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Prompt-Neutron Emission from Single Fission Fragments*

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With a Cf^{252} source placed at the edge of a large cadmium-loaded liquid scintillator, it has been possible to obtain a measure of the number of prompt neutrons emitted from a single fission fragment of measured mass. The mass determination results from a concurrent time-of-flight measurement of the two fragment velocities. The 30-in. scintillator has a high detection efficiency, little dependent on the energy of the neutrons, and permits detection in a full hemisphere about the direction of one of the fragment flight paths. The average number of neutron counts per fission is found to increase with the mass number of the fragment approaching the neutron detector in much the same way in both the light- and heavy-fragment groups, with a sharp decrease occurring in passing from the light to the heavy group. A correction for the geometry of the neutron detection, assuming isotropic emission of the neutrons in the fragment frames, enhances slightly the sawtooth dependence of $\bar{\nu}(A)$ and gives for the ratio of the average number of neutrons from the light fragment to that from the heavy fragment $\bar{\nu}_L/\bar{\nu}_H=1.02\pm0.02$. One is led to believe that either the emission of the neutrons is far from isotropic or that the slightly lighter fragments possess considerably more excitation energy than the slightly heavier fragments when the mass division is nearly symmetric. Perhaps a new picture of the mass division is indicated.

I. INTRODUCTION

THE prompt-neutron-emission probabilities in fission are expected to be closely related to the average excitation energies of the fission fragments. A variation of the average total excitation energy of the fragments with mass ratio has recently been reported.[1] Evidence has now been found that the total excitation energy may be shared unequally by the two fragments in a way dependent on the mass division. A picture of the fission process, somewhat different from that commonly held, may be indicated.

The present work[2] is an extension of that previously reported,[1] in which the average number of neutrons emitted from both fragments was measured as a function of the mass and kinetic-energy modes of fission. With a Cf^{252} source placed at the edge of a large cadmium-loaded liquid scintillator, it is possible to obtain a good measure of the number of neutrons emitted from a single fission fragment, provided that the neutrons are emitted reasonably isotropically in the fragment frames. There is known to be a strong correlation between fragment and neutron directions,[3] which is assumed to result from the emission of the neutrons from fast-moving fragments. The masses and kinetic energies of the fragments are determined by time-of-flight measurement of the fragment velocities.

II. APPARATUS

The apparatus is shown schematically in Fig. 1. Except for the change in position of the Cf^{252} source with respect to the liquid scintillator, the source, detection apparatus, and electronic recording equipment are essentially as described in reference 1. To summarize briefly: The source consists of a small amount ($\sim10^3$ spontaneous fission/min) of Cf^{252} deposited over a small area (~1 cm^2) on a thin (~0.1 mg/cm^2) nickel foil. The time-of-fission signal is obtained from electrons (ejected from the source foil by one of the frag-

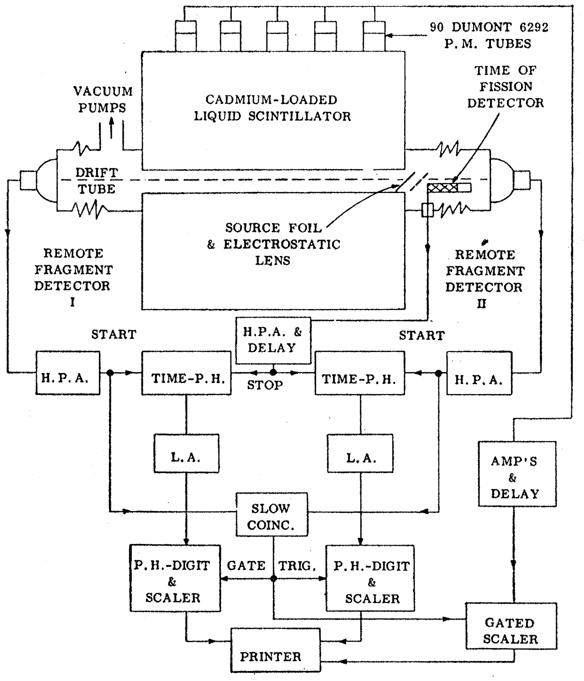

FIG. 1. Schematic diagram of the apparatus. H. P. A. = Hewlett-Packard Model 460*B* distributed amplifier; H.P.A. and DELAY = amplifiers and 200-ohm cable; TIME-P.H. = time-to-pulse-height converter; L.A. = modified Los Alamos Model 101*A* linear amplifier; P.H.-DIGIT = pulse-height-to-digital converter; AMP's and DELAY = modified Los Alamos 503*A* preamplifier and amplifier, H.P. Model 460*B* amplifier, and cable.

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

[1] W. E. Stein and S. L. Whetstone, Jr., Phys. Rev. **110**, 476 (1958).

[2] A preliminary account of this work was presented at the American Physical Society meeting in Vancouver, British Columbia on August 28, 1958 [S. L. Whetstone, Jr., Bull. Am. Phys. Soc. Ser. II, **3**, 337 (1958)].

[3] J. S. Fraser, Phys. Rev. **88**, 536 (1952).

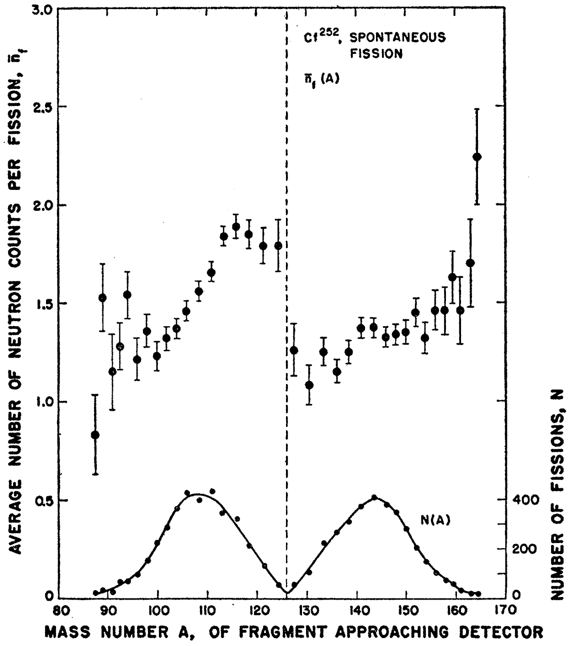

FIG. 2. The average number of neutron counts per fission $\bar{n}_f$ as a function of the mass number A of the fragment approaching the neutron detector. Uncertainties shown are relative standard errors.

ments) which are accelerated and focused onto a thin plastic scintillator. The fragments travel, in this case, 154 cm in evacuated drift tubes before they are detected in 4-in.-diameter thin plastic scintillators. The fragment flight times are measured using time-to-pulse-height and pulse-height-to-digital converters. The analyzer channels are about 1.2 mμsec wide, about one-third the full width at half-maximum of the present timing-resolution curve. The calibration of these instruments is checked automatically during the course of the experiment by having a series of pulses, derived from a single pulser, introduced at one- to two-hour intervals into the signal outputs of the three detectors. The cylindrical liquid scintillator that detects the neutrons is 30 in. long and $28\frac{1}{2}$ in. in diameter and subtends a 2π solid angle at the source. This solid angle is centered about the flight path of a fragment that traverses the $2\frac{5}{8}$-in. diameter tube along the axis of the detector. The scintillator solution consists of triethylbenzene, *p*-terphenyl, POPOP, and cadmium octoate. The neutron counts (resulting from thermalized-neutron captures in the dissolved cadmium) that occur within a 50-μsec time interval following (by 0.75 μsec) the prompt pulse due to fission γ rays and proton recoils in the scintillator are recorded in a gated scaler. The background is sampled continuously by counting for the same length of time in another scaler 500 μsec after the prompt pulse. The data are printed automatically on a paper tape, transferred to punched cards, and processed with the help of an IBM-704.

III. RESULTS

A total of 7158 events has been analyzed. Comparing the observed average neutron counting rate, corrected for the background counting rate and the detection geometry, with the measured value[4] of $\bar{\nu}=3.86\pm0.07$ averaged over all fission modes, one finds that the efficiency of the neutron detector was ~68%. The ratio of the number of background to fission-neutron counts was ~0.10.

The average number of neutron counts per fission $\bar{n}_f$ as a function of the mass number A of the fragment approaching the detector is shown in Fig. 2. The most striking feature is the large decrease in $\bar{n}_f$ in going from the light- to the heavy-fragment group at the mass corresponding to the symmetric mass division, A=126.

To correct for the geometry of the neutron detection, that is, for the fraction of the neutrons emitted from fragments approaching the neutron detector that are not emitted into the forward hemisphere in the laboratory, and for the fraction of the neutrons emitted into the backward hemisphere in the laboratory from departing fragments, it seems most reasonable, still, to assume that neutrons are emitted from the fully accelerated fragments *isotropically* in the fragment frames, with an energy spectrum that will produce the measured laboratory spectrum. The calculation, which uses the center-of-mass neutron-energy spectrum deduced by Terrell,[5] is summarized in Fig. 3. The fraction of neutrons carried into the forward hemisphere from fragments of velocities $\bar{v}_f$ is given by the curve $G_+(\bar{v}_f)$. The curve $\bar{v}_f(A)$ shows the relation between the average velocity and mass of the fragments obtained from the data of this experiment. The correlation between the velocity and mass of a fragment is well defined; the standard deviation in the fragment velocity for a given mass, $\sigma_{vf}\sim0.005\times10^9$ cm/sec, is relatively small. The heavy curve $G_+(A)$ gives the geometric correction factor as a function of the fragment mass number. The cor-

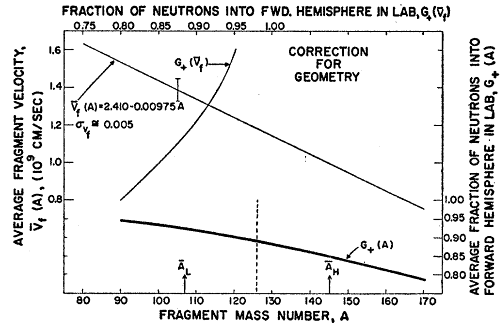

FIG. 3. Summary of the correction for the geometry of the neutron detection, assuming isotropic emission of the neutrons in the fragment frames.

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[4] Obtained from the results of Diven, Martin, Taschek, and Terrell, Phys. Rev. **101**, 1012 (1956), and Hicks, Ise, and Pyle, Phys. Rev. **101**, 1016 (1956).

[5] J. Terrell, Phys. Rev. **113**, 527 (1959).

rection factor is seen to be nearly unity, with slightly more than 90% of the neutrons carried forward from the light fragments and something like 85% from the heavy fragments. The corrected distribution $\bar{n}_c(A)$ is obtained from the relation

$$\bar{n}_c(A)=\{G_+(M-A)[\bar{n}_f(A)+\bar{n}_f(M-A)]-\bar{n}_f(M-A)\}/\{G_+(M-A)+G_+(A)-1\},$$

where $M=252$ is the mass number of the fissioning nucleus. Furthermore, since the neutron-detection efficiency is not expected to be strongly energy dependent and therefore is expected to be nearly independent of the fragment mass, the corrected data may with some confidence be normalized to the measured average number of neutrons per fission for all fission modes, $\bar{\nu}=3.86$ (reference 4).

The result of these corrections is shown in Fig. 4, which gives the average number of neutrons per fragment $\bar{\nu}$ as a function of the fragment mass number A. The correction for the geometry enhances slightly the discontinuity at the symmetric mass and causes the slopes of $\bar{\nu}$ *vs* A to become quite similar in the light- and heavy-fragment groups. The ratio of the average number of neutrons from the light-fragment group to the average number from the heavy group is 1.02±0.02 for the corrected data of Fig. 4 and 1.17±0.02 for the uncorrected data of Fig. 2, where the uncertainties are the statistical standard errors.

It should be noted that a very similar behavior of the average neutron-emission probability near the symmetric mass mode has been reported previously[6] for the neutron-induced fission of U^{233}. The light-to-heavy-fragment neutron ratio in this case, however, was claimed to be much larger, $\bar{\nu}_L/\bar{\nu}_H=1.24$. The dependence of the average number of neutrons (from *both* fragments) per fission on the mass ratio, moreover, was found to be qualitatively different from that observed in this and the previous Cf^{252} experiment.[1] Most suspect was the small dependence of the average number of neutrons on the total kinetic energy of the fragments reported in the U^{233} work. The neutron detectors used in the U^{233} work[6] were handicapped by relatively low detection efficiency (of the order of a few percent, rather strongly dependent on neutron energy), high low-energy cutoff (~0.38 Mev), and small solid angle.

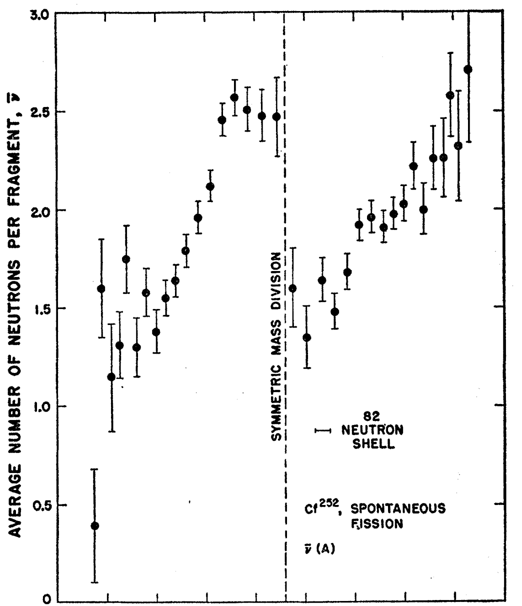

FIG. 4. The average number of neutrons per fragment as a function of the fragment mass number. Isotropic emission of the neutrons in the fragment frames has been assumed and the curve has been normalized to give $\bar{\nu}=3.86$, averaged over all fission modes. Uncertainties shown are relative standard errors.

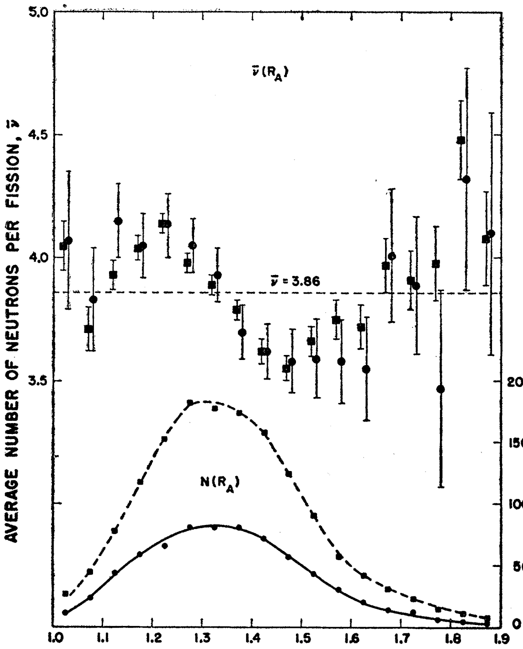

FIG. 5. The average number of neutrons per fission $\bar{\nu}$ as a function of the mass ratio R_A. Squares represent the data obtained in reference 1, where neutrons were detected from both fragments with the Cf^{252} source centered in the liquid scintillator. Circles show the result of combining the light- and heavy-fragment data of Fig. 4. Uncertainties shown are relative standard errors.

Figure 5 compares the dependence of the average number of neutrons per fission on the mass ratio of fragments found in the present experiment with that found previously.[1] The excellent agreement is held to indicate that probably no additional systematic errors of appreciable magnitude have been introduced by the change in the neutron-detection geometry.

The dispersion δA in the measurement of the mass number of a fragment is related to the relative dispersions $\delta v_L/v_L$ and $\delta v_H/v_H$ in the determination of the

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[6] J. S. Fraser and J. C. D. Milton, Phys. Rev. 93, 818 (1954).

light- and heavy-fragment velocities by the expression

$$\delta A = MR_A[(\delta v_L/v_L)^2 + (\delta v_H/v_H)^2]^{\frac{1}{2}}/[1+R_A]^2,$$

where $M = A_L + A_H = 252$ and $R_A = A_H/A_L$. A lower limit for the magnitude of δA can be obtained by first considering only the dispersive effects caused by the instrumental limitations of the time-of-flight measurements and by the emission of neutrons from the fragments. One obtains, using relations given in reference 1 and a time resolution of $(3.5 \pm 0.5) \times 10^{-9}$ sec (full width at half-maximum) $\delta A = 3.1 \pm 0.5$ mass units (lower limit). An approximate upper limit, needed because the additional dispersive effect due to the source is so difficult to determine, can be obtained by comparing the present time-of-flight data with the estimate of the true relative width of the total kinetic energy distribution of the fragments $(9.5 \pm 1.0)\%$ at $R_A = 1.35$ obtained by Milton and Fraser.[7] One obtains $\delta A = 5.7 \pm 0.4$ mass units (upper limit). A correction of the data of Fig. 2 for a mass dispersion of $5.0_{-2.0}^{+1.0}$ mass units (full width at half-maximum) will have the effect of increasing the average slope of $\bar{n}_f(A)$ in each of the fragment groups by $(11_{-7}^{+4})\%$. This correction does not alter the ratio $\bar{\nu}_L/\bar{\nu}_H$, but can increase the size of the "step down" in $\bar{\nu}$ in going from the light to the heavy group near the symmetric mass by as much as 0.2 neutron.

Since the magnitude of the dispersion in the measurement of the mass of a single fragment due to neutron emission is proportional to the square root of the sum of the numbers of neutrons emitted by *both* fragments, this dispersion is nearly constant with mass number. If there are large anisotropies in the emission of the neutrons in the frame of the fragments, this will affect the resulting perturbation of the fragment velocities. It is not believed, however, that any dispersive effect could remove the principal characteristics of the observed distribution $\bar{n}_f(A)$.

When the data are analyzed in separate 10-Mev intervals of the total fragment kinetic energy E_K, a family of nearly parallel linear curves $\bar{\nu}(A,E_K)$ is obtained, with the curves for smaller values of E_K lying higher in $\bar{\nu}$ than those for larger values. This is another example of the inverse correlation between the kinetic and excitation energies; see reference 1. The slopes of straight-line fits to the data in the intervals of A containing the majority of the events are $\partial\bar{\nu}(A,E_K)/\partial A = 0.11$ (neutrons/fission)/mass unit in the light-fragment group, and perhaps 0.04 in the heavy group, after corrections have been made for the dispersion in the mass measurement and the geometry and efficiency of the neutron detection. In the light-fragment group, these slopes are somewhat larger than the slope $\partial\bar{\nu}(A)/\partial A = 0.08$ (for all E_K) but in the heavy-fragment group not significantly different. A consideration of the weighting involved in producing $\bar{\nu}(A)$, (all E_K), due to the correlation between $\bar{E}_K$ and A, explains the decreased slope $\partial\bar{\nu}(A)/\partial A$, (all E_K), in the light-fragment group, but would predict an increased slope in the heavy group. The slopes assumed in the heavy-fragment group are not well defined by the data, perhaps due to perturbations induced by the 82-neutron shell.

IV. DISCUSSION

If the neutron-emission probability as a function of fragment mass is like that given in Fig. 4, the following remarks can be made:

1. The existence of an 82-neutron shell in primary fragments with mass numbers in the neighborhood of 133–134 may contribute to the depression in neutron emission from the heavy fragment near mass symmetry.
2. The drastic change in neutron emission at the symmetric mass should be reflected in the radiochemical mass-yield curve. In particular, the valley should be measurably distorted and deepened at mass numbers just below $A = 126$.[8]

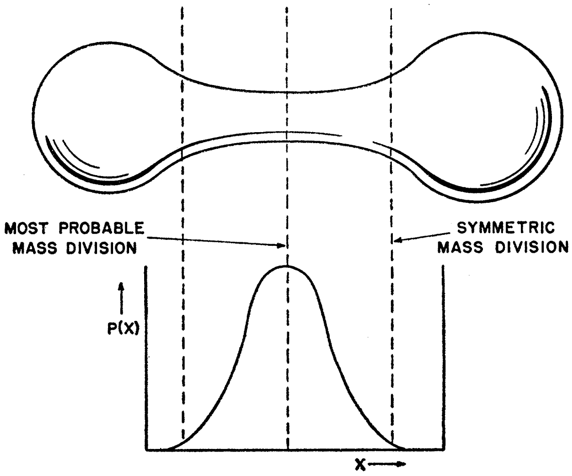

FIG. 6. A "picture" of a spontaneously fissioning nucleus of Cf^{252} shortly before it breaks in two. The relative sizes of the two ends and the neck for the "most probable configuration" were calculated using every-day classical concepts. The "most probable configuration" is defined as that partition of nucleons between ends and neck which, when split at the midpoint of the neck, will result in the most probable mass division. The number of nucleons in the neck is found to be equal to 38 (if the neck is defined as that part of the nucleus, in the most probable configuration, which must be exchangeable between the ends to reproduce the observed mass distribution). The picture is drawn for an average neck diameter equal to approximately two "nucleon diameters." A neck 1-"nucleon-diameter" thick can be four times as long. $P(x)$ indicates schematically the probability for splitting at different places along the neck. Since the neck is elongating during the time before the split, $P(x)$ is also a time-dependent function; the time dependence of which might be used to explain correlations between fragment kinetic energy and mass ratio, for example.

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[7] J. C. D. Milton and J. S. Fraser, Phys. Rev. **111**, 877 (1958).

[8] The radiochemical mass-yield data for the valley region in U^{235}, as compiled by S. Katcoff in Nucleonics **16**, No. 4, 78 (1958), although the most complete of such data available, do not appear to be adequate to indicate the presence or absence of a discontinuity in the average neutron-emission probability near the symmetric mass mode.

3. Encouraged by a recent argument[9] that fissioning nuclei may actually prefer asymmetric shapes when critically deformed, one can devise a simple model of the fission process which will reproduce both the observed mass distribution and the strange dependence of $\bar{\nu}$ on mass number. One can easily imagine that just before the fissioning nucleus breaks in two, there exists a fairly long neck[10] connecting two relatively large volumes, and that usually, if not always, these volumes are of unequal size[11] (see Fig. 6). The nucleus will be expected to break with greatest probability somewhere near the middle of the neck, which will favor the asymmetric mass divisions observed and which will partition the deformation energy of the neck fairly equally between the two fragments. Since the two ends of the nucleus would be expected to have fairly small internal excitation energies before the split, the excitation energies of the fragments after the split, and therefore the number of neutrons emitted from each fragment, should be, on the average, equal for the most probable mass division. The shape and volume of the neck can now be tailored to imply a point-of-splitting probability, such as drawn schematically in Fig. 6, which will reproduce the observed fragment mass distribution. It is obvious that symmetric mass division will correspond to the relatively very rare splitting close to the large end of the nucleus, and it is seen that this kind of a split gives almost all of the large amount of deformation energy to the light fragment. Splittings very far from mass symmetry correspond to breaking points close to the small end, with the deformation energy of the neck given to the heavy fragment. Thus the observed $\bar{\nu}(A)$ dependence is obtained. It is, of course, possible in principle for splitting to occur even closer to the large end of the nucleus than for the case of symmetric mass division, which will, in effect, reverse the roles of the ends of the nucleus, with now the heavy fragment gaining the deformation energy of the neck. It need only be assumed (for the case of Cf^{252}, at least)[12] that the probability of splitting falls off sufficiently fast as the point of symmetric mass splitting is crossed, so that the effect on the average number of neutrons emitted is small even quite close to symmetry.

Whether there be a peculiar angular anisotropy of the neutron emission or a strange division of the excitation energy between the fragments, it appears that something unexplained, and therefore interesting, is involved.

ACKNOWLEDGMENTS

In addition to the many people whose help was acknowledged in the previous work,[1] thanks are due Zane C. Motteler for the additional IBM-704 programming required.

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[9] V. V. Vladimirskii, J. Exptl. Theoret. Phys. U.S.S.R. **32**, 822 (1957) [translation: Soviet Phys. JETP **5**, 673 (1957)].

[10] See D. L. Hill, *Proceedings of the Second United Nations Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, to be published), Paper *P*/660, for an example of such a neck.

[11] It is perhaps easiest to imagine that the high probability of such configurations results from these evolving directly from shapes assumed near an asymmetric saddle-point.

[12] To extend the splitting model to other fissioning nuclides, it may be noted that the model is sensitive to the most probable mass ratio and to the probability of symmetric mass division. The most probable mass ratio largely determines the volume of the effective neck and the probability of symmetric mass division influences the sharpness of the $\bar{\nu}(A)$ transition. The "most probable configurations" (see caption of Fig. 6) for thermal-neutron-induced fission of U^{233}, U^{235}, and Pu^{239} are not appreciably different from that for Cf^{252}. However, the fact that the deformation energy of the neck seems to account for about two of the four neutrons on the average for Cf^{252} would suggest that the neutron emission from nuclides emitting only two or three neutrons on the average per fission may be almost completely determined by the "hydrodynamics" of the splitting process. In addition, the strange behavior of $\bar{\nu}$ near the symmetric mass division may be altered for fission processes more favoring symmetric mass division, since then, near symmetry, a light fragment, for example, could be formed more easily from the large end as well as from the small end of the fissioning nucleus.