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<sup>28</sup>Figure 7 in LSS gives a plot of the stopping variance  $(\Delta \rho_T)^2 / \gamma$  vs  $\epsilon$  with contour lines of constant k.

PHYSICAL REVIEW C

#### VOLUME 6, NUMBER 1

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# Range Measurements of Fission Products. II. Kinetic Energy Deficit and Width of the Energy Distributions\*

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Range measurements of fission products  $(Zr^{97}, Mo^{99}, Rh^{105}, Pd^{112}, In^{115}, Te^{132}, Ce^{143})$  in gases have been compared with the range-energy tables of Northcliffe and Schilling. A reevaluation of the kinetic energy deficit for symmetric fission has been made, using a modification of these range-energy relationships. Experimental measurements of range distributions of fission fragments stopped in gases have been used to determine the width of the kinetic energy distributions as a function of product mass. The values obtained are compared with those from other workers.

# I. INTRODUCTION

This paper is the second of two dealing with the ranges of fission fragments in gases.<sup>1</sup> In this paper the measured average ranges for eight different fission products are compared with range-energy tables of Northcliffe and Schilling.<sup>2</sup> The ratio of measured to tabulated ranges depends only slightly on the mass of the fragment. This has

enabled us to convert previously measured ranges<sup>3-12</sup> for low-yield fission products into energies. Ranges for species whose energies are well known from counter measurements<sup>13-15</sup> have been used to normalize the range-energy relationships. These range-energy curves in turn are then used to determine energies of products in near-symmetric fission from radiochemical range data. For these low-yield products only radiochemical measurements have good mass resolution. This approach has been used previously,  $^{3-10, 12}$  but was made difficult by the uncertainties in conversion of the measured ranges into energies. Results are consistent with the most recent counter experiments,  $^{13-15, 16}$  but inconsistent with earlier work.  $^{17-24}$ 

The Northcliffe-Schilling tabulation of electronic stopping powers has also been used to convert the previously determined fission-produced straggling into the width of the kinetic energy distribution produced in the fission process. The radiochemical technique makes it possible to determine the width of the kinetic energy distribution for a known product without any mass-resolution ambiguities. The width values that we have obtained seem to be  $\approx 10\%$  greater than the values of Schmitt, Neiler, and Walter.<sup>14</sup>

# Iİ. EXPERIMENTAL

The experimental measurements are described in the first paper and details may be found there.<sup>1</sup> In summary, measurements were made of the ranges and range straggling in four gases for eight different fission products from  $Cf^{252}$ . An electrostatic collection technique was used, and the distribution of radioactivity on the collecting plate could be simply related to the distribution of product ranges. The observed range distributions were corrected for various effects such as geometry, sample width, and source thickness, as described in the previous paper.

The final data are mean range, and variance of the range (straggling in range). The range variance arises from two sources. One is the variation in kinetic energy produced by the fission process itself. The other is that produced by the stopping process. In the previous paper, the stopping effect was compared with the Lindhard, Scharff, and Schi $\phi$ tt (LSS)<sup>25</sup> theory, which is an *a priori* theory based on a statistical approach to the stopping process. In this paper the mean ranges are used to obtain the kinetic energy deficit in near-symmetric fission and the range variances are used to obtain the width of kinetic energy distributions. The third section of this paper is devoted to the detailed comparison of the fission-produced straggling with previous studies of the width of the energy distribution in fission.

## III. COMPARISON OF RANGE MEASUREMENTS WITH THE SEMIEMPIRICAL RANGES OF NORTHCLIFFE AND SCHILLING

In this section a comparison is made of measured ranges and the semiempirical stopping systematics of Northcliffe and Schilling.<sup>2</sup> The Northcliffe-Schilling tables give a comprehensive listing of mean range (actually path length) of ions of Z from 1 to 103 in 24 different stopping materials. The region covered is from 0.0125 to 12.0 MeV/amu and both electronic stopping power and mean range are listed in small steps. The data on which the systematization is based are mostly the range and stopping-power data for ions of Z less then 10, and Ar, Br, and I. In the large regions where there are no experimental measurements, reasonable extrapolations and interpolations have been made. The intent of Northcliffe and Schilling is to provide a precise guide for predicting stopping properties that have not been measured. In this section a direct comparison is made between the predicted range  $R_{\rm NS}$  of Northcliffe and Schilling and the fission-product measurements from this work and the literature.

Northcliffe and Schilling<sup>2</sup> have tabulated range values for one isotope of each element in various

Observed		Stop		Ion		
product	H <sub>2</sub>	Не	Ne	Ar	$E/A^{a}$	in Ref. 2
Zr <sup>97</sup>	0.909	0.859	0.722	0.905	1.072	Y <sup>89</sup> 39
M0 <sup>99</sup>	0.897	0.857	0.718	0.961	1.051	${ m Z}{ m r}_{40}^{90}$
$\mathbf{Rh}^{105}$	0.905	0.876	0.721	0.993	0.985	$Mo_{42}^{98}$
Pd <sup>112</sup>	0.879	0.849	0.701	0.957	0.906	$Rh_{45}^{103}$
Cd <sup>115</sup>	0.875	0.843	0.706	0.947	0.874	$Ag_{47}^{107}$
$Te^{132}$	0.877	0.851	0.725	0.954	0.674	$Sb_{51}^{121}$
$Ce^{143}$	0.880	0.876	0.743	0.964	0.549	$Ba_{56}^{138}$
Sm <sup>153</sup>	0.853	0.841	0.731	0.935	0.448	$Nd_{60}^{142}$

TABLE I. Ratio of Northcliffe-Schilling ranges  $R_{\rm NS}$  to measured ranges  $\langle R \rangle$  for Cf<sup>252</sup> fragments in various gases.

<sup>a</sup> Energy per atomic mass unit E/A taken from energies in Ref. 13.



FIG. 1. The ratio  $R_{\rm NS}/\langle R \rangle$  for various fission products in various stoppers. The data are from this work. Straight lines have been fitted by hand to show the slope.

stopping media. To convert to another isotope, one must use the following prescription:

$$R_{\rm NS} = \frac{A}{A_{\rm NS}} R_{\rm tab} (E/A), \qquad (1)$$

where  $R_{tab}(E/A)$  is the range interpolated from the tables for the appropriate energy-mass ratio, and A and  $A_{NS}$  are the atomic masses of the product of interest and the tabulated ion whose Z is closest to  $Z_p$ , the most probable atomic number of the fission product before  $\beta$  decay. (The estimation of  $Z_p$  is discussed in the preceding paper.) Fission-product energies were taken from the work of Schmitt and co-workers.<sup>14,15</sup> To make the comparison of range values clearer, the ratio of  $R_{\rm NS}$  to  $\langle R \rangle$  has been taken. This ratio is displayed together with the E/A values in Table I, and as a function of product mass in Fig. 1.

This comparision has also been made for several other systems for which fission-product ranges have been measured ( $Pu^{239}$ ,  $U^{235}$ , and  $Cf^{252}$  fission). This latter comparison provides normalized range-energy relationships which can be used for evaluating the kinetic energy release in nearsymmetric fission. Since this purpose requires good accuracy, a detailed consideration of  $Z_p$  has been included in the evaluation of the factor  $R_{\rm NS}/$  $\langle R \rangle$ . There is a dependence of the value of  $R_{\rm NS}$  on  $Z_p$ . Therefore, for cases in which the  $Z_p$  is intermediate between two integers,  $R_{\rm NS}$  has been computed for both Z values, and a weighted average taken. If the  $Z_{p}$  were 56.7, for example, the  $R_{\rm NS}$ was the sum of 0.7 times the  $R_{NS}$  for 57 and 0.3 times the  $R_{\rm NS}$  for 56. The difference between two  $R_{\rm NS}$  values differing by one charge is typically about 3% in the fission-product mass region. The various quantities used in this calculation are given in Tables II–IV and the dependence of  $R_{\rm NS}/\langle R \rangle$ on product mass is shown in Fig. 2. A detailed discussion of the application of these values of  $R_{\rm NS}/\langle R \rangle$  will be found in Sec. IV A.

The span of the values of  $R_{\rm NS}/\langle R \rangle$  for different stoppers is summarized in Table V. The ratios are reasonably close to unity for light stoppers except for Ne, but the trend toward large predict-

TABLE II. Ratio of Northcliffe-Schilling ranges  $R_{NS}$  to measured ranges  $\langle R \rangle$  for U, Al, and air. Thermal-neutroninduced fission of  $U^{235}$ .

Product	Range	Zp	E/A	$R_{\rm NS}/\langle R \rangle$	f <sup>a</sup>	Product	Range	Zp	E/A	$R_{\rm NS}/\langle R \rangle$	f <sup>a</sup>
Sr <sup>89</sup> Sr <sup>90</sup>	11.55 <sup>b</sup> 11.90 <sup>b</sup>	35.66 36.06	$\begin{array}{c} 1.129 \\ 1.117 \end{array}$	$\begin{array}{c} 1.377 \\ 1.332 \end{array}$	$\begin{array}{c} 1.367 \\ 1.366 \end{array}$	Ba <sup>140</sup> Ce <sup>141</sup>	8.74 <sup>b</sup> 8.56 <sup>b</sup>	54.59 55.00	$\begin{array}{c} 0.482 \\ 0.471 \end{array}$	$\begin{array}{c} \textbf{1.309} \\ \textbf{1.322} \end{array}$	$\begin{array}{c} \textbf{1.311} \\ \textbf{1.310} \end{array}$
Sr, Y <sup>91</sup> Y <sup>91</sup>	11.54 <sup>b</sup> 11.35 <sup>b</sup>	$36.46 \\ 37.27$	$\begin{array}{c} \textbf{1.099} \\ \textbf{1.074} \end{array}$	1.364 1.375	$\begin{array}{c} 1.364 \\ 1.362 \end{array}$	$Ce^{143}$	8.42 <sup>b</sup>	55.78 56.18	0.450 0.437	1.313 1.307	1.308
$Zr^{95}$	11.36 <sup>b</sup>	38.,08	1.057	1.369	1.360	$Nd^{147}$	8.07 <sup>b</sup>	57.46	0.404	1.299	1,303
Mo <sup>99</sup>	11.36 <sup>b</sup>	38.97 39.65	1.034	1.381	1.356	Sr <sup>90</sup> T131	4.02 °	36.46	1.099	1.039	1.032
$Ru^{105}$ $Ru^{106}$	11.28 <sup>b</sup> 10.90 <sup>b</sup>	41.11 42.29	0.963	1.341 1.333	1.351	Г <sup>101</sup> Ва <sup>140</sup>	2.98 °	50.80 54.59	0.481	0.978	0.985
Те <sup>123</sup> Те <sup>132</sup>	9.75 ° 9.63 <sup>b</sup>	50.02 51.21	0.596	1.330	1.323	Mo <sup>99</sup> Ba <sup>140</sup>	$3.98^{d}$ 2.98 d	$39.65 \\ 54.59$	$1.025 \\ 0.481$	$1.025 \\ 0.978$	$\begin{array}{c} 1.023 \\ 0.975 \end{array}$
$Cs^{137}$	9.18 <sup>b</sup>	53,39	0.521	1.294	1.314						

<sup>&</sup>lt;sup>a</sup> Fitted value of  $R_{\rm NS}/\langle R \rangle$  (see Sec. III of the text).

<sup>c</sup> Mean ranges in mg/cm<sup>2</sup> of Al from Ref. 6.

<sup>d</sup> Mean ranges in mg/cm<sup>2</sup> of Al from Ref. 10.

<sup>&</sup>lt;sup>b</sup> Mean ranges in mg/cm<sup>2</sup> of U from Ref. 8.

ed ranges is pronounced for the heavy stoppers Au and U. This probably results from the fact that Northcliffe and Schilling's treatment ignores the differences between the fragment's projected range and its path length. This difference is caused by multiple scattering as the ion travels through the medium. Because of repeated scattering, the measured range will always be shorter than the actual distance traveled. This projected range effect is negligible for fission fragments in light gases, but for heavy stoppers like gold and uranium, the effect becomes pronounced.<sup>2,6,8</sup>

For each stopping material there seems to be very little dependence of the ratio of  $R_{\rm NS}/\langle R \rangle$  on the fragment mass as shown in Figs. 1 and 2. In air there is no difference between the light and heavy fragments, and in the other gases, no mass effect is readily visible from the available measurements. For both U and Al a straight-line fit of  $R_{\rm NS}/\langle R \rangle$  vs A gives a small negative slope, the ratios being slightly lower for the heavy fragments. These slopes may reflect a slight energy dependence of the factor  $R_{\rm NS}/\langle R \rangle$ .

In general, the correlation observed between the experimental data and the table of Northcliffe and Schilling<sup>2</sup> seems to be quite good. Since  $R_{\rm NS}/\langle R \rangle$  is only weakly dependent on product mass, it should be possible to interpolate this ratio for other products and, with less accuracy, to extrapolate it. Because  $R_{\rm NS}/\langle R \rangle$  is close to unity, these ratios can be used to normalize the tabulated values of  $R_{\rm NS}$  and  $(\partial E/\partial x)_{\rm NS}$ . This approach has been used in the following two sections for two

TABLE III. Northcliffe ranges over measured ranges,  $R_{\rm NS}/\langle R \rangle$  for fission products. Thermal-neutron-induced fission of Pu<sup>239</sup>.

Product	Range <sup>a</sup>	$Z_{p}$	E/A	$R_{\rm NS}/\langle R \rangle$	f <sup>b</sup>
Sr <sup>91</sup>	3.125	36.76	1.128	1.035	1.036
Y <sup>92</sup>	3.125	37.16	1.115	1.034	1.036
Y <sup>93</sup>	3.099	37.57	1.103	1.035	1.036
Y <sup>94</sup>	3.081	37.97	1.091	1.036	1.036
$\mathbf{Z} \mathbf{r}^{97}$	3.064	39.17	1.058	1.034	1.036
Mo <sup>99</sup>	3.040	39.95	1.041	1.033	1.037
$\mathbf{Rh^{105}}$	2,965	42.18	0.976	1.041	1.037
$Pd^{109}$	2,880	44.05	0.876	1.015	1.037
${ m Sb}^{129}$	2,561	50.32	0.656	1.058	1.039
$Te^{132}$	2.512	51.51	0.625	1.060	1.039
$Te^{133}$	2.500	51.95	0.612	1.055	1.039
$Te^{134}$	2.500	52.42	0.600	1.048	1.039
$Ba^{140}$	2.353	54.89	0.508	1,043	1.040
$Ce^{143}$	2.315	56.08	0.476	1,037	1.040
$Pm^{149}$	2.230	58.59	0.395	1.001	1.041

<sup>a</sup> Mean ranges in  $mg/cm^2$  of air from Ref. 3.

<sup>b</sup> Fitted value of  $R_{\rm NS}/\langle R \rangle$  (see Sec. III of the text).

purposes. The ratio  $R_{\rm NS}/\langle R \rangle$  is used to probe the dependence of total kinetic energy release on the mass ratio in Sec. IV A; and in Sec. IV B, the stopping power, appropriately normalized, is used to convert the fission-produced straggling to variation in energy for single fragments.

#### **IV. DISCUSSION**

## A. Total Kinetic Energy Release in Near-Symmetric Fission

The objective of this section is to reevaluate the kinetic energy release for near-symmetric fission of Cf<sup>252</sup>, Pu<sup>239</sup>, and U<sup>235</sup>. Counter measurements are especially uncertain in this region because of mass-resolution problems and low counting rates; radiochemical methods are not affected by these particular difficulties. The problems that must be overcome in using radiochemical range measurements for this purpose are the determination of accurate range-energy relationships and precise mean ranges. It was shown above that the Northcliffe-Schilling range tables<sup>2</sup> are reasonably accurate for fission products. It was also shown that the ratio  $R_{\rm NS}/\langle R \rangle$  is only a slowly varying function of fragment mass. This latter fact makes it feasible to interpolate in order to arrive at a good approximation to the range-energy relationships.

TABLE IV. Northcliffe ranges over measured ranges,  $R_{\rm NS}/\langle R \rangle$  for fission products. Spontaneous fission of Cf<sup>252</sup>.

Product	Range	$Z_{p}$	E/A	$R_{\rm NS}/\langle R \rangle$	f <sup>a</sup>
M0 <sup>99</sup>	3.000 <sup>b</sup>	39.85	1.072	1.070	1.072
Ag <sup>111</sup>	$2.860^{b}$	44.60	0.920	1.059	1.067
$Ag^{112}$	2.840 <sup>b</sup>	44.90	0.905	1.059	1.067
$Cd^{115}$	$2.793^{b}$	45.86	0.883	1.073	1.066
$Te^{132}$	$2.586^{b}$	51.41	0.689	1.081	1.060
Ba <sup>139</sup>	$2.495^{b}$	54.39	0.589	1.051	1.057
Ba <sup>140</sup>	2.457 <sup>b</sup>	54.79	0.578	1.060	1.057
Ce <sup>141</sup>	2.430 <sup>b</sup>	55.19	0.566	1.065	1.056
$Ce^{143}$	2.405 <sup>b</sup>	55.98	0.548	1.064	1.056
Nd <sup>147</sup>	2.405 <sup>b</sup>	57.66	0.500	1.027	1.054
M0 <sup>99</sup>	3.85 <sup>c</sup>	39.85	1.072	1.086	1.079
Rh <sup>105</sup>	3.80 c	42.07	0.985	1.065	1.074
Ag <sup>111</sup>	3.64 <sup>c</sup>	44.60	0.920	1.078	1.069
Cd <sup>113</sup>	3.63 c	45.30	0.893	1.068	1.067
$Cd^{115}$	3.63 c	45.86	0.883	1.068	1.066
T <sup>131</sup>	3 46 <sup>c</sup>	51.01	0.692	1.024	1.052
$Te^{132}$	3 38 <sup>c</sup>	51.41	0.689	1.048	1.051
Ba <sup>140</sup>	3.03 °	54.79	0.578	1.068	1.044

<sup>a</sup> Fitted value of  $R_{\rm NS}/\langle R \rangle$  (see Sec. III of the text).

<sup>b</sup> Mean ranges in mg/cm<sup>2</sup> of air from Ref. 4.

<sup>c</sup> Mean ranges in mg/cm<sup>2</sup> of Al from Ref. 12.

The details of the normalization procedure were discussed above. The range data from Niday<sup>8</sup>; Alexander and Gazdik<sup>6</sup>; Aras, Menon, and Gordon<sup>10</sup>; Katcoff, Miskel, and Stanley<sup>3</sup>; Marsh and Miskel<sup>4</sup>; and Birgül, Ölmez, and Aras<sup>12</sup> divided into two groups: (1) products whose fission yield was higher than 1% (the "calibration" region); (2) products with lower yield for which the range-energy relationship is to be interpolated or extrapolated. The ratios  $R_{\rm NS}/\langle R \rangle$  were computed with the estimates of  $Z_p$  described in Sec. III and in Ref. 1. These values of  $R_{\rm NS}/\langle R \rangle$  vs A were then fitted with a least-squares straight line. The smoothed values of  $R_{\rm NS}/\langle R \rangle$  were then used to calculate an interpolated correction factor for each of the range measurements outside the calibration region. Then the following prescription was used to convert the measured ranges for points outside the calibration region into ranges appropriate for



FIG. 2. The ratio  $R_{\rm NS}/\langle R \rangle$  for various fission products in various stoppers. Different symbols denote different references as follows:  $\Box$ , fission fragments from  ${\rm Pu}^{239}$ in air, Ref. 3;  $\bigcirc$ , fission fragments from  $U^{235}$  in U, Ref. 8;  $\nabla$ ,  $\Delta$ , fission fragments from  $U^{235}$  in Al from Refs. 10 and 6, respectively;  $\blacksquare$ , fission fragments from Cf<sup>252</sup> in air, Ref. 4;  $\diamondsuit$ , fission fragments from Cf<sup>252</sup> in Al, Ref. 12. The straight lines have been fitted by least squares.

the table  $R'_{NS}$ :

$$R'_{\rm NS} = f\left(\frac{A_{\rm NS}}{A}\right) \langle R \rangle$$
, (2)

where f is the interpolated correction factor  $R_{\rm NS}/\langle R \rangle$ , and  $A_{\rm NS}$  and A are the mass of the tabulated ion and the mass of the product, respectively. This is the inverse of the procedure used to calculate  $R_{\rm NS}$ . The single-fragment energy,  $E_{Ki}^*$ , was then obtained by interpolation from the table, and converted to the total kinetic energy released,  $E_K^*$ , by the relation

$$E_{k}^{*} = \frac{E_{ki}^{*}}{A} (m_{i}^{*}) (A_{c} - m_{i}^{*})^{-1} A_{c}, \qquad (3)$$

where  $m_i^*$  is the primary mass of the fragment and  $A_c$  is the mass of the fissioning nucleus. The primary-fragment masses were obtained by adding the product mass and the value of  $\nu$  for each mass for each mass chain.<sup>15,26</sup>

In cases where the  $Z_p$  was between two integers, a weighted average of the total kinetic energies has been used. This weighting is parallel to that used for  $Z_p$ 's. For example, for a  $Z_p$  of 56.47 the following average would be taken:  $E_K^* = 0.47[E_K^*(57)]$  $+0.53[E_K^*(56)]$ , where  $E_K^*$  is the total kinetic energy. The whole set of calculations is summarized in Tables VI-VIII.

The total kinetic energies for all fragments are shown in Figs. 3-5. The yield curve from Refs. 14 and 15 is also displayed on the same abscissa. The curves of Schmitt and co-workers<sup>14,15</sup> from

TABLE V. Ratio of the Northcliffe-Schilling range  $R_{\rm NS}$  to the measured range  $\langle R \rangle$ .

Stopper	$R_{ m NS}/\langle R \rangle$	Fissioning nuclide	References
H <sub>2</sub>	0.85-0.91	Cf <sup>252</sup>	This work
He	0.84-0.88	Cf <sup>252</sup>	This work
Mylar	1.01 - 0.92	$U^{235}$	a
Air	0.98 - 1.07	$Cf^{252}$ , $Pu^{239}$	b
Ne	0.70 - 0.74	$Cf^{252}$	This work
Al	0.96 - 1.05	$U^{235}$	С
Al	1.02-1.09	Cf <sup>252</sup>	d
Ar	0.90-0.96	$Cf^{252}$	This work
Au	1.24 - 1.28 e	$U^{235}$	f
U	1.38 - 1.32 g	$U^{235}$	h

<sup>a</sup> Energies taken from Ref. 14; ranges from Ref. 11. <sup>b</sup> Energies taken from Refs. 14 and 15; ranges from

Refs. 4 and 3 for  $Cf^{252}$  and  $Pu^{239} + n$ , respectively. <sup>c</sup> Energies taken from Ref. 14; ranges from Refs. 6 and 10.

<sup>d</sup> Energies taken from Ref. 14; ranges from Ref. 12.

<sup>e</sup> Corrected for scattering as described in Ref. 6.

<sup>f</sup> Energies from Ref. 14; ranges from Ref. 6.

<sup>g</sup> Uncorrected for scattering as described in Ref. 8.

<sup>h</sup> Energies from Ref. 14; ranges from Ref. 8.

In the case of  $U^{235}+n$ , there are two sets of applicable range measurements. The values of Niday<sup>8</sup> were obtained with natural uranium as the stopper; they show an energy at symmetric fission of about 150 MeV. This is about 10 MeV higher than that of Milton and Fraser<sup>24</sup> and about 7 MeV lower than that of Schmitt. The range values from aluminum stoppers are more erratic (±6 MeV average deviation) but seem to be in agreement with Schmitt.

The kinetic energies from the data of Katcoff, Miskel, and Stanley<sup>3</sup> for  $Pu^{239} + n$  show no significant difference from the results of Schmitt and co-workers.<sup>15</sup> Both lead to energies of about 163 MeV at symmetric fission as opposed to 148 MeV from the experiment of Milton and Fraser.<sup>24</sup> The

TABLE VI. Parameters leading to the value of the total kinetic energy release. Thermal-neutron-induced fission of  $U^{235}$ .

Product	Range	$Z_{p}$	$ u^{a}$	f <sup>b</sup>	Total kinetic energy
Ag <sup>111</sup>	3.55 <sup>c</sup>	44.40	2.48	1.009	172.3
Cd <sup>115</sup>	3.32 <sup>c</sup>	45.65	2.04	1.004	158.3
Sn <sup>121</sup>	3.18 <sup>c</sup>	47.60	1.52	0.997	155.6
Sn <sup>125</sup>	3.21 <sup>c</sup>	48.71	0.61	0.992	159.3
Sb <sup>127</sup>	3.25 <sup>c</sup>	49.32	0.34	0.990	164.7
Ag <sup>111</sup>	$3.51^{d}$	$\begin{array}{r} 44.40 \\ 45.66 \\ 36.46 \\ 54.59 \\ 30.71 \end{array}$	2.48	1.009	169.1
Cd <sup>115</sup>	$3.33^{d}$		2.04	1.004	159.1
Sr <sup>91</sup>	$4.02^{d}$		1.38	1.032	165.1 <sup>e</sup>
Ba <sup>140</sup>	$2.98^{d}$		1.19	0.975	168.4 <sup>e</sup>
As <sup>77</sup>	$12.90^{f}$		0.62	1.379	167.1
Pd <sup>109</sup>	10.09 <sup>f</sup>	$\begin{array}{r} 43.75 \\ 44.40 \\ 44.70 \\ 45.66 \\ 48.71 \end{array}$	2.67	1.344	163.1
Ag <sup>111</sup>	9.74 <sup>f</sup>		2.48	1.342	154.3
Pd <sup>112</sup>	9.61 <sup>f</sup>		2.32	1.341	151.0
Cd <sup>115</sup>	9.52 <sup>f</sup>		2.04	1.338	151.6
Sn <sup>125</sup>	9.09 <sup>f</sup>		0.61	1.327	148.3
Sb, Te <sup>127</sup> Sn <sup>153</sup> Eu <sup>156</sup> Zr <sup>97</sup> Ru <sup>103</sup> Ba <sup>140</sup>	$9.58^{f} \\ 7.43^{f} \\ 7.10^{f} \\ 11.36^{f} \\ 11.28^{f} \\ 8.74^{f}$	$\begin{array}{r} 49.32 \\ 59.88 \\ 60.96 \\ 38.87 \\ 41.11 \\ 54.59 \end{array}$	$0.34 \\ 1.73 \\ 1.53 \\ 1.55 \\ 1.36 \\ 1.19$	$1.325 \\ 1.296 \\ 1.293 \\ 1.357 \\ 1.351 \\ 1.310$	167.0 154.5 148.5 175.8 <sup>e</sup> 182.7 <sup>e</sup> 169.7 <sup>e</sup>

<sup>a</sup> The values of  $\nu$  were taken from V. F. Apalin *et al.*, Nucl. Phys. <u>71</u>, 553 (1965); and H. R. Bowman *et al.*, Phys. Rev. <u>129</u>, 2133 (1963).

<sup>b</sup> This is the extrapolated factor needed to correct the Northcliffe ranges (see Sec. IVA of text).

<sup>c</sup> Mean ranges in mg/cm<sup>2</sup> of Al from Ref. 10.

<sup>d</sup> Mean ranges in  $mg/cm^2$  of Al from Ref. 6.

<sup>e</sup> Total kinetic energy release of this species calculated to check normalization.

<sup>f</sup> Mean ranges in  $mg/cm^2$  of U from Ref. 8.

TABLE VII. Parameters leading to the value of the total kinetic energy release. Thermal-neutron-induced fission of  $Pu^{239}$ .

Product	Range <sup>a</sup>	$Z_{p}$	ν <sup>b</sup>	f <sup>c</sup>	Total kinetic energy
Br <sup>83</sup>	3.247	33.47	0.50	1.035	158.6
$Pd^{112}$	2.745	45.01	2.90	1.037	180.6
$In^{117}$	2.549	46.65	2.30	1.038	162.4
$\mathrm{Sb}^{127}$	2.561	49.63	0.60	1.038	174.1
Eu <sup>157</sup>	2.194	61.62	3.50	1.041	194.6
$Pd^{109}$	2.878	44.06	2.20	1.037	188.8 <sup>d</sup>
$Mo^{99}$	3.039	39.96	1.35	1.036	180.2 d
I <sup>133</sup>	2,500	51.96	0.90	1.039	179.3 <sup>d</sup>
$Ce^{143}$	2.316	56.09	2.00	1.040	175.8 <sup>d</sup>

<sup>a</sup> Mean ranges in  $mg/cm^2$  of air from Ref. 3.

<sup>b</sup> The values of  $\nu$  were taken from V. F. Apalin *et al.*, Nucl. Phys. <u>71</u>, 553 (1965); and H. R. Bowman *et al.*, Phys. Rev. <u>129</u>, 2133 (1963).

<sup>c</sup> This is the extrapolated factor needed to correct the Northcliffe ranges (see Sec. IV A of text).

<sup>d</sup> Total kinetic energy release of this species calculated to check normalization.

TABLE VIII.	Parameters	leading to t	the value of the
total kinetic ene	ergy release.	Spontaneo	us fission of
$Cf^{252}$ .			

Product	Range	$Z_p$	ν <sup>a</sup>	f <sup>b</sup>	Total kinetic energy
${ m Sr}^{91}$	3.142 <sup>c</sup>	36.66	1.20	1.074	174.7
Y <sup>91</sup>	3.129 <sup>c</sup>	36.66	1.20	1.074	173.6
$Y^{93}$	3.129 <sup>c</sup>	37.47	1.30	1.073	178.2
${ m Z}{ m r}^{97}$	3.064 <sup>c</sup>	39.07	1.40	1.072	180.9
$\operatorname{Sn}^{121}$	$2.767^{\circ}$	47.80	3.40	1.063	200.8
$\mathrm{Sb}^{127}$	2.612 c	49.52	1.10	1.061	180.8
$\mathrm{Sm}^{153}$	2.327 <sup>c</sup>	60.08	2.30	1.052	192.6
Eu <sup>156</sup>	2.302 <sup>c</sup>	61.16	2.70	1.050	195.0
$\mathrm{Tb}^{161}$	2.263 <sup>c</sup>	63.06	3,10	1.049	201.8
$Ag^{111}$	2.860 c	44.60	2.50	1.067	192.7
$Ce^{143}$	2.405 <sup>d</sup>	55.98	1.80	1.055	183.5
Nd <sup>147</sup>	$2.405^{\text{d}}$	57.66	1.85	1.054	192.6
$\mathrm{Sb}^{127}$	3.310 e	49.61	1.10	1.055	179.5
$Te^{129}$	3.320 <sup>e</sup>	50.31	0.80	1.054	183.1

<sup>a</sup> The values of  $\nu$  were taken from V. F. Apalin *et al.*, Nucl. Phys. <u>71</u>, 553 (1965); and H. R. Bowman *et al.*, Phys. Rev. <u>129</u>, 2133 (1963).

<sup>b</sup> This is the extrapolated factor needed to correct the Northcliffe ranges (see Sec. IV A of text).

<sup>c</sup> Mean ranges in mg/cm<sup>2</sup> of air from Ref. 4.

<sup>d</sup> Total kinetic energy release of this species calculated to check normalization.

<sup>e</sup> Mean ranges in mg/cm<sup>2</sup> of Al from Ref. 12.



FIG. 3. Yields and total kinetic energies for fission of  $U^{235}$ . Points are from range data of Ref. 8,  $\Box$ ; Ref. 6,  $\Delta$ ; Ref. 10,  $\nabla$ ; filled points are heavy fission products, open points are light products, the dot and dash curve is from Ref. 22, the solid curves from Ref. 14.



FIG. 4. Yields and total kinetic energies for fission of  $Pu^{239}$ . Points are from range data of Ref. 3. Filled points are for heavy fission products, open points for light products. Dot and dash curve is from Ref. 22; solid curves from Ref. 15.

energies from the data of Marsh and Miskel<sup>4</sup> and from Birgül and co-workers<sup>12</sup> for  $Cf^{252}$  are also consistent with those of Schmitt, Neiler, and Walter,<sup>14</sup> and Bennett and Stein,<sup>20</sup> except for the high values for A values 121 and 147. Nevertheless, the value of the total kinetic energy at symmetry seems to be clearly larger than that reported by Stein and Whetstone (160 MeV).

For both Pu and Cf very high apparent kinetic energies are obtained for the fragments of mass >150. This may be due to systematic errors in the range (a 2% error in a range value in U for example will cause a 5.3-MeV difference at mass ratio 1.5). However, this seems unlikely as these deviations from the counter results and from the ranges of heavy fragments are very large (10– 35 MeV). It seems more likely that errors may appear in the extrapolation of the ratios  $R_{\rm NS}/\langle R \rangle$ to very large masses. Consistent with this possibility is the fact that  $R_{\rm NS}/\langle R \rangle$  for masses 147 and 149 are both significantly smaller than the smooth fit values. A similar sort of extrapolation error may exist for As<sup>77</sup> stopped in U (U<sup>235</sup> + *n*).<sup>8</sup>

To summarize, one can conclude that range measurements lead to kinetic energies at nearsymmetric fission that are essentially consistent



FIG. 5. Yields and total kinetic energies for fission of  $Cf^{252}$ . Points are from range data of Ref. 4,  $\Box$ ; Ref. 12,  $\bigcirc$ . Filled points are for heavy fission products, open points for light products. Dot and dash curve is from Ref. 20; dashed curve from Ref. 16; solid curves from Ref. 14.

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$ ho$ in $ m H_2$	ρ <sub>f</sub>	$\left(\frac{\partial E}{\partial x}\right)_{\rm NS}$ (MeV cm <sup>2</sup> /mg)	$R_{ m NS}/\langle R  angle$	σ <sub>EK</sub> (MeV)
0.047	0.040	203.452	0.909	$11.4 \pm 1.8$
0.036	0.034	208.966	0.897	$10.0 \pm 0.6$
0.037	0.039	220.920	0.904	$12.3 \pm 0.6$
0.036	0.034	237.438	0.879	$11.6 \pm 0.5$
0.036	0.036	247.728	0.875	$12.9 \pm 1.7$
0.035	0.035	264.845	0.877	$14.8 \pm 0.7$
0.033	0.027	264.120	0.880	$11.9 \pm 3.5$
0.036	0.039	249.855	0.852	$17.0 \pm 2.2$
	$\rho$ in H <sub>2</sub> 0.047 0.036 0.037 0.036 0.036 0.035 0.033 0.033	$\begin{array}{c c} \rho \text{ in } \mathrm{H_2} & \rho_{\mathrm{f}} \\ \hline 0.047 & 0.040 \\ 0.036 & 0.034 \\ 0.037 & 0.039 \\ 0.036 & 0.034 \\ 0.036 & 0.036 \\ 0.035 & 0.035 \\ 0.033 & 0.027 \\ 0.036 & 0.039 \\ \hline \end{array}$	$\begin{array}{c c} & \begin{pmatrix} \partial E \\ \partial x \end{pmatrix}_{\rm NS} \\ \rho \mbox{ in } {\rm H}_2 & \rho_{\rm f} & ({\rm MeVcm^2/mg}) \\ \hline 0.047 & 0.040 & 203.452 \\ 0.036 & 0.034 & 208.966 \\ 0.037 & 0.039 & 220.920 \\ 0.036 & 0.034 & 237.438 \\ 0.036 & 0.036 & 247.728 \\ 0.035 & 0.035 & 264.845 \\ 0.033 & 0.027 & 264.120 \\ 0.036 & 0.039 & 249.855 \\ \hline \end{array}$	$\begin{array}{c c} & \begin{pmatrix} \partial E \\ \partial x \end{pmatrix}_{\rm NS} \\ \rho \mbox{ in } {\rm H}_2 & \rho_{\rm f} & ({\rm MeVcm^2/mg}) & R_{\rm NS}/\langle R \rangle \\ \hline 0.047 & 0.040 & 203.452 & 0.909 \\ 0.036 & 0.034 & 208.966 & 0.897 \\ 0.037 & 0.039 & 220.920 & 0.904 \\ 0.036 & 0.034 & 237.438 & 0.879 \\ 0.036 & 0.036 & 247.728 & 0.875 \\ 0.035 & 0.035 & 264.845 & 0.877 \\ 0.033 & 0.027 & 264.120 & 0.880 \\ 0.036 & 0.039 & 249.855 & 0.852 \\ \hline \end{array}$

TABLE IX. Values related to the standard deviation of the kinetic energy distribution.

with the most recent experiments of Schmitt and co-workers<sup>13-15</sup> and Bennett and Stein.<sup>20</sup> These values are, in turn, usually inconsistent with the older work.<sup>20,22,24</sup>

## B. Widths of Single-Fragment Energy Distributions

In this section the fission-produced component of the straggling parameter  $\rho_{\rm f}$  is transformed into the width of the total kinetic energy distribution  $\sigma_{\rm EK}$  for the fragments. The separation of the fission-produced straggling parameter from the experimentally determined  $\rho_{\rm corr}$  was made in the preceding paper. This approach to the determination of  $\sigma_{\rm EK}$  from  $\rho_{\rm f}$  is important because the radiochemical method enables the determination of the energy distribution of a given mass chain, with no un-



FIG. 6. The standard deviation of the energy distribution. Filled points are from heavy fragments, open points from light fragments. The solid line is taken from Schmitt, Neiler, and Walker (Ref. 14); the dash and dot line from Whetstone (Ref. 23).

certainty in the overlap of adjacent masses. A detailed comparison is made with the results of Schmitt, Nieler, and Walter,<sup>14</sup> who used a combination of the time of flight and the counter method, and with those of Whetstone<sup>23</sup> who used time of flight. A less detailed comparison is made with the older work. The Northcliffe-Schilling range tables are reasonably accurate for fission products in light gases. Therefore, after normalization by the experimental ratio  $R_{\rm NS}/\langle R \rangle$ , one can also expect quite good accuracy for the electronic stopping powers. The method to be used rests on several other assumptions. First, electronic stopping is taken as the major contributor to the total stopping power. This is consistent with the empirical estimates of Northcliffe and Schilling and with the theory of LSS. Second, the momentum variance produced by the emission of neutrons from the fragments is small and has been neglected. Third, we have assumed that the stopping power  $(\partial E/\partial x)_{NS}$  is essentially constant near the mean energy. This does not introduce major systematic errors, since according to Northcliffe and Schilling, the stopping power does not vary more than 10% over the span of energies involved.

To convert the range distribution into an energy distribution, one uses the relationship

$$\sigma'_{E} = R_{\rm NS} \left( \frac{\partial E}{\partial x} \right)_{\rm NS} \rho_{\rm f} = \frac{R_{\rm NS}}{\langle R \rangle} \left( \frac{\partial E}{\partial x} \right)_{\rm NS} \sigma_{\rm f}, \qquad (4)$$

where  $R_{\rm NS}$  is the range from the Northcliffe-Schilling table, adjusted for mass as described in Sec. III, and  $(\partial E/\partial x)_{\rm NS}$  is the electronic stopping power for H<sub>2</sub>. Since momentum must be conserved, the standard deviation in momentum for the light fragment  $\Delta p_L$  must be equal to the standard deviation in momentum for the heavy fragment  $\Delta p_H$ . Hence the total rms width  $\sigma_{\rm EK}$  will be

$$\sigma_{EK} = \frac{(\Delta p_L)^2}{2m_L^*} + \frac{(\Delta p_H)^2}{2(252 - m_L^*)} = \frac{\Delta p^2(252)}{2m^*(252 - m^*)}, \quad (5)$$

where  $m^*$  is the mass of the primary fragment (before neutron emission).<sup>27</sup>

	$\sigma_{EK}$	Remarks	Reference	Date
Cf <sup>252</sup>	7.4	At mass ratio 1.07 (mass 130)	22	1958
$Cf^{252}$	8.5	At mass ratio 1.07 (mass 130)	20	1958
$Cf^{252}$	12.0	At mass ratio 1.07 (mass 130)	23	1963
$Cf^{252}$	13.9	At mass ratio 1.07 (mass 130)	14	1966
$U^{235} + n$	8.8	At mass 97	a	1956
$U^{235} + n$	8.70	At mass 97	24	1962
$U^{235} + n$	8.70	At mass 97	14	1966
$U^{233} + n$	8.80	At mass 97	24	1962
$Pu^{239} + n$	9.18	At mass 97	24	1962
$Pu^{239} + n$	9.0	At mass 97	15	1966
$Cf^{252}$	8.5	At mass 97	20	1958
$Cf^{252}$	10.	At mass 97	20	1963
$Cf^{252}$	9.8	At mass 97	23	1966
$Cf^{252}$	11.	At mass 97	This work	1970

TABLE X. Comparison of the widths of the kinetic energy distributions from various sources.

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Since the square of the momentum variance  $(\Delta p)^2$  is related to the standard deviation of the energy  $\sigma_{EK}$  [by  $(\Delta p)^2 = 2m * \sigma'_E$ ], the above expression reduces to

$$\sigma_{EK} = \frac{\sigma'_E(252)}{(252 - m^*)} \,. \tag{6}$$

The mass of the fragment before neutron emission  $m^*$  is related to the post-neutron mass by the relation

$$m_i^* = m_i + \nu, \tag{7}$$

where  $\nu$  is the average number of neutrons emitted by the primary fragment. From Eqs. (4) and (6) the value of  $\sigma_{EK}$  can be obtained from each measured value of  $\sigma_{f}$ .

The results for the standard deviation of the energy distribution,  $\sigma_{\rm EK},$  are given in Table IX. Values are also given for  $(\partial E/\partial x)_{\rm NS}$  the stopping power, the fission-produced straggling  $\rho_{\rm f}$  (from the first paper of this series), and the normalization factor for the stopping power,  $R_{\rm NS}/\langle R \rangle$  (computed in Sec. III). The results for the energy straggling have been plotted in Fig. 6 together with the same quantity measured by Schmitt, Neiler, and Walter<sup>14</sup> and Whetstone.<sup>23</sup> A comparison of the energy straggling with values from earlier work is given in Table X. The reported values of  $\sigma_{EK}$  have increased over the years, as the resolution corrections have improved. The values of  $\sigma_{EK}$  in this work are all about 10% higher than the results of Schmitt, Neiler, and Walter.<sup>14</sup> This difference is of the order of the expected uncertainties in this work. Since neither Schmitt and co-workers, nor Whetstone estimate their uncertainties, it is possible that all the results agree within experimental errors.<sup>28</sup> Although the results are not conclusive, it appears that the

present work is more in accord with a gradual rise of  $\sigma_{EK}$  for near-symmetric masses (reported by Schmitt, Neiler, and Walter<sup>14</sup>) rather than a very steep rise (reported by Whetstone<sup>23</sup>).

#### V. SUMMARY

We have compared our experimentally determined mean ranges with the compilation of Northcliffe and Schilling.<sup>2</sup> There are no marked effects of product mass on the ratio between experimentally determined ranges and predicted ranges. The apparent superiority of the Northcliffe rangeenergy relationship has led us to use it to reexamine the range data of previous workers. By "normalization" of the Northcliffe range-energy formulation with products whose energies are well known, we conclude that the radiochemical range data indicate a rather small deficit of total kinetic energy release for near-symmetric fission. This is consistent with more-recent data obtained by other methods, but inconsistent with older results.

The Northcliffe-Schilling compilation of stopping powers has been used to convert experimental range stragglings to kinetic energy variances as a function of fragment mass. These data show slightly higher energy variances than previous work.

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<sup>27</sup>The mean change in momentum will be zero if the neutrons are emitted isotropically, but the contribution to the rms width of the energy distribution will be nonzero. If the direction of travel is the x direction and  $\phi$ is the angle of neutron emission with respect to this direction, the net addition of the  $\sigma_{EK}^2$  is given by

$$\Delta p_x^2 = \frac{\int_0^{\pi} (p_n \cos \phi)^2 \sin \phi d\phi}{\int_0^{\pi} \sin \phi d\phi} = \frac{\overline{p_n^2}}{3},$$

where  $p_n$  is the momentum of an individual neutron. The quantity  $\overline{p}_n^2$  can be calculated, since

 $\overline{p}_n^2 = 2M_n E_n$ ,

where  $M_n$  is the mass of the neutron and  $E_n$  its energy. The contributions to the quantity  $\overline{p}_n^2$  by successive neutron emissions will be

 $\overline{p_n}^2 = 2M_n E_n \nu$ .

<sup>28</sup>One major test of internal consistency of the radiochemical measurements is the matching of the standard deviation of the energy distributions for complementary heavy and light fragments. This test has been applied to the data from this work and the internal consistency is excellent. The same test applied to the results of Petrzhak *et al.* gives a discrepancy of  $\approx 25\%$ .