Correlations between the Neutron Multiplicities and Spontaneous Fission Modes of Californium-252

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The numbers of prompt neutrons associated with specific fission modes of Cf^{252} have been measured. The average number of neutrons per fission depends largely upon the total kinetic energy of the fission fragments, decreasing by at least 0.06 neutron per fission for an increase of 1 Mev in the total kinetic energy. A less marked variation with the ratio of fragment masses is observed.

I. INTRODUCTION

BECAUSE of the complicated nature of the fission process, the most promising theoretical approaches[1-8] at the present time are of a semiempirical nature. Existing experimental data that can be incorporated in or used as tests of such theories include spontaneous-fission half-lives,[9,10] nuclear-mass surfaces,[11,12] relative probabilities of fission modes (including the kinetic energy[13-16] and mass-ratio[17,18] distributions of the fission fragments), average numbers[19-21] and energies of prompt neutrons,[22] energies of prompt gamma rays,[23] and average probabilities of prompt-neutron emission from spontaneous or low-energy-induced fission.[19,20,24] The work described here was performed to determine the neutron multiplicities as functions of fission modes; such numbers are closely related to the distribution of excitation energy at the time of fission. To provide ease of handling and low backgrounds, we used the spontaneously fissioning isotope of Cf^{252}. Preliminary results have been reported.[25]

The mass equation of neutron-induced binary fission, which holds just after the fission has occurred but before the emission of neutrons, is

$$M(A,\delta,Z)+E_n+B=M(A^H,\delta^H,Z^H)+M(A^L,\delta^L,Z^L)+E_K+E_x,$$

where the atomic masses M are functions of the atomic number A, the charge Z, and the even-odd parameter δ of the semiempirical mass formula. The superscripts L and H refer to the light and heavy fragments, respectively. E_K is the total kinetic energy, and E_x the total excitation energy of the two fragments. E_n, the kinetic energy of the incident neutron, and B, the neutron binding energy, are absent in spontaneous fission, but we include them here for the sake of the later discussion.

For a given mass ratio M_H/M_L, a distribution in E_K is observed which is due to a real distribution (caused primarily by a small charge fluctuation) and dispersion from (a) the momentum distribution due to the recoil of the fission fragments when neutrons are emitted, (b) the rather poor energy resolution of fission chambers, and (c) ionization defect. From the true distribution in E_K, the distribution in E_x can be determined immediately from the mass equation of fission, if it is assumed that the small charge distribution that is observed does not affect the total energy released. From the distributions in E_x, the neutron-emission probabilities can be determined by use of neutron-evaporation theory.

From a comparison of fission-chamber measurements and chemical fission-product data, Leachman[5] has attempted to correct for the dispersions caused by (b) and (c). Normalizing his calculations to the measured average numbers of neutrons per fission, he proceeded

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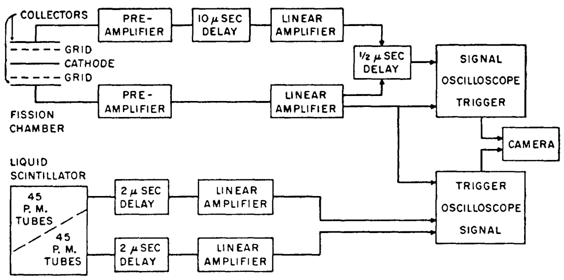

FIG. 1. Block diagram of apparatus.

as outlined above[8] to obtain the probabilities $P(\nu)$ the ν neutrons are emitted in a fission event, for three particular mass ratios. When averaged to correspond to an actual mass-ratio distribution, these results agree well with experiment. The measurements reported here will make it possible to extend the comparison between theory and experiment to specific fission modes.

II. METHOD AND APPARTAUS

The neutron-detection apparatus is a tank of cadmium-loaded liquid scintillator 30 in. in diameter and 30 in. high, viewed by photomultipliers distributed over the curve surface. A 3-in.-diameter well allows a small double ("back-to-back") fission chamber to be placed at the center of the detector.[20] The arrangement of the apparatus is shown schematically in Fig. 1. A pulse from one side of the back-to-back fission chamber triggers the sweeps of two oscilloscopes, and the pulses from the two sides of the fission chamber are displayed on one oscilloscope. The prompt gamma rays and proton recoils from the fission neutrons produce a pulse at the beginning of the trace of the second oscilloscope, which is followed by the neutron-capture pulses. Both sweeps are photographed on a single strip of continuously moving film (Fig. 2).

The fission chamber is of the double Frisch-gridded type, operated at 25 lb above atmospheric pressure. The gas was composed of 95% argon and 5% nitrogen. All fission fragments were stopped in the regions between the source and the grids. An amount of Cf^{252} sufficient to give 100 spontaneous fissions per minute was evaporated onto a 5-μg/cm² VYNS film flashed with 5-μg/cm² gold. The foil was in contact with a 10%-transmission Lectromesh grid, which served as a collimator for the fission fragments. Pulses from the collimated side of the fission chamber are used to trigger the recording apparatus.

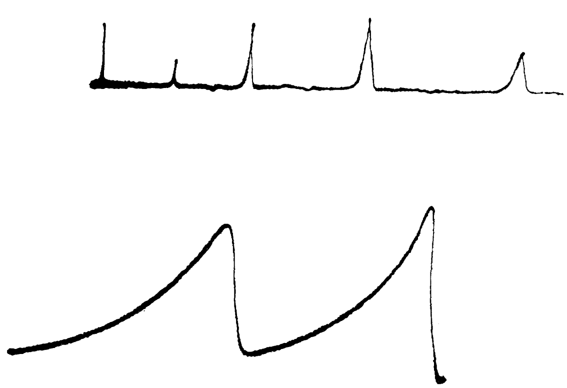

FIG. 2. Sweeps triggered by a fission-chamber pulse. Top: prompt-γ and recoil-proton pulse on the right, followed by neutron-capture pulses. Bottom: pulses from the two sides of the fission chamber. Parts of the traces have been reinforced with ink.

The oscilloscope sweeps were projected and read, and the data obtained in this way were sorted on an IBM Type 650 computer. Resolution and background corrections were introduced into the neutron-multiplicity calculations in the manner described in reference 20.

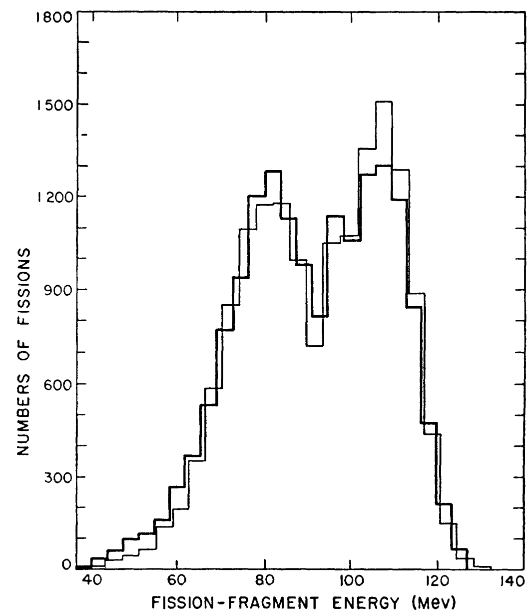

FIG. 3. Fragment-energy spectra (corrected for ionization defect) from both sides of the back-to-back fission chamber.

III. RESULTS AND DISCUSSION

Sixteen thousand spontaneous fissions were recorded and analyzed. Although the electronic pulse amplifications from the two sides of the fission chamber were approximately equalized with a calibrated pulser, it was later necessary to adjust all the pulses from one side by a constant multiplicative factor of about 1.07 to make the peaks of the energy distributions coincide. The energy scale was obtained from the back-to-back fission chamber data of Smith *et al.*[16] by multiplying all corrected pulse heights by a constant (the same for all pulses) to make the peaks of our number-*vs*-energy distribution coincide with theirs, namely at 100 and 77 Mev.[26,*] The ionization-defect correction was then

[26] Ionization-chamber measurements by Harry R. Bowman (private communication) indicate peaks in the Cf^{252} energy distribution at about 67 and 92 Mev before correction for ionization defect. Fragment-velocity measurements are now in progress at several laboratories.

* *Note added in proof.*—Alan B. Smith (private communication) has remarked that our fragment energy distributions are

obtained from an extrapolation of Leachman's velocity-selector measurements, and 5.0 Mev and 6.5 Mev were added to pulse heights in the high- and low-energy groups respectively. The resultant fission-fragment energy distributions from the two sides of the fission chamber are shown in Fig. 3, and the relative probabilities of the fission modes are shown in the contour diagram of Fig. 4.

Because of the necessary restriction on the fission chamber size, the gas pressure in the chamber was high, and as a result the energy resolution was poorer than that obtained by Smith *et al.*, as can be seen by comparing the fission-mode probability contour diagrams. However, the ratio of the high- to low-energy peaks is 1.36, in agreement with the results of Smith *et al.*

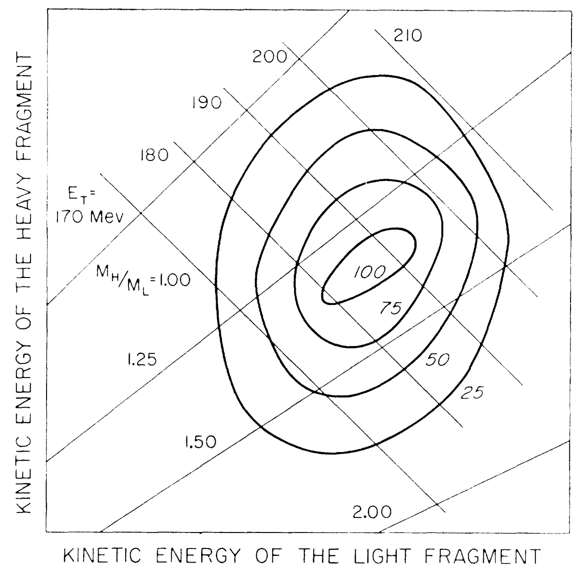

FIG. 4. The observed relative probabilities of the fission modes. Lines of constant total energy and constant mass ratio are shown also.

The observed average number of neutrons per fission, $\bar{\nu}$, and the number distribution of fission events as functions of the total kinetic energy are shown in Fig. 5. The events that have total energies less than about 140 Mev are suspect for two reasons: (a) the measurements by Smith and by Bowman with Cf^{252} and by other workers with various transuranic elements do not show such events, and (b) the values of $\bar{\nu}$ obtained in this region approach the average for all fission modes, probably indicating that these counts arise from a large dispersion.

If we consider only the events with total energies greater than about 160 Mev, a strong linear correlation

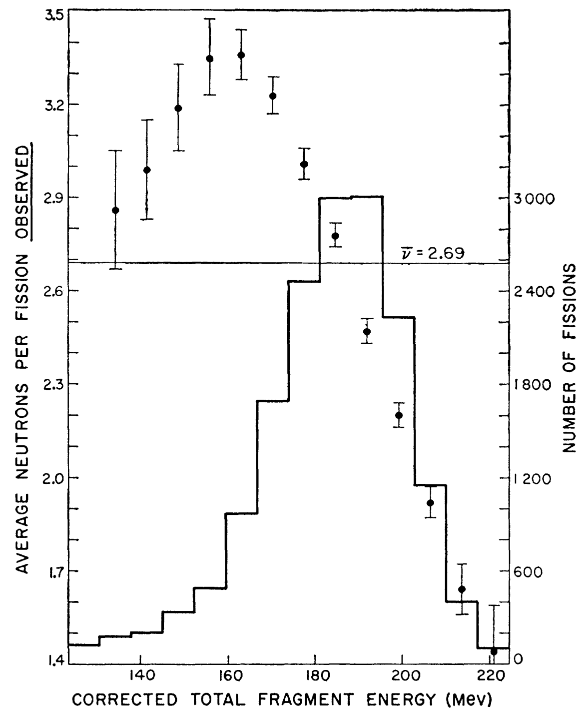

FIG. 5. Observed average numbers of neutrons per fission with standard errors, and the number distribution of fission events as functions of fission-fragment total kinetic energy (corrected for ionization defect).

of $\bar{\nu}$ with total kinetic energy is observed: in particular, $d\bar{\nu}/dE \approx 0.039$ observed neutrons per fission per Mev. The *observed* average number of neutrons per fission

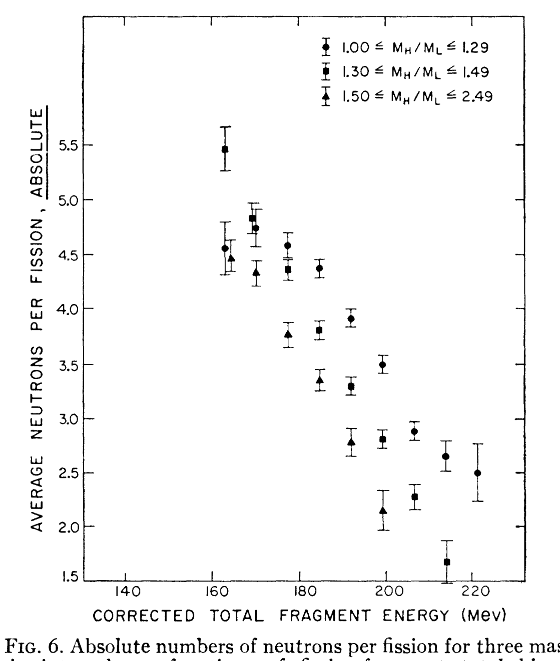

FIG. 6. Absolute numbers of neutrons per fission for three mass-ratio intervals as functions of fission-fragment total kinetic energies. The standard errors do not include the uncertainty in the neutron-detector efficiency.

characteristic of sources which may be thin when averaged over the total area, but on which the fissionable material is deposited in clumps. More recently, H. Bowman has prepared Cf^{252} sources by the same technique as used for the present one and obtained fragment energy peaks at 73.5 and 97.6 Mev. We conclude that our source was indeed thick to the fission fragments.

TABLE I. The calculated variation with total fragment kinetic energy, E_T, of the average number of prompt neutrons per spontaneous fission of Cf^{252}. 160 Mev $<E_T<$ 230 Mev. $\Delta\equiv$ the full width at half-maximum of the assumed Gaussian total-energy resolution function.

Δ(Mev):	0	13	16	20
$-\frac{\partial\bar{\nu}}{\partial E}$(neutrons/fission Mev):	0.055	~0.06	~0.08	~0.11

averaged over all fission modes is 2.69, and a comparison with the previously determined true value, $\bar{\nu}=3.82\pm0.12$,[20] gives a neutron-detection efficiency of $70.4\pm2.2\%$ for these measurements (the efficiency had fallen from a previous value of about 80% because of the deterioration of the scintillator solution). The value of $d\nu/dE$, corrected for efficiency but still not corrected for energy resolution, is therefore $d\nu/dE\approx0.055$. As a further refinement of the data, we plot the values of $\bar{\nu}$ *vs* total energy, corrected for the neutron-detection efficiency, for three different mass-ratio bands in Fig. 6, and it is seen that there is an inverse correlation between the mass ratio[27] and the average number of neutrons per fission for any given total energy.

It is difficult to correct our measurements for energy dispersion, because this effect is a function of the mass ratio and therefore is not constant within any kinetic energy interval. In an attempt to learn the type of effect that the energy dispersion has on $d\bar{\nu}/dE$, we assume that the dispersion does not vary with mass ratio or total kinetic energy, and that it can be represented by a Gaussian with a full width Δ at half-maximum. After the unfolding of this dispersion, the values of $d\bar{\nu}/dE$ shown in Table I are obtained for several assumed values of Δ. Leachman[5] has obtained an approximate value for the dispersion in the U^{235} fission-chamber measurements by Brunton and Hannah[13] by comparing their results with his measurements of fission-fragment velocities. With the aid of Cf^{252} and U^{235} fission-fragment energy distributions measured by Bowman,[26] we have obtained a crude estimate for the energy dispersion in our measurements from Leachman's conclusions. This value, $\Delta=13$ Mev, gives $d\bar{\nu}/dE\approx0.06$, but this figure may be low by perhaps 30% or more, depending upon the manner in which the dispersion varies with the fission mode.

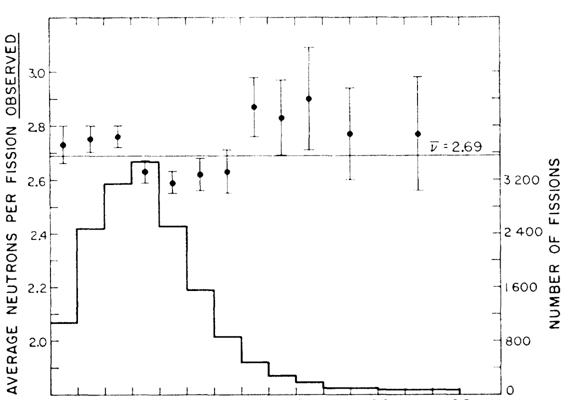

FIG. 7. Observed average numbers of neutrons per fission (standard errors) and numbers of fission events as functions of the ratio of fragment kinetic energies.

Fowler[28] has observed that the experimentally determined average kinetic energy of the fission fragments from neutron-induced fission does not depend on the energy of the neutron causing fission, showing that the neutron kinetic energy is distributed as excitation energy. He and Leachman have calculated the variation in $\bar{\nu}$ with the energy of the incident neutron. Leachman[8] has obtained a value for $d\nu/dE_n$ of about 0.13 neutron/fission Mev for a nuclear temperature of approximately 1.4 Mev. This energy dependence is in good agreement with measurements by Fowler, by Terrell, and by Diven, Martin, and Terrell. From an examination of the mass equation of fission, one is tempted to assume that the dependence on kinetic energy in spontaneous fission might be similar (as indeed it seems to be from our measurements), but inasmuch as the total available energy depends on mass ratio, it is not possible to explain the dependence of $\bar{\nu}$ on E_K in such a simple way.†

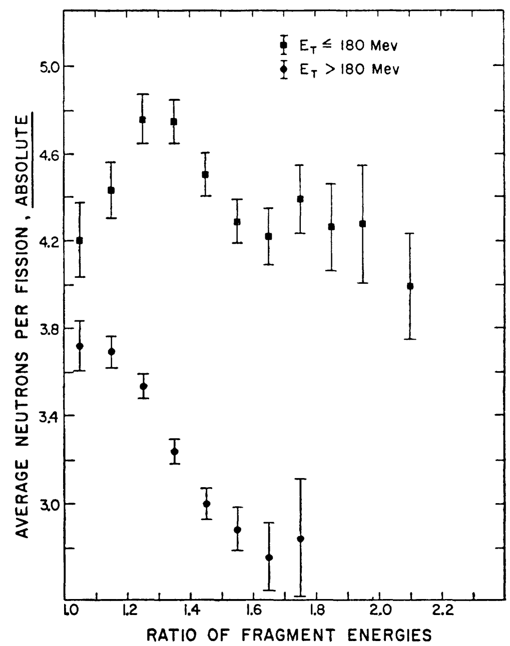

FIG. 8. Absolute numbers of neutrons per fission for two total-kinetic-energy intervals as functions of the fragment-kinetic-energy ratios. The standard errors do not include the uncertainties in the neutron-detector efficiency.

[27] From conservation of momentum, we have $E_H/E_L=M_L/M_H$.

[28] J. L. Fowler, quoted by R. Leachman, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Paper No. 592, Vol. 2, p. 193.

† *Note added in proof.*—R. B. Leachman and C. S. Kazek, Jr. [Phys. Rev. (to be published)] have now calculated the value $\partial\bar{\nu}/\partial E=-0.116$ for Cf^{252}.

The dependence of the average numbers of neutrons per fission on the ratio of masses or kinetic energies of the fragments is given in Fig. 7. It is seen that there is at most a small variation with mass ratio when no discrimination is made on the basis of total energy. However, when the fissions are first divided into two roughly equal groups with total kinetic energies greater than or less than 180 Mev, there is an obvious dependence on mass ratio (Fig. 8). The effect of the energy resolution of the apparatus has not been subtracted from these data.

Finally, the variation of the mean total kinetic energy of the fragment pairs with mass or energy ratio is given in Fig. 9.‡

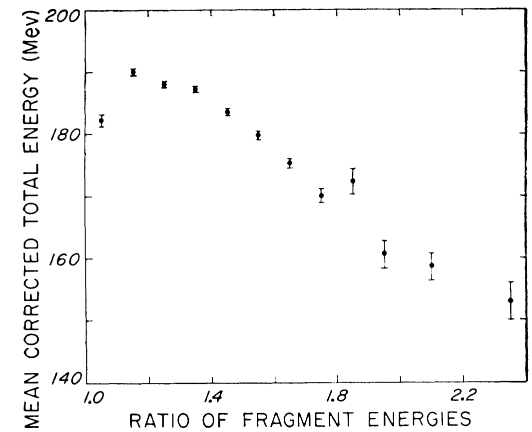

FIG. 9. The mean total kinetic energy of the fission-fragment pairs as a function of the ratio of kinetic energies of the fragments. Errors are statistical rather than absolute.

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‡ *Note added in proof.*—H. Bowman (private communication) has carried out a similar experiment which includes a check of the present results with improved energy resolution. A partial analysis of the data indicates that there will be fairly good quantitative agreement with this work.

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Neutron Emission from Fission Modes*

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The number and energy of neutrons and the average energy of prompt gamma rays emitted from various fission modes are estimated from the excitation-energy distributions of fragments from these modes. These excitation-energy distributions are derived from the mass equation of fission and the measured kinetic energies of the fragments. Simple considerations of neutron boil-off are used with these excitations.

For the most probable mass ratios of fragments, the variation in the average number of neutrons $\bar{\nu}$ with the total kinetic energy E_K of the fragments is found to be $d\bar{\nu}/dE_K = -0.121$ Mev^{-1} for thermal-neutron fission of U^{235} and -0.116 Mev^{-1} for spontaneous fission of Cf^{252}. The spectra of neutron energies resulting from this analysis are found to have negligible change with E_K, but the neutrons from Cf^{252} fission are more energetic than those from thermal-neutron fission of U^{235}. The average energy E_γ of prompt gamma rays from Cf^{252} fission is found to be 4.0 Mev, with a variation $dE_\gamma/dE_K = -0.0167$ for the most probable mass ratios of fragments.

INTRODUCTION

THE number and energy of neutrons emitted from fission depend in a complex manner on the excitation of the fragments, the nuclear identity of the fragments and the channels through which the excitation is expended. As an aid to understanding the detailed observations[1] of multiplicities of fission neutrons, calculations[2] have been made from a greatly simplified model of fission involving a statistical approach to the determination of the excitation and to neutron emission. In this analysis, empirical data of the energetics of fission fragments and the masses of nuclides were used to determine the distribution in total excitation energy shared by the two fragments, and then statistical assumptions were applied to estimate the distributions of excitation energy of the individual fragments and the neutron emission. The estimates of the probabilities P_ν of emitting $\nu = 0$, $\nu = 1$,

* Work performed under the auspices of the U. S. Atomic Energy Commission.

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