

Mass number and prompt neutron emission of individual fission fragments as functions of nuclear charge, both involving parameters determinable from radiochemical data

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We lack an equation relating fission fragment mass before prompt neutron emission to the mass of the resulting fission product. It is shown that by using conveniently defined auxiliary functions and partly neglecting fine structure effects, expressions may be derived for mass number, charge density, and prompt neutron yields of individual fission fragments. All expressions involve parameters which can be evaluated from radiochemical fission product yield data, without recourse to any physical measurement whatsoever. The expressions for neutron yields from individual fragments reproduce the well-known saw-tooth curve. The fragment mass number as a function of charge is composed of two parallel straight lines with a simple discontinuity at symmetric charge division. Similarly, the fragment charge density versus charge has two branches extending in the heavy and light fragment regions, respectively. The corresponding relationship is a homographic function of charge, and is discontinuous at symmetric charge division, where Dirichlet's theorem applies. In the fission of ^{238}U , the two branches come closer together at symmetric charge division as excitation energy of the fissioning nucleus increases. The expressions mentioned above have been applied to nine different low excitation energy (≤ 14 MeV) fission processes for which selected recommended data are available. Comparison is made with published data wherever available; in general, good agreement is observed. The expression predicted by the liquid drop model for mass asymmetry of fission is shown to be identically valid for charge and neutron asymmetry also. Two new identities are also reported. In addition, two quantities are defined, namely, *the inverse charge density* with respect to nucleons and that with respect to neutrons. It is shown that the arithmetic mean of either of these quantities for the average light and heavy fragments equals the corresponding quantity for the fissioning nucleus, and that this equality holds true with notable accuracy in all low-energy fission processes considered.

[NUCLEAR REACTIONS, FISSION ^{232}Th , ^{233}U , ^{235}U , ^{238}U , ^{239}Pu ,
 ^{252}Cf . Expressions for fragment mass, charge density, and neutron
yields versus charge involving parameters determinable from radiochem-
ical yields.]

INTRODUCTION

First experimental determinations of prompt neutrons emitted from individual fission fragments were carried out by Fraser and Milton¹ who used physical techniques for determining both preneutron emission fragment masses and neutron yields. However, direct measurements of neutron yields suffer from uncertainty in the corrections for angular correlation of neutrons. By using radiochemical mass-yield data along with fragment masses determined by time-of-flight techniques, Terrell² was able

to give refined information about neutron yields from individual fragments as a function of fragment mass. Similarly, Wahl³ uses both radiochemical and physical data to infer the number of neutrons emitted by individual fragments for thermal fission of ^{235}U . Thus, so far, the fact that preneutron emission fragment masses cannot be deduced from radiochemical results has generally been taken for granted. Consequently, an equation relating fragment mass and the mass of the resulting fission product is, at present, lacking. It is the purpose of the present paper to show that preneutron and post-

neutron emission masses could be related solely through parameters determinable from radiochemical data. Certainly, this cannot be achieved rigorously, but a smooth linear relationship between fragment mass and charge can be derived from radiochemical results, and this would be valid if secondary effects for some particular A and Z values corresponding to strong shells could be partly neglected. Fortunately, these particular values are very few, so that the linear mass versus charge relationship reproduces satisfactorily all the information that time-of-flight measurements for fragment masses would provide, including the well-known saw-tooth curve for neutron yields.

CALCULATIONS

Experimental parameters. The charge densities $\bar{\delta}_H$ and $\bar{\delta}_L$ of the average fragments are used as auxiliary parameters which give concise and symmetric formulas. These are given by

$$\bar{\delta}_H = \frac{\bar{Z}_H}{\bar{M}_H} \quad (1a)$$

and

$$\bar{\delta}_L = \frac{\bar{Z}_L}{\bar{M}_L}, \quad (1b)$$

where \bar{Z}_H and \bar{M}_H are the charge and mass number of the average heavy fission fragment, and \bar{Z}_L and \bar{M}_L are the corresponding quantities for the light fragment, respectively. We have

$$\bar{M}_H = \bar{A}_H + \bar{\nu}_H$$

and

$$\bar{M}_L = \bar{A}_L + \bar{\nu}_L,$$

where \bar{A}_H and \bar{A}_L are the mass numbers of the average heavy and light products and $\bar{\nu}_H$ and $\bar{\nu}_L$ are the average numbers of emitted neutrons.

It may be assumed that the heavy and light fragments emit approximately equal numbers of neutrons on the average.²

$$\bar{\nu}_H = \bar{\nu}_L = \frac{\bar{\nu}_H + \bar{\nu}_L}{2} = \frac{\bar{\nu}}{2}.$$

Accordingly, Eq. (1a) may be written as

$$\bar{\delta}_H = \frac{\bar{Z}_H}{\bar{A}_H + \frac{\bar{\nu}}{2}}.$$

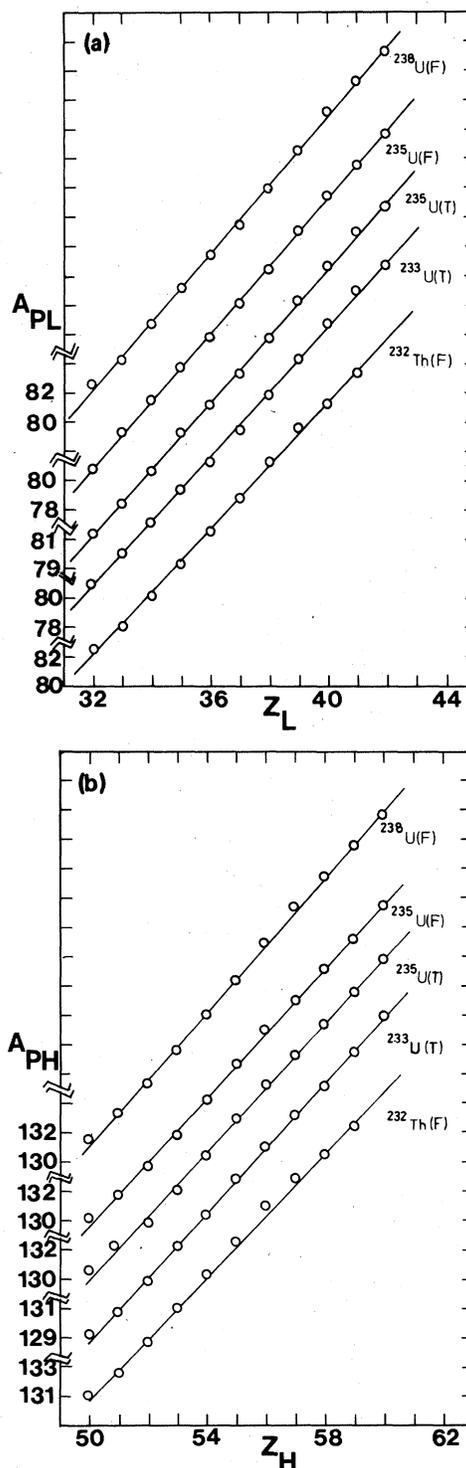


FIG. 1(a). The most probable light product mass numbers versus charge in thermal-neutron-induced fission of ^{233}U and ^{235}U , and fission-spectrum-neutron-induced fission of ^{232}Th , ^{235}U , and ^{238}U . (b) The most probable heavy product mass numbers versus charge in thermal-neutron-induced fission of ^{233}U and ^{235}U , and fission-spectrum-neutron-induced fission of ^{232}Th , ^{235}U , and ^{238}U .

In writing Eq. (1b) explicit use will be made of the conservation condition $\bar{A}_H + \bar{A}_L + \bar{\nu} = A$ and of the equality $\bar{Z}_H + \bar{Z}_L = Z$:

$$\bar{\delta}_L = \frac{Z - \bar{Z}_H}{A - \bar{A}_H - \frac{\bar{\nu}}{2}}$$

The experimental parameters to be used in the calculations are essentially $\bar{\nu}$, \bar{Z}_H , and \bar{A}_H , where Z_H is obtained from recommended yield data⁴⁻⁶ by the formula

$$\bar{Z}_H = \frac{\sum_{Z_H} Z_H y_A(Z_H)}{\sum_{Z_H} y_A(Z_H)}$$

\bar{A}_H could be calculated similarly; however, the same information will be used indirectly through the linear relationships

$$A_{PH} = a_H Z_H + b_H \quad (2a)$$

and

$$A_{PL} = a_L Z_L + b_L \quad (2b)$$

between the most probable mass numbers of the heavy and light isotopic products A_{PH}, A_{PL} , and the charge numbers Z_H, Z_L for a given mode of charge division [Figs. 1(a) and 1(b)]. We believe that this procedure provides more reliable values for \bar{A}_H . Detailed information concerning determination of the most probable product mass numbers which play an important part in the calculations is given elsewhere.⁷ Having found \bar{Z}_H as above we calculate

\bar{A}_H by means of Eq. (2a):

$$\bar{A}_H = a_H \bar{Z}_H + b_H$$

The numerical values of the fundamental parameters used throughout are given in Tables I–V.

Fragment mass versus charge. We start by writing for the mass numbers M_H and M_L of the two fragments the expressions

$$M_H = \frac{Z_H}{\delta_H} + \Delta v_H \quad (3a)$$

and

$$M_L = \frac{Z_L}{\delta_L} + \Delta v_L \quad (3b)$$

where Δv_H and Δv_L are corrective terms introduced to account for the fact that the average densities $\bar{\delta}_H$ and $\bar{\delta}_L$ have been written instead of the actual charge densities δ_H and δ_L . Adding Eqs. (3a) and (3b) side by side and using the relations $M_H + M_L = A$ and $Z_H + Z_L = Z$ we obtain

$$\begin{aligned} \Delta v_H + \Delta v_L &= A - \frac{Z}{\bar{\delta}_L} - \left[\frac{1}{\bar{\delta}_H} - \frac{1}{\bar{\delta}_L} \right] Z_H \\ &= A - \frac{Z}{\bar{\delta}_H} + \left[\frac{1}{\bar{\delta}_H} - \frac{1}{\bar{\delta}_L} \right] Z_L \end{aligned} \quad (4)$$

Two identities. The following relationships are two new identities applicable to any mode of fission

TABLE I. The most probable product mass number as a function of charge for light and heavy groups in thermal-neutron-induced fission of ²³³U, ²³⁵U, and ²³⁹Pu.

²³³ U				²³⁵ U				²³⁹ Pu			
Z _L	A _{PL}	Z _H	A _{PH}	Z _L	A _{PL}	Z _H	A _{PH}	Z _L	A _{PL}	Z _H	A _{PH}
32	80.82	60	151.00	32	81.43	60	151.84	33	82.07	61	154.54
33	83.08	59	148.60	33	83.42	59	149.60	34	84.68	60	152.11
34	85.11	58	146.34	34	85.74	58	147.46	35	87.39	59	149.92
35	87.46	57	144.20	35	88.25	57	145.32	36	89.57	58	147.65
36	89.31	56	142.12	36	90.23	56	143.29	37	91.93	57	145.28
37	91.51	55	139.88	37	92.35	55	141.00	38	94.39	56	142.67
38	93.81	54	137.52	38	94.75	54	138.52	39	96.84	55	140.45
39	96.31	53	135.25	39	97.29	53	136.11	40	99.44	54	137.68
40	98.71	52	132.83	40	99.71	52	133.89	41	101.91	53	135.30
41	100.95	51	130.86	41	101.90	51	132.24	42	104.18	52	133.43
42	102.80	50	129.25	42	103.70	50	130.49	43	106.38	51	131.58
43	105.80	49	126.20	43	105.36	49	128.97	44	107.55	50	129.67
44	109.20	48	122.70	44	109.70	48	123.50	45	110.69	49	126.70
45	112.70	47	118.40	45	112.79	47	118.25	46	113.99	48	121.50

TABLE II. The most probable product mass number as a function of charge for light and heavy groups in fission-spectrum-neutron-induced fission of ^{232}Th , ^{235}U , and ^{238}U .

^{232}Th				^{235}U				^{238}U			
Z_L	A_{PL}	Z_H	A_{PH}	Z_L	A_{PL}	Z_H	A_{PH}	Z_L	A_{PL}	Z_H	A_{PH}
31	80.32	59	149.47	32	80.96	60	151.56	32	82.62	60	153.72
32	82.46	58	147.58	33	83.31	59	149.32	33	84.34	59	151.63
33	84.09	57	145.98	34	85.56	58	147.20	34	86.71	58	149.55
34	86.13	56	144.02	35	87.79	57	145.08	35	89.20	57	147.39
35	88.31	55	141.59	36	89.79	56	143.08	36	91.48	56	144.95
36	90.47	54	139.39	37	92.17	55	140.76	37	93.56	55	142.44
37	92.81	53	137.03	38	94.47	54	138.32	38	95.93	54	140.06
38	95.24	52	134.66	39	97.11	53	135.87	39	98.69	53	137.79
39	97.65	51	132.63	40	99.55	52	133.65	40	101.25	52	135.26
40	99.24	50	131.10	41	101.72	51	131.81	41	103.38	51	133.39
41	101.38	49	128.82	42	103.70	50	130.18	42	105.27	50	131.51

in any *binary* fission process:

$$A - \frac{Z}{\delta_L} = \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] Z_H$$

and

$$A - \frac{Z}{\delta_H} = - \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] Z_L.$$

They are proved by direct substitution and simplification.

Now, let us apply these identities to the particular fictive mode of fission which leads to the average heavy and light complementary fission fragments. The resulting equalities, namely,

$$A - \frac{Z}{\delta_L} = \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_H$$

and

$$A - \frac{Z}{\delta_H} = - \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_L,$$

permit Eqs. (4) to be reduced to

$$\begin{aligned} \Delta v_H + \Delta v_L &= \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] (\bar{Z}_H - Z_H) \\ &= \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] (Z_L - \bar{Z}_L). \end{aligned} \quad (5)$$

TABLE III. The most probable product mass number as a function of charge for light and heavy groups in 14 MeV-neutron-induced fission of ^{235}U and ^{238}U , and spontaneous fission of ^{252}Cf .

Z_L	^{235}U (14 MeV)			Z_L	^{238}U (14 MeV)			Z_L	^{252}Cf (spontaneous)		
	A_{PL}	Z_H	A_{PH}		A_{PL}	Z_H	A_{PH}		A_{PL}	Z_H	A_{PH}
33	83.99	59	148.75	32	82.08	60	152.36	35	89.17	63	159.06
34	86.06	58	146.69	33	84.04	59	150.12	36	91.29	62	156.79
35	88.33	57	144.71	34	86.10	58	147.83	37	93.66	61	154.49
36		56	142.26	35	88.65	57	145.46	38	96.05	60	152.14
37	92.47	55	139.96	36	90.95	56	143.25	39	98.45	59	149.84
38	95.16	54	137.06	37	93.40	55	140.86	40	100.71	58	147.39
39	97.57	53	134.58	38	95.73	54	138.17	41	103.11	57	145.14
40	99.82	52	132.86	39	98.33	53		42	105.37	56	142.83
41	102.11	51	130.62	40	100.85	52	133.75	43	107.62	55	140.49
42	104.25	50	128.46	41		51	131.87	44	109.85	54	138.23
43	108.01	49	124.69	42	105.38	50	129.77	45	112.23	53	135.75
				43	107.69	49	127.82	46	114.54	52	133.64

TABLE IV. Parameters of the linear expressions $A_{PL} = a_L Z_L + b_L$ and $A_{PH} = a_H Z_H + b_H$ for the most probable product mass numbers.

Fissioning system	a_L	a_H	b_L	b_H
^{232}Th (fission spectrum neutron)	2.141	2.110	13.658	25.364
^{233}U (thermal neutron)	2.221	2.208	9.628	18.362
^{235}U (thermal neutron)	2.268	2.182	8.715	20.875
^{235}U (fission spectrum neutron)	2.296	2.189	7.423	20.238
^{235}U (14 MeV neutron)	2.277	2.305	8.649	12.994
^{238}U (fission spectrum neutron)	2.328	2.272	7.727	17.549
^{238}U (14 MeV neutron)	2.366	2.282	5.949	15.406
^{239}Pu (thermal neutron)	2.377	2.309	4.008	13.537
^{252}Cf (spontaneous)	2.316	2.323	8.065	12.761

Note that in the foregoing formulas it is assumed that the charge of the light fragment is lower than the average light fragment charge by the same amount as the heavy fragment charge is higher than the average heavy fragment charge, a requirement of complementarity of the fragments, neglecting ternary fission which is an exceedingly rare occurrence any way.

We may assume again²

$$\Delta v_H = \Delta v_L = \frac{\Delta v_H + \Delta v_L}{2}$$

Applying Eqs. (5)

$$\begin{aligned} \Delta v_H = \Delta v_L &= \frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] (\bar{Z}_H - Z_H) \\ &= \frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] (Z_L - \bar{Z}_L) . \end{aligned}$$

Accordingly, the equations for the fragment mass versus charge relationship become finally

$$M_H = \frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] Z_H + \frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_H$$

and

$$M_L = \frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] Z_L - \frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_L .$$

(6)

The fragment mass versus charge relationship, as seen, consists of two parallel straight lines extending in the heavy and the light group of fission fragments. The function is discontinuous at $Z_H = Z_L = Z/2$ (symmetric fission). At this point two different fragment masses are given by the two lines. But, since the discontinuity is a simple one the Fourier theorem (sometimes also called

TABLE V. Numerical values used in the calculations for the parameters \bar{Z}_H and \bar{v} .

Fissioning system	\bar{Z}_H	\bar{v}
^{232}Th (fission spectrum neutron)	54.30	2.5 ^a
^{233}U (thermal neutron)	54.29	2.492 ^b
^{235}U (thermal neutron)	54.05	2.416 ^b
^{235}U (fission spectrum neutron)	54.02	2.61 ^c
^{235}U (14 MeV neutron)	53.77	4.51 ^c
^{238}U (fission spectrum neutron)	53.46	3.0 ^c
^{238}U (14 MeV neutron)	53.64	4.7 ^a
^{239}Pu (thermal neutron)	54.10	2.884 ^b
^{252}Cf (spontaneous)	55.74	3.784 ^b

^aJ. P. Unik *et al.* in *Proceedings of the International Atomic Energy Agency Symposium on the Physics and Chemistry of Fission, Rochester, New York, 1973* (IAEA, Vienna, 1974) Vol. 2, p. 19.

^bJ. W. Boldeman and A. W. Dalton, *Aust. At. Energy Comm. Report AAEC/E 172*, (1967).

^cM. Lefort, *Nuclear Chemistry* (Van Nostrand, London, 1968).

Dirichlet's theorem) applies and gives for the common mass number of symmetric fragments the value

$$M_H \left(\frac{Z}{2} \right) = M_L \left(\frac{Z}{2} \right) \\ = \frac{M_H(Z/2) + M_L(Z/2)}{2} = \frac{A}{2}.$$

Applications follow:

(1). *Fragment charge density versus charge.* The charge density expressions of the actual fission fragments can now be written explicitly, using Eqs. (6) for fragment mass versus charge just obtained

$$\delta_H = \frac{Z_H}{M_H} = \frac{2Z_H}{\left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] Z_H + \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_H}, \quad (7)$$

$$\delta_L = \frac{Z_L}{M_L} = \frac{2Z_L}{\left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] Z_L - \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_L}.$$

Both are homographic functions of charge. The whole function is composed of two branches. The branch which lies in the light group is descending, the other ascending. The function is discontinuous at $Z_H = Z_L = Z/2$ (symmetric fission).

(2). *Fragment neutron yields versus charge.* The number of prompt neutrons emitted by a fission fragment is given by the difference between its mass number and that of the most probable primary product for the same Z :

$$v_H = \frac{1}{2} \left[\left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] Z_H + \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_H \right] - A_{PH}, \quad (8)$$

$$v_L = \frac{1}{2} \left[\left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] Z_L - \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_L \right] - A_{PL}.$$

In situations in which the individuality of fragments is of importance and fine structure effects should not be neglected the discrete numerical values given in Tables I–III must be used for A_{PH} and A_{PL} . This introduces indirectly fine structure effects since the discrete values calculated for A_{PH} and A_{PL} carry with them such effects through the measured fission

yields used in their calculation. In other situations the smooth values of A_{PH} and A_{PL} defined by the linear expressions (2a) and (2b) may be used. In this case Eqs. (8) become

$$v_H = \left[\frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] - a_H \right] Z_H \\ + \left[\frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_H - b_H \right], \quad (9) \\ v_L = \left[\frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] - a_L \right] Z_L \\ - \left[\frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_L + b_L \right].$$

The numerical values of the parameters a_H , b_H , a_L , and b_L for various fission processes are given in Table IV. Undoubtedly, the linear expressions (9) can only be applied within definite ranges of the variables Z_H and Z_L . The lower limits of these ranges are obtained by putting $v_H = 0$ and $v_L = 0$ in Eqs. (9), and are found to be

$$Z_{HO} = \frac{-\frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_H + b_H}{\frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] - a_H} \quad (10)$$

and

$$Z_{LO} = \frac{\frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \bar{Z}_L + b_L}{\frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] - a_L}.$$

Upper limits for Z_H and Z_L should be considered also, since the total number of emitted neutrons must not exceed $\bar{\nu}$. These upper limiting charges must also satisfy the complementarity conditions from which they can be calculated:

$$Z_{HM} = Z - Z_{LO} \quad (11)$$

and

$$Z_{LM} = Z - Z_{HO}.$$

(3). *Fission asymmetry with respect to mass, charge, and neutron contents.* These are defined by

$$\sigma_a = \frac{\bar{A}_H - \bar{A}_L}{A}, \quad (12)$$

$$\sigma_z = \frac{\bar{Z}_H - \bar{Z}_L}{Z},$$

and

$$\sigma_n = \frac{\bar{N}_H - \bar{N}_L}{N},$$

respectively.

Swiatecki⁸ has shown that the liquid drop model predicts for low energy fission a mass asymmetry expression of the form

$$\sigma_a = c_a \left[\left(\frac{Z^2}{A} \right)_a - \left(\frac{Z^2}{A} \right) \right]^{1/2}, \quad (13)$$

where the parameter Z^2/A is a measure for the ratio of Coulombic to surface energy, and $(Z^2/A)_a$ is a limiting value for this parameter below which the symmetrical saddle point shape becomes unstable against asymmetric distortions; c_a is a constant of proportionality.

The fission asymmetry may also be defined in terms of nuclear charge or neutron contents of the average products. It has been shown⁹ that in both cases these asymmetries are given by expressions similar to Eq. (13). This similarity is almost a complete analogy or identity, since the numerical values of the constants are practically the same in the three cases

$$\left(\frac{Z^2}{A} \right)_a = 40.2 \pm 0.7, \quad \left(\frac{Z^2}{A} \right)_z = 39.65,$$

$$\left(\frac{Z^2}{A} \right)_n = 39.50.$$

$$c_a = 0.090, \quad c_z = 0.089, \quad c_n = 0.103.$$

These values were obtained as slopes and intercepts of Z^2/A versus $\sigma_{a,z,or,n}$ plots, and an explanation for the analogy in question is lacking. Now, we are going to show that the expressions derived above for the fragment mass versus charge afford an explanation for the equality of the respective constants in the three cases. Let us apply Eqs. (6) to the average fragments, putting $Z_H = \bar{Z}_H$ and $Z_L = \bar{Z}_L$, and subtract side by side

$$\begin{aligned} \bar{M}_H - \bar{M}_L &= \frac{1}{2} \left[\frac{1}{\bar{\delta}_H} + \frac{1}{\bar{\delta}_L} \right] (\bar{Z}_H - \bar{Z}_L) \\ &+ \frac{1}{2} \left[\frac{1}{\bar{\delta}_H} - \frac{1}{\bar{\delta}_L} \right] Z. \end{aligned} \quad (14)$$

In accordance with the assumption that $\bar{v}_H = \bar{v}_L$, the left-hand side of Eq. (14) is simply $\bar{A}_H - \bar{A}_L = \sigma_a A$. The difference $\bar{Z}_H - \bar{Z}_L$ on the right-hand side of Eq. (14) is similarly $\sigma_z Z$, so that Eq. (14) may be written as

$$\sigma_a = \frac{1}{2} \left[\frac{1}{\bar{\delta}_H} + \frac{1}{\bar{\delta}_L} \right] \frac{Z}{A} \sigma_z + \frac{1}{2} \left[\frac{1}{\bar{\delta}_H} - \frac{1}{\bar{\delta}_L} \right] \frac{Z}{A}. \quad (15)$$

For the expressions for σ_a and σ_z to be identical, on the right of Eq. (15) the coefficient of σ_z must be unity

$$\frac{1}{2} \left[\frac{1}{\bar{\delta}_H} + \frac{1}{\bar{\delta}_L} \right] \frac{Z}{A} = 1 \quad (16)$$

and the second term negligible. Table VI shows that the coefficient in question is indeed practically unity for all fission processes considered. On the other hand, the uncertainty ± 0.7 in the parameter $(Z^2/A)_a$ causes a corresponding absolute uncertainty of 0.0304 in σ_a . Table VI shows that the numerical value of the second term is in all cases less than this, so that this term may be neglected. Thus Eq. (15) reduces drastically to $\sigma_a = \sigma_z$, signifying full analogy of the mass and charge asymmetry expressions.

Furthermore, from definitions (12) it follows that

$$N \sigma_n = A \sigma_a - Z \sigma_z.$$

Replacing σ_a by Eq. (15) and simplifying

$$\begin{aligned} \sigma_n &= \left[\frac{1}{2} \left[\frac{1}{\bar{\delta}_H} + \frac{1}{\bar{\delta}_L} \right] - 1 \right] \frac{Z}{N} \sigma_z \\ &+ \frac{1}{2} \left[\frac{1}{\bar{\delta}_H} - \frac{1}{\bar{\delta}_L} \right] \frac{Z}{N}. \end{aligned}$$

Full analogy requires, as above, for the coefficient of σ_z the condition

$$\begin{aligned} &\left[\frac{1}{2} \left[\frac{1}{\bar{\delta}_H} + \frac{1}{\bar{\delta}_L} \right] - 1 \right] \frac{Z}{N} \\ &= \left[\frac{1}{2} \left[\frac{1}{\bar{\delta}_H} + \frac{1}{\bar{\delta}_L} \right] - 1 \right] \frac{Z}{A - Z} = 1, \end{aligned} \quad (17)$$

TABLE VI. Numerical results indicating the analogy of different expressions for fission asymmetry.

Fissioning system	$\frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] \frac{Z}{A}$	$\frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \frac{Z}{A}$	$\frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] - 1$	$\frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \frac{Z}{A-Z}$	$\frac{1}{2} \left[\frac{1}{\delta_H} - \frac{1}{\delta_L} \right] \frac{Z}{A-Z}$
²³² Th (fission spectrum neutron)	0.9989	0.0055	0.9982	0.0089	0.0089
²³³ U (thermal neutron)	0.9978	0.0123	0.9963	0.0203	0.0203
²³⁵ U (thermal neutron)	0.9979	0.0120	0.9966	0.0196	0.0196
²³⁵ U (fission spectrum neutron)	0.9981	0.0107	0.9970	0.0175	0.0175
²³⁵ U (14 MeV neutron)	0.9982	0.0109	0.9969	0.0179	0.0179
²³⁸ U (fission spectrum neutron)	0.9977	0.0140	0.9963	0.0228	0.0228
²³⁸ U (14 MeV neutron)	0.9988	0.0070	0.9981	0.0114	0.0114
²³⁹ Pu (thermal neutron)	0.9977	0.0151	0.9963	0.0248	0.0248
²⁵² Cf (spontaneous)	0.9991	0.0065	0.9985	0.0107	0.0107

and for the second term on the right, namely,

$$\frac{1}{2} \left(\frac{1}{\delta_H} - \frac{1}{\delta_L} \right) \frac{Z}{A-Z}$$

to be less than 0.030. Table VI again shows that both requirements are fully satisfied. This result combined with the previous one proves that indeed the equalities $\sigma_a = \sigma_z = \sigma_n$ are correct at least within the limits imposed by the observational uncertainties.

RESULTS AND DISCUSSION

We performed the calculations for nine low-energy fission processes, namely, spontaneous ²⁵²Cf; thermal-neutron induced, ²³³U, ²³⁵U, and ²³⁹Pu; fission-spectrum-neutron induced, ²³²Th, ²³⁵U, and ²³⁸U; 14 MeV-neutron-induced, ²³⁵U and ²³⁸U.

The straight lines representing the fragment mass number as a function of charge, Eqs. (6), for thermal-neutron-induced fission of ²³⁵U are shown in Fig. 2, where some points which represent theoretical results from the Wilkins-Steinberg model^{10,11} have also been recorded. The deviations between the values calculated in this work and those given by the Wilkins-Steinberg model do not exceed 0.3%, except for the pair $Z = 56 - 36$ in which case the deviation is approximately 1%. This larger deviation stems from the deformed-strong-neutron shell at $N \sim 88$ which plays an important role in the Wilkins-Steinberg model. The most probable fragment mass for $Z = 56$ is $M_H = 144$; this corresponds to $N = 144 - 56 = 88$ neutrons. According to the model the fragments born in the $N \sim 88$

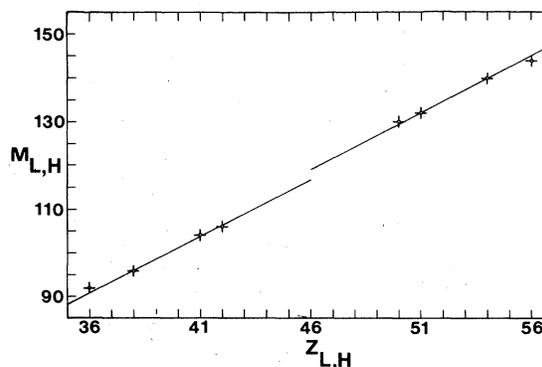


FIG. 2. Mass number versus charge for light and heavy fragments in thermal-neutron-induced fission of ²³⁵U. The points represent values deduced from the Wilkins-Steinberg model (Refs. 10 and 11).

region are highly deformed, and have a special tendency to higher neutron emission. Another strong shell (spherical in this case) is present at $Z \sim 50$ and is jointly operative with the spherical-strong-neutron shell at $N \sim 82$. The absence of deformation energy in this case prevents high neutron emission and the mass numbers predicted by the model agree well with the smooth values given by the linear relationships, Eqs. (6).

The charge density versus charge curves were calculated using Eqs. (7). There is a discontinuity at symmetric charge division which arises from the corresponding discontinuity of the fragment mass relationship mentioned above. The common density at discontinuity is obtained again by applying Fourier's theorem, and is found to be Z/A , as would be expected, i.e., equal to that of the fissioning nucleus. Figure 3 illustrates the variation of

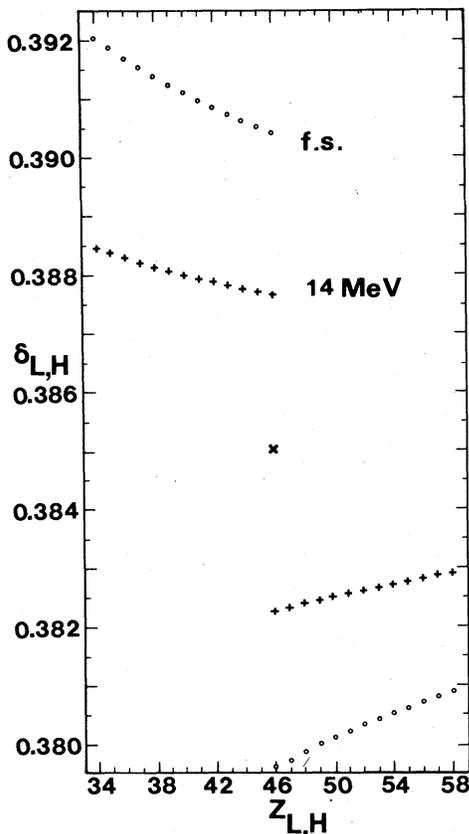


FIG. 3. Charge density versus charge in fission-spectrum-neutron-induced fission of ^{238}U (circles) and 14 MeV-neutron-induced fission of ^{238}U (crosses). Note the discontinuity at symmetric charge division $Z = 46$, where Fourier's theorem predicts the common charge density of fragments to be 0.385 for both processes, agreeing remarkably with $91/239 = 0.3849$.

charge density with charge for fission-spectrum-neutron-induced and 14 MeV-neutron-induced fission of ^{238}U . It is seen that the two branches of the curve approach closer at symmetry as the excitation energy of the fissioning nucleus increases. Similarly, in the case of ^{235}U , for 14 MeV- and fission-spectrum-neutron-induced fission the corresponding branches which lie almost inseparably as a single curve, approach to come closer together at symmetric division, leaving behind the curve for thermal-neutron-induced fission. In other words, a picture similar to the case of ^{238}U is observed, where the outer curve belongs to thermal fission while the inner curve is a double one whose components belong to 14 MeV- and to fission-spectrum-neutron fissions.

The number of neutrons emitted from the heavy and light fragments as a function of charge in spontaneous fission of ^{252}Cf was calculated from Eqs. (8). The results are compared with experimental data¹² in Table VII; a good agreement is observed. In most cases the experimental data are reported merely in terms of fragment mass, not in terms of charge number. By using Eqs. (12) as transformation equations, along with Eqs. (18) or (9), it is possible to calculate the functions ν_H vs M_H and ν_L vs M_L , and other functions as well. This was done for thermal-neutron-induced-fission processes of ^{233}U , ^{235}U , and ^{239}Pu . The discrete values of emitted neutrons deduced from Eqs. (8) and the smooth numbers implied by the straight lines, Eqs. (9), are compared with experimental results¹³ in Figs. 4–6. Notice the good agreement between the experimental points and the points corresponding to the discrete values. Note that, at

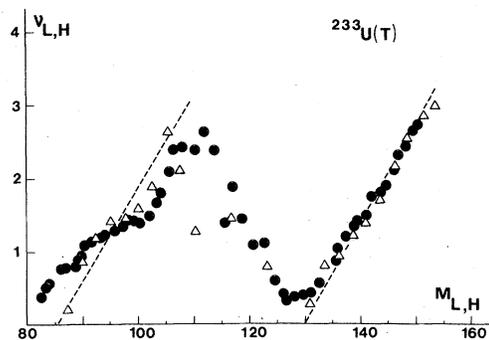


FIG. 4. Emitted neutrons versus fragment mass for thermal-neutron-induced fission of ^{233}U . Circles: experimental (Ref. 13). Triangles: this work. Straight lines: linear approximations; note the better agreement for the heavy-group line.

TABLE VII. Emitted neutrons per fragment as a function of fragment mass for light and heavy groups in spontaneous fission of ^{252}Cf .

Z_L	ν_L [expt. (Ref. 12)]	ν_L (this work)	Z_H	ν_H [expt. (Ref. 12)]	ν_H (this work)
40	1.46 ± 0.146	1.34	58	2.07 ± 0.08	2.55
41	1.61 ± 0.1	1.51	57	1.842 ± 0.05	2.24
42	1.86 ± 0.06	1.83	56	1.89 ± 0.07	1.98
43	1.93 ± 0.03	2.14	55	1.76 ± 0.035	1.75
44	2.22 ± 0.05	2.48	54	1.38 ± 0.09	1.44
45	2.33 ± 0.12	2.67	53	1.37 ± 0.06	1.34

the heavier fragment side of the light group (near $Z = 44$) the neutron yields fall down rapidly and reach the lighter fragment side of the heavy group, a fact in harmony with experimental data, reproducing the well-known saw-tooth curve. In addition, it is seen that the functions ν_H vs M_H and ν_L vs M_L are represented fairly well by two straight lines with slightly different slopes. Inspection of Table IV shows that, except in 14 MeV-neutron-induced fission of ^{238}U , $a_L > a_H$. Consequently, the slope $d\nu_L/dM_L$ for the light group is in general smaller than the corresponding slope $d\nu_H/dM_H$ for the heavy group; this is in accordance with experimental results. Wahl's saw-tooth curve for thermal fission of ^{235}U based on both radiochemical and physical measurements gives significantly higher neutron yields for the lightest and correspondingly lower neutron yields for the complementing heaviest fragments. On the other hand, the very good agreement between Wahl's $\nu = \nu_H + \nu_L$ data and our $\nu = A - (A_{PH} + P_{PL})$

values, especially near $A_{PH} \sim 130$, rules out the possibility for the difference in the results derived from physical and radiochemical measurements to be due to the use by Wahl of Terrell's method in regions of rapidly changing yields.

The neutron yields for highly symmetric division calculated for 14 MeV-neutron-induced fission differ considerably. The high neutron emission observed in this case is consistent with the known fact that the saw-tooth curve "washes out" with increasing excitation energy. A similar situation was encountered in high-energy proton-induced fission.¹⁴

The validity ranges of linear expressions for ν_H vs Z_H and ν_L vs Z_L calculated from Eqs. (10) and (11) are given in Table VIII. It is seen that the lowest charges Z_{LO} below which neutron emission ceases lie in a relatively narrow range centering at $Z = 32 - 33$; which should be attributed to the high stability of the spherical strong-neutron shell with 50 neutrons, as already noted by Terrell. The calculated values for Z_{HO} reflect a similar effect due

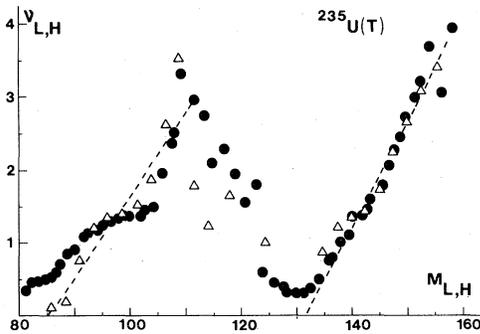


FIG. 5. Emitted neutrons versus fragment mass for thermal-neutron-induced fission of ^{235}U . Circles: experimental (Ref. 13). Triangles: this work. Straight lines: linear approximations; note the better agreement for the heavy-group line.

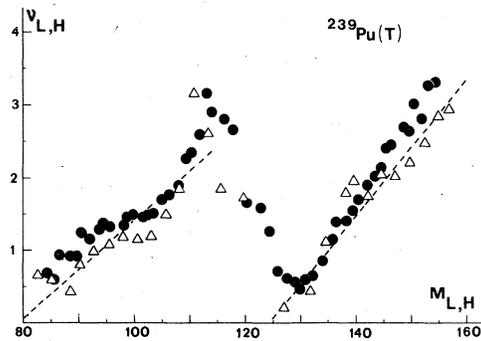


FIG. 6. Emitted neutrons versus fragment mass for thermal-neutron-induced fission of ^{239}Pu . Circles: experimental (Ref. 13). Triangles: this work. Straight lines: linear approximations; note the better agreement for the heavy-group line.

TABLE VIII. Ranges of validity of linear relationships of emitted neutrons versus fragment charge, in the processes investigated.

Fissioning system	Z_{LO}	Z_{LM}	Z_{HO}	Z_{HM}
^{232}Th (fission spectrum neutron)	31.8	38.3	51.7	58.2
^{233}U (thermal neutron)	34.1	41.5	50.5	57.9
^{235}U (thermal neutron)	33.9	41.2	50.9	58.1
^{235}U (fission spectrum neutron)	32.0	41.5	50.5	60.0
^{235}U (14 MeV neutron)	34.3			57.7
^{238}U (fission spectrum neutron)	34.6	43.2	48.8	57.4
^{238}U (14 MeV neutron)	29.1			62.9
^{239}Pu (thermal neutron)	31.5	45.9	48.1	62.4
^{252}Cf (spontaneous)	34.7	49.9	48.0	63.3

to the well-known strong-proton shell $Z = 50$, and the jointly operative neutron shell $N = 82$.

Observe that from Eq. (16) or Eq. (17) we obtain

$$\frac{1}{2} \left[\frac{1}{\delta_H} + \frac{1}{\delta_L} \right] = \frac{A}{Z}, \quad (18)$$

which means that the arithmetic mean of reciprocal charge densities of the average heavy and light fragments equals the reciprocal charge density of the fissioning nucleus. Note that the left-hand side of Eq. (18) appears in several formulas given above and that it could be replaced simply by A/Z with high

accuracy in all fission processes considered, making a remarkable reduction of the formulas possible. In addition, from Eqs. (6),

$$dM_H/dZ_H = dM_L/dZ_L = A/Z.$$

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