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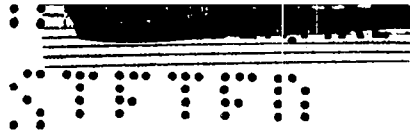
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ON THE EMISSION PROBABILITIES OF
FISSION NEUTRONS

by

R. B. Leachman

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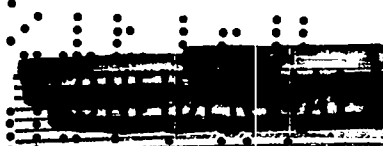
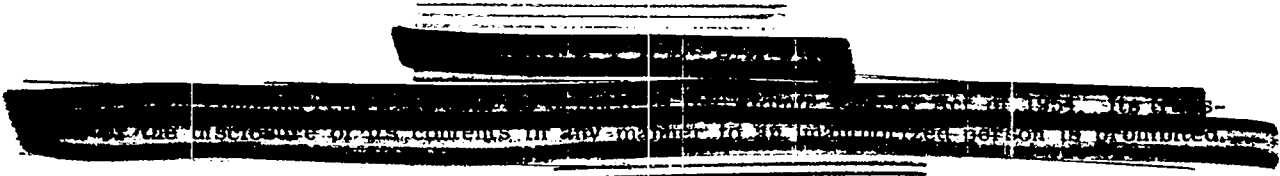
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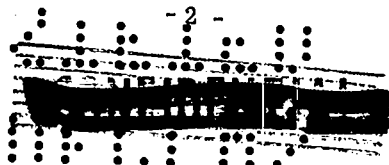
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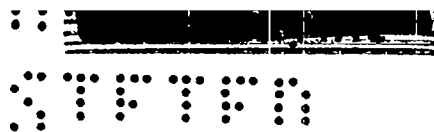
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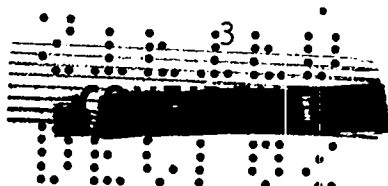


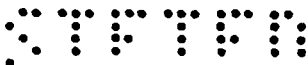
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ABSTRACT

Emission probabilities of 0, 1, 2, 3 ... prompt neutrons from fission are determined from the calculated excitation energy distributions of the fission products, together with the evaporation model of the nucleus. Shapes of these excitation energy distributions are derived from the kinetic energy distributions observed for the fragment pairs; absolute excitation energies are determined by normalizing the calculated average number of fission neutrons to measured values. Neutron emission data are calculated for thermal-neutron induced fission of U^{235} and Pu^{239} and for spontaneous fission of U^{238} , Cm^{242} , Cm^{244} , and Cf^{252} . The fragment kinetic energies resulting from the normalization are in satisfactory agreement with recent measurements and, similarly, the neutron emission probabilities are in reasonable agreement with measured values.

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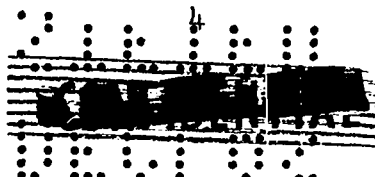
AUTHOR'S NOTE

The experimental results of neutron emission from spontaneous fission of Cm^{242} and Cm^{244} , which are now being prepared by Hicks, Ise, and Pyle at the Radiation Laboratory at Berkeley, will be included in a revision of this report as they become available in unclassified form. Similarly, the present calculations will be re-normalized to the unclassified $\bar{\nu}$ values resulting from these measurements.

ACKNOWLEDGMENTS

The author wishes to acknowledge the suggestions made by Professor C. D. Coryell on the mass surface, by Professor V. F. Weisskopf on neutron temperatures, and Professor H. H. Barschall on the use of the evaporation model. The machine calculations were made by I. J. Cherry.

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INTRODUCTION

Recent measurements of the neutron emission probabilities of spontaneous fission show wide distributions in the number of neutrons emitted in each fission event. Geiger and Rose¹ observed the ratios of the second and third moments of neutron emission to the average number of neutrons emitted for spontaneous fission of U^{238} . Hicks et al.² used a large tank containing a liquid scintillator with cadmium and measured the probabilities $P_0, P_1, P_2 \dots$ of the occurrence of 0, 1, 2... neutrons from the spontaneous fission of Cf^{252} . Their data show appreciable probabilities of emitting 1, 2, 3, and 4 neutrons per fission. A calculation of the neutron emission probabilities by the evaporation model of the nucleus and the calculated excitation energy distribution of the fission products is made in this paper for comparison with the fluctuations in the observed number of fission neutrons.

METHOD

In this section is presented an outline of the method by which the neutron emission probabilities are calculated. The details of the calculations are presented in the following sections.

To calculate the neutron emission probabilities, the distribution X^L in the excitation energy E_x^L of the light fragment and the corresponding distribution for the heavy fragment are required. In this paper the superscripts L and H are used to designate the light and

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heavy fragments, respectively, and all distributions are normalized to unity. These excitation energy distributions are obtained by first determining the distribution X in the total excitation energy E_X from the distribution K in the sum E_K of the kinetic energy of the two fission fragments and from the mass equation of fission,

$$M(A, \delta, Z) + B = M(A^L, \delta^L, Z^L) + M(A^H, \delta^H, Z^H) + E_K + E_X, \quad (1)$$

in which an atomic mass unit is 931.15 Mev and M represents the atomic mass. In Eq. 1 the mass number A of the compound nucleus undergoing fission is related to the mass number of the fragments by $A = A^L + A^H$ and the nuclear charge Z of the compound nucleus is, similarly, $Z = Z^L + Z^H$. The δ terms are the even-odd mass parameters. With the binding energy of a neutron to the compound nucleus as B , Eq. 1 thus applies to thermal-neutron induced fission. With $B = 0$, Eq. 1 applies to spontaneous fission.

The total excitation energy E_X determined from Eq. 1 is shared by the two fission products of binary fission. To determine the neutron emission probabilities, the excitation energy distributions X^L and X^H of the excitation energies E_X^L and E_X^H of the respective fragments are determined from the convolution

$$X(E_X, \delta, R_A) = \int_{-\infty}^{\infty} dE_X^L X^L(E_X^L, \delta^L, R_A) X^H(E_X - E_X^L, \delta^H, R_A), \quad (2)$$

where $R_A = A^L/A^H$. In Eq. 2, $E_X - E_X^L$ is substituted for the E_X^H parameter in X^H .

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The resulting excitation energy distributions are combined with the evaporation model of the nucleus to determine the probabilities of numbers of fission neutrons from each fragment. For simplicity, the simple neutron evaporation expression

$$n(\epsilon) \propto \epsilon \exp(\epsilon/T) \quad (3)$$

is used where $n(\epsilon)$ is the probability of emitting neutrons of energy ϵ and T is the nuclear "temperature." From the neutron emission expression (Eq. 3) are determined the neutron emission probabilities $N^L(E_X^L, \delta^L, \nu^L, R_A)$ and the similar probability for the heavy fragment, both independent of the neutron energy ϵ . The number of emitted neutrons is ν . The probabilities P_{ν}^L and P_{ν}^H of emitting ν neutrons from the light and heavy fragment, respectively, are determined by combining the excitation energy distributions X^L and X^H with the neutron emission probabilities N_{ν}^L and N_{ν}^H by

$$P_{\nu}^L(\delta^L, \nu^L, R_A) = \int_{-\infty}^{\infty} dE_X^L X^L(E_X^L, \delta^L, R_A) N_{\nu}^L(E_X^L, \delta^L, \nu^L, R_A) \quad (4)$$

and the similar expression for the heavy fragment. The δ parameters are removed by summation over the various even-odd conditions with appropriate weighting. The composite probability P_{ν} of emitting ν neutrons from the fission products of mass ratio R_A is determined from the summation

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$$P_{\nu}(\nu, R_A) = \sum_{\eta=0}^{\nu} P_{\eta}^L(\nu^L, R_A) P_{\nu-\eta}^H(\nu^H, R_A), \quad (5)$$

where η is a summation parameter analogous to ν . Finally, the combined neutron emission probabilities of all fission modes is considered by summation of each P_{ν} over all R_A , with the weighting of each case determined from the measured fission yields.

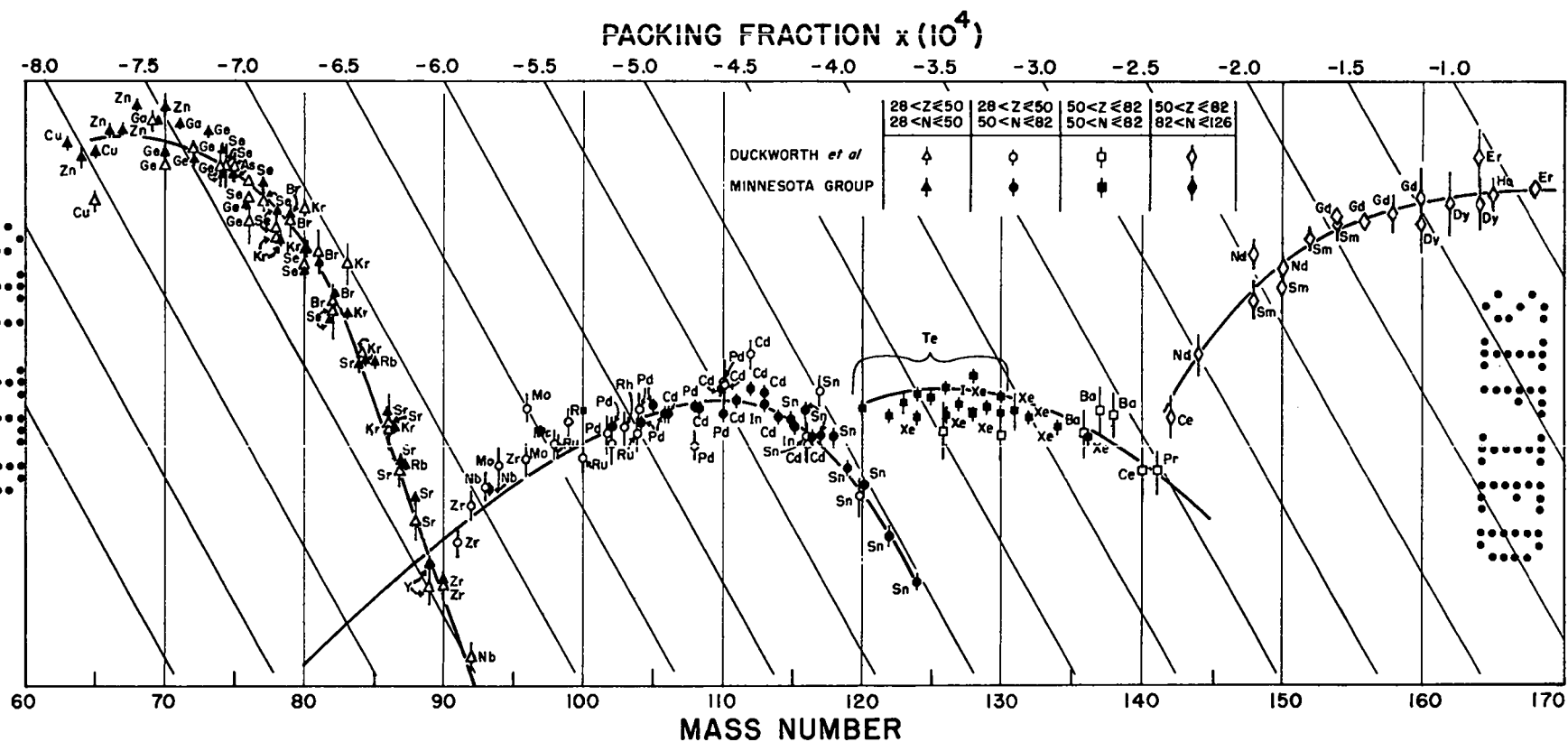
Inaccuracies in the empirical values of E_K , M , and T in the above expressions cause errors in the resulting neutron emission probabilities. Consequently, in this analysis the results are normalized to the measured $\bar{\nu}$ values.

MASS SURFACE

For the short-lived, neutron-rich fission products, essentially no experimental data of the atomic masses or the binding energies of neutrons exist. Consequently, semi-empirical data are used to determine these masses and binding energies. The masses M and the binding energy B used in Eq. 1 are from the compilations by Huizenga and Magnusson and by Glass,³ but with the latter values normalized to the U^{236} mass of the former for consistency. The semi-empirical mass surface for the fission products is a combination of the valley of the mass surface shown in Fig. 1, the parabolic mass surface of isobars, and the even-odd parameter of nuclear constitution. The valley of the mass surface in Fig. 1 is based on the mass-spectrographic measurements by Duckworth et al.⁴ and by the Minnesota group,⁵

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Fig. 1--Valley of the semi-empirical mass surface. Points are mass-spectrographic data converted to odd A and to the non-integer Z corresponding to the valley for each mass number A.

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converted to masses of the non-integer stable Z of each isobar by the parabolic constants and nuclear charges tabulated by Coryell⁶ and to odd A by the even-odd parameters of Fermi.⁷ These parabolic surface constants are determined from data closer to the mass valley than are the fission products and thus, in this sense, the M and B values used in the calculations are of unknown accuracy.

The position on the mass surface for which the masses and neutron binding energies are computed is determined in A by the mass ratio R_A chosen and in nuclear charge Z by observed charge displacement values from the non-integer stable charge. However, these nuclear charges of fragments observed by Glendenin et al.⁸ and Pappas⁹ are of fragments after neutron emission. To determine the nuclear charges of the primary (before neutron emission) fission fragments of mass number A , the small effect of the average number of neutrons emitted by both the light and heavy fragment was considered. The division of the fission neutrons between the light and heavy fragment was based upon the division observed by Fraser and Milton.¹⁰ In the calculations the distribution in Z was not taken into account, but only the most probable nuclear charge for each ratio R_A was used. It can be shown from the nuclear charge distribution of Glendenin et al.⁸ that for each isobar the energy sum $E_K + E_X$ from Eq. 1 is a narrow distribution, compared to other distributions to be considered, and can be neglected.

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The validity of using a semi-empirical mass surface to compute neutron binding energies has been shown¹¹ by comparison with measured binding energies. When shell effects in the mass surface are not considered, these comparisons show a discrepancy between calculated and measured binding energies in the regions of nuclear shells. Based on Fig. 1, the present determinations of binding energies take into account these shell effects. In Table I are the resulting binding energies of the most probable R_A of $U^{235} + n$ fission, where n represents thermal-neutron induced fission.

TABLE I. NEUTRON BINDING ENERGIES CALCULATED FROM THE SEMI-EMPIRICAL MASS SURFACE

Values are for the fission products of $U^{235} + n$ when $R_A = 141/95$.

The even-odd term δ_1 represents primary fission products with an odd number of neutrons; δ_2 represents an even number of neutrons.

ν	$B^L(\delta_1)$	$B^L(\delta_2)$	$B^H(\delta_1)$	$B^H(\delta_2)$
1	4.17	5.71	4.05	5.63
2	10.26	10.26	9.93	9.93
3	15.02	16.56	14.30	15.88
4	21.60	21.60	20.63	20.63
5	26.91	28.45	25.46	27.04

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and the width of the dispersion. However, since the comparisons indicate a Gaussian dispersion is suitable, a Gaussian dispersion

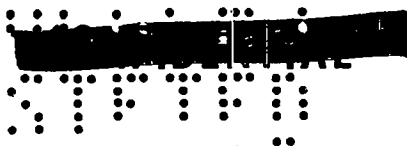
$$D(E_K, E_I + \Delta) \propto \exp \left[- \left(\frac{E_I + \Delta - E_K}{u} \right)^2 \right] \quad (7)$$

is used in Eq. 6 to simplify calculations. A width $u = 7.2$ Mev is used as a weighted average between the widths determined from the two comparison methods. However, within the uncertainty of these width determinations the calculated neutron emission probabilities vary negligibly with u .

The distribution $X(E_X, \delta, R_A)$ in the total excitation energy is obtained from $K(E_K, R_A)$ by a substitution of E_X for E_K by Eq. 1. Due to the limited experimental and theoretical information available on the relative excitations of the light and heavy fragments, a somewhat arbitrary division of X into the excitation energy distributions X^L and X^H of the individual fragments is made. The division of the excitation energy is into identical distributions $X^L = X^H$ of the respective arguments E_X^L and E_X^H , which are related by $E_X = E_X^L + E_X^H$. These distributions are from $X(E_X, \delta, R_A)$ by the convolution of Eq. 2.

In practice, the use of the empirical distribution $I(E_I, R_A)$ in the convolutions of Eqs. 5 and 6 to obtain the excitation energy distributions X^L and X^H is difficult. Instead, the ionization energy distributions were fitted by a sum of 13 Gaussian functions of equal

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width and various amplitudes, regularly spaced along the energy axis. Such a sum of Gaussian expressions in Eqs. 5 and 6 results in the excitation energy distribution

$$X^L(E_X^L) = \sum_{\alpha=0}^6 a_{\alpha} \exp \left[- \left(\frac{E_0 - E_X^L + \alpha d}{w} \right)^2 \right] \quad (8)$$

and the corresponding excitation energy distribution X^H of the heavy fragment, where α is a summation parameter. The amplitude coefficients a_{α} , the width w , and the energy spacing d determining the shape of the distribution are in Table II for the three R_A values used for $U^{235} + n$ and $Pu^{239} + n$ fission. The magnitude of the excitation energy is given by E_0 , where $E_0 = E_0(\delta)$. In Fig. 2 is shown the excitation energy corresponding to the most probable R_A of $U^{235} + n$ fission. Although the negative excitation energies and negative probabilities resulting from this approximate method of analysis have no physical meaning, both have mathematical meaning and are carried in the analysis.

For each fissile nuclide, the neutron emission probabilities were calculated for three mass number ratios R_A , with appropriate weighting for each group. The separation between the higher mass number ratios was the 82-neutron shell.

Since no distributions of ionization energy as a function of mass number ratio have been reported for spontaneous fission, the



TABLE II. VALUES USED IN THE FIT OF X^L AND X^H BY EQUATION 8

The tabulated E_0 values are for fission into primary fragments of odd A. Even-A fission products result in E_0 values differing by the odd-even mass term.

Fission Case	R_A	E_0 (Mev)	d (Mev)	w (Mev)	a_0	a_1	a_2	a_3	a_4	a_5	a_6
$U^{235} + n$	133/103	-22.16	11.0	8.33	-.0001	-.0006	.0023	.0587	.0039	.0052	-.0020
	141/95	-16.48	9.0	6.25	.0007	-.0030	.0088	.0735	.0034	.0049	.0021
	149/87	-13.11	7.5	4.36	-.0023	.0010	.0134	.1059	.0086	.0073	-.0044
$Pu^{239} + n$	131/109	-20.52	11.0	8.33	.0027	-.0073	.0153	.0450	.0061	.0058	-.0004
	139/101	-13.42	9.0	6.25	.0068	-.01530	.0229	.0671	.0050	.0066	-.0027
	147/93	- 8.91	7.5	4.36	-.0032	.0025	.0131	.1096	.0016	.0099	-.0042

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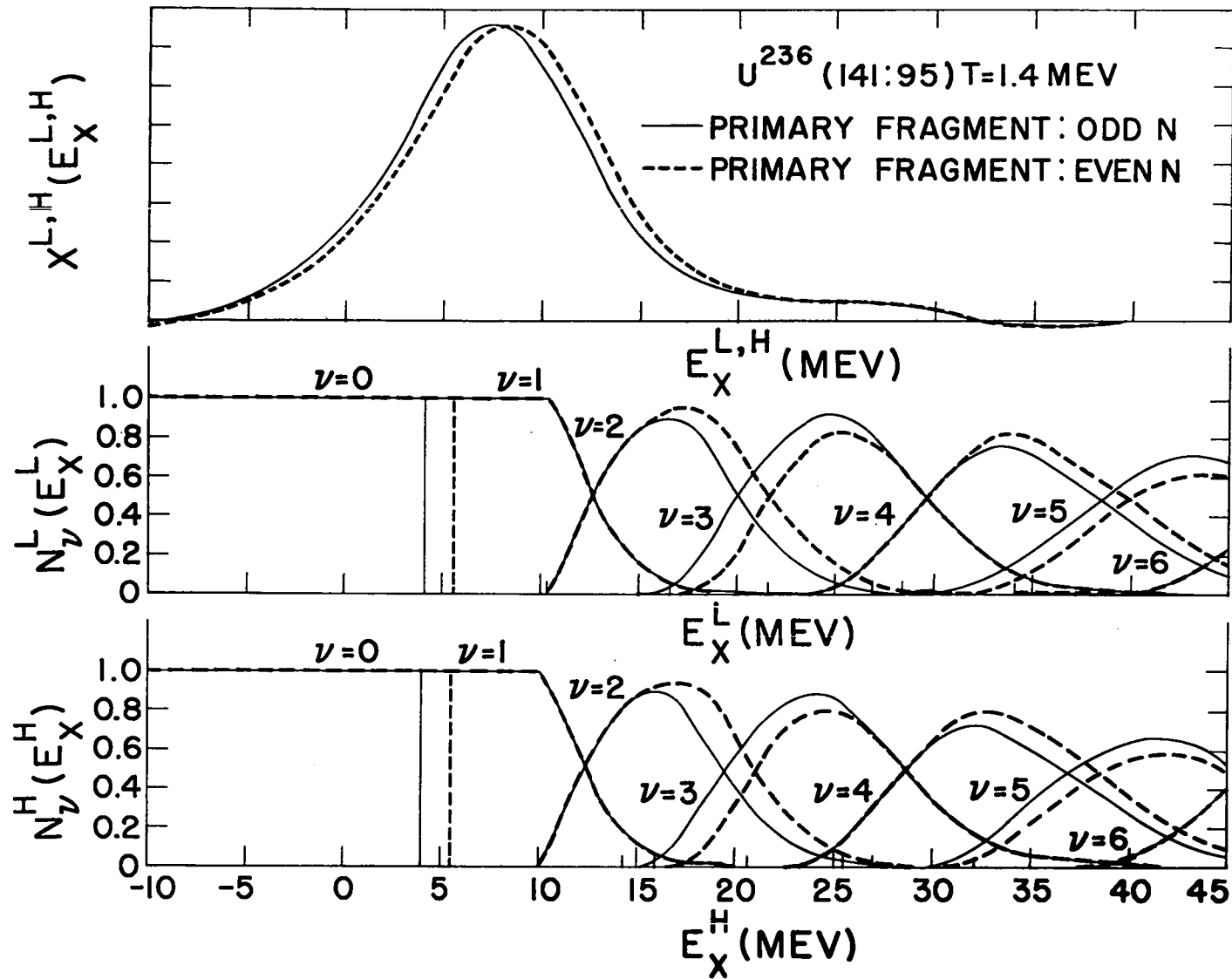


Fig. 2--Excitation energy distributions and neutron emission probabilities for $U^{235} + n$ fission. The excitation energy distributions, X^L and X^H , are based on $E_K = 169.1$ Mev and result in $\bar{\nu} = 2.5$.

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distribution obtained for $U^{235} + n$ fission is applied to the case of spontaneous fission of U^{238} and, similarly, that of $Pu^{239} + n$ is applied to the cases of Cm^{242} , Cm^{244} , and Cf^{252} spontaneous fission. It has been shown^{12,13} that the energy distributions are very similar for all the fission cases measured, and so this application of data is expected to cause only a small error. In this application of the data, the mass distribution of the heavy fragment is considered to be fixed¹⁸ for the fission of $U^{235} + n$ and U^{238} and fixed for the fission of $Pu^{239} + n$, Cm^{242} , Cm^{244} , and Cf^{242} .

NEUTRON EMISSION

The theoretical considerations of the evaporation model¹⁹ of the nucleus have been compared with experimental data by Cohen.²⁰ In this comparison, the model as expressed by Eq. 3 was found to agree with observed neutron emission probabilities, but failed to explain all aspects of particle evaporation with a constant T . In the present considerations, however, the T appropriate for neutron evaporation resulting from excitations of roughly 5 to 15 Mev is of importance. The neutron "temperature" T is thus determined from Eq. 3 and the measured $(n,2n)$ excitation functions,²¹ which are of these excitation energies. A "temperature" $T = 1.4$ Mev was found to provide the best fit of these data in the region of A required. Although this T is for nuclides with neutron-proton ratios corresponding to the valley of the mass surface, the T of the neutron-rich fission products with

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somewhat larger neutron-proton ratios is not expected to differ greatly.

The neutron evaporation expression (Eq. 3) is integrated over all possible neutron energies in the full succession of neutron emissions possible and the emission probabilities

$$N_0^L(E_X^L, \delta^L, R_A) = 1 \text{ for } E_X^L < B_1^L \quad (9)$$

$$= 0 \text{ for } E_X^L \geq B_1^L$$

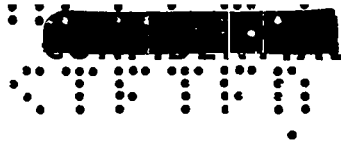
$$N_1^L(E_X^L, \delta^L, R_A) = 0 \text{ for } E_X^L < B_1^L$$

$$= 1 \text{ for } B_1^L \leq E_X^L < B_2^L$$

$$\approx \left\{ \left[\exp \left(- \frac{E_X^L - B_2^L}{T} \right) \right] \left[\left(\frac{E_X^L - B_2^L}{T} \right) + 1 \right] \right\}$$

$$\text{for } E_X^L \geq B_2^L$$

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$$N_{\nu}^L(E_X^L, \delta^L, \nu^L, R_A) = 0 \text{ for } E_X^L < B_{\nu}^L \text{ and } \nu > 1 \quad (9)$$

(Cont'd)

$$\approx \left\{ 1 - \left[\exp \left(- \frac{E_X^L - B_{\nu}^L}{T} \right) \right] \left[1 + \sum_{\mu=1}^{2\nu-3} \frac{\left(\frac{E_X^L - B_{\nu}^L}{T} \right)^{\mu}}{\mu!} \right] \right\}$$

$$\text{for } B_{\nu}^L \leq E_X^L < B_{\nu+1}^L \text{ and } \nu > 1$$

$$\approx \left\{ \left[\exp \left(- \frac{E_X^L - B_{\nu+1}^L}{T} \right) \right] \left[1 + \sum_{\mu=1}^{2\nu-1} \frac{\left(\frac{E_X^L - B_{\nu+1}^L}{T} \right)^{\mu}}{\mu!} \right] - \left[\exp \left(- \frac{E_X^L - B_{\nu}^L}{T} \right) \right] \left[1 + \sum_{\mu=1}^{2\nu-3} \frac{\left(\frac{E_X^L - B_{\nu}^L}{T} \right)^{\mu}}{\mu!} \right] \right\}$$

$$\text{for } E_X^L \geq B_{\nu+1}^L \text{ and } \nu > 1$$

are thus obtained, where μ is a summation parameter. As discussed in the Mass Surface Section (page 9) the binding energies B_{ν}^L are calculated from the mass surface. Similar expressions of neutron emission N_{ν}^H for the heavy fragment are obtained. These neutron emission probabilities N_{ν}^L and N_{ν}^H , with the binding energies of Table I, are



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shown in Fig. 2.

The probabilities P_{ν}^L and P_{ν}^H of emitting ν^L and ν^H neutrons from the light and heavy fragment, respectively, are calculated from the integral Eq. 4 and the corresponding expression for the heavy fragment. In the present calculations an IBM 701 was used for the integration. After the probabilities for the various even-odd conditions are combined, the neutron emission probabilities, P_{ν}^L and P_{ν}^H of each fragment were combined by the summation Eq. 5 into the emission probabilities P_{ν} of both fragments. The mass number ratio R_A was removed by summation over its three values. With the calculations discontinued after $\nu = 9$, the average number of neutrons was obtained by the summation $\bar{\nu} = \sum_{\nu=0}^9 P_{\nu}$. The second and third moments, $\overline{\nu^2}$ and $\overline{\nu^3}$, respectively, were similarly obtained.

RESULTS

The calculations of neutron emission probabilities were normalized to the measured values of $\bar{\nu}$ by adjusting Δ . Normalization of the thermal-neutron induced fission cases were to $\bar{\nu} (U^{235} + n) = 2.5 \pm 1$ and $\bar{\nu} (Pu^{239} + n) = 3.0 \pm .1$ (Ref. 22). The average number of neutrons from the spontaneous fission of U^{238} was measured as $2.2 \pm .3$ by Segrè²³ and $2.5 \pm .2$ by Littler²⁴ and normalization has been made to both these values. Normalization was made to the following measured values of $\bar{\nu}$ for spontaneous fission: $\bar{\nu} (Cm^{242}) = 3.1 \pm 10$ -15 percent (Ref. 25), $\bar{\nu} (Cr^{252})/\bar{\nu} (Cm^{244}) = 1.39 \pm 2$ percent (Ref. 26), and $\bar{\nu} (Cr^{252}) = 3.10 \pm .18$ (Ref. 27). The measured average

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ionization energies (E_I) are compared in Table III with the average kinetic energies (E_K) obtained from these normalizations. The Δ values obtained as a difference between these average kinetic and ionization energies are in reasonable agreement with the 12-Mev ionization defects obtained from the comparison of ionization data¹² with velocity¹⁴ and calorimetric¹⁵ data.

TABLE III. AVERAGE ENERGIES OF THE FISSION PRODUCTS

The average energies (E_I) from ionization chamber measurements contain source and collimator corrections. The indicated uncertainties in the calculated average kinetic energies (E_K) are from the uncertainties in $\bar{\nu}$ only.

Fission Case	E_K (Mev)	E_I (Mev)	Δ (Mev)	E_I Reference
$U^{235} + n$	$169.1 \pm .8$	154.7	14.4	12
U^{238}	$165.4 \pm 2.4^a, 162.9 \pm 1.6^b$	151.7	$13.7,^a 11.2^b$	28 ^c
$Pu^{239} + n$	$174.2 \pm .8$	159.8	14.4	13
Cm^{242}	181.4 ± 3	160	21.4	29
Cm^{244}	186.9 ± 1.2			
Cr^{252}	192.7 ± 1.3			

^aNormalized to = $2.2 \pm .3$

^bNormalized to = $2.5 \pm .2$

^cThese data are relative to the energy of fragments from fission of $U^{235} + n$, for which the energy of Ref. 12 is used.

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TABLE IV. CALCULATED AND EXPERIMENTAL NEUTRON EMISSION PROBABILITIES

Both data are normalized to the same $\bar{\nu}$ values given in the text.

Fission Case Method		P_0	P_1	P_2	P_3	P_4	P_5	P_6	P_7	P_8	P_9
$U^{235} + n$	Calc.	.0227	.1543	.3475	.2875	.1429	.0460	.0043	-.0031	-.0016	-.0005
U^{238}	Calc. ^a	.0430	.2168	.3731	.2430	.1015	.0282	-.0008	-.0033	-.0012	-.0003
	Calc. ^b	.0223	.1508	.3490	.2946	.1391	.0455	.0047	-.0038	-.0017	-.0005
$Fu^{239} + n$	Calc.	.0106	.0627	.2412	.3597	.2383	.0840	.0154	-.0063	-.0041	-.0015
Cm^{242}	Calc.	.0095	.0536	.2347	.3708	.2498	.0821	.0110	-.0068	-.0035	-.0011
Cm^{244}	Calc.	.0418	.1826	.3967	.2512	.1165	.0224	-.0041	-.0051	-.0018	-.0003
Cf^{252}	Calc.	.0129	.0651	.2286	.3516	.2132	.0911	.0411	-.0009	-.0008	-.0020
	Exp. ^c	-.0077 +.0118	.1176 +.0400	.1656 +.0906	.3496 +.1118	.3253 +.1354	-.0126 +.0706	.0621 +.0294	0		

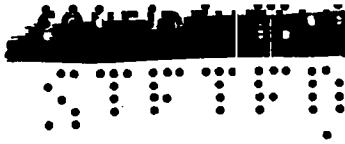
^aNormalized to $\bar{\nu} = 2.2$

^bNormalized to $\bar{\nu} = 2.5$

^cReference 2

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The emission probabilities P_ν of ν neutrons from fission as obtained from these calculations are compared in Table IV with directly measured values and the agreement is seen to be good. The negative emission probabilities in this table result from the negative probabilities of excitation energy when values of Table II are used in Eq. 8. Similarly the ratios $\overline{\nu^2/\nu}$ and $\overline{\nu^3/\nu}$ from spontaneous fission of U^{238} are compared in Table V. These comparisons in Tables IV and V indicate the use of the calculated excitations in the evaporation model of the nucleus satisfactorily explains the observed fluctuations in the number of neutrons emitted from fission.

TABLE V. MULTIPLICITIES OF NEUTRONS FROM THE SPONTANEOUS
FISSION OF U^{238}

Method	$\overline{\nu^2/\nu}$	$\overline{\nu^3/\nu}$
Calc. ^a	2.70	8.02
Calc. ^b	2.96	9.58
Exp. ^c	$3.26 \pm .2$	$12.73 \pm .9$

^aNormalized to $\bar{\nu} = 2.2$.

^bNormalized to $\bar{\nu} = 2.5$.

^cReference 1.



[REDACTED]

[REDACTED]

The average number of neutrons from the light and heavy fragments obtained from these calculations gives $\overline{\nu^L}/\overline{\nu^H} = 1.05$ for fission of $U^{235} + n$ and, similarly, 1.03 for $Pu^{239} + n$. In contrast to the equal excitation energies assumed in this calculation, a comparison of these neutron ratios with the $\overline{\nu^L}/\overline{\nu^H} = 1.3$ ratios measured by Fraser and Milton¹⁰ indicates a larger excitation energy for the light fragment than for the heavy fragment. However, such a change in the excitation energies can be shown to change the neutron emission probabilities by only a small amount.

[REDACTED]

[REDACTED]

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[REDACTED]
[REDACTED]

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