamma,f}} \int_{E^*-E_b}^{E^*-E_f} E_\gamma N(E_\gamma) dE_\gamma \right]. \quad (9)$$

At  $E_n = 2$  Mev,  $\bar{E}_{\gamma,f}$  is 140 kev. If it is assumed that  $\Gamma_\gamma/\Gamma_f$  decreases with increasing energy in such a way that the calculated and experimental values of  $\alpha$  agree, then  $\bar{E}_{\gamma,f}$  will decrease by at least a factor of 2.

Even considering the approximate nature of the above results, it appears that the prefission gamma-rays can account for only a part of the missing energy. Some additional energy will enter into the harder fission-neutron spectrum [Eq. (3)] but this will only amount to  $\sim 75$  kev at  $E_n = 2$  Mev. Gamma-ray emission by the fragments may account for a small amount of additional energy due to the increase in the neutron separation energy. In addition, increases in the kinetic energy of the fragments cannot be disregarded. Experiments<sup>19,20</sup> have shown no evidence of this, but it is unlikely that the small amounts of energy involved would have been detected.

<sup>18</sup> B. C. Diven, J. Terrell, and A. Hemmendinger, Phys. Rev. **109**, 144 (1958).

<sup>19</sup> J. S. Wahl, Phys. Rev. **95**, 126 (1954).

<sup>20</sup> E. Segrè and C. Weigand, Phys. Rev. **94**, 157 (1954).