

# MASS DISTRIBUTION AND KINETICS OF $U^{235}$ THERMAL-NEUTRON-INDUCED THREE-PARTICLE FISSION

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A three-parameter correlation experiment has been performed in which the energies of the two heavy fragments and of the third light particle (usually a long-range alpha particle) have been measured in coincidence. A comparison of the mass distributions thus obtained for particles from binary and ternary fission indicates that the long-range alpha particles are emitted principally at the expense of nucleons in the light fragment for near-symmetric fission and at the expense of nucleons in the heavy fragment for more asymmetric fission. Strong influence of the closed  $N=82$  and  $N=50$  shells is indicated.

Surface barrier detectors were used in conjunction with standard low-noise amplifier systems; events were serially recorded by a  $128 \times 128 \times 4$ -channel punched-paper-tape correlation recorder. The data were sorted, summed, and correlated through the use of computers, and results have been obtained giving the mass distribution, mass-energy correlations, and other kinetic parameters associated with  $U^{235}$  thermal-neutron-induced three-particle fission.

In order to assure high resolution in these measurements, a narrow, highly collimated beam of thermal neutrons from the Oak Ridge Research Reactor was incident on the  $50\text{-}\mu\text{g}/\text{cm}^2$   $U^{235}$  deposit, backed by a  $70\text{-}\mu\text{g}/\text{cm}^2$  self-supporting nickel foil. All detectors were located outside the neutron beam. The two fission fragment detectors, made of 500 ohm-cm silicon, were selected and calibrated with respect to the time-of-flight data of Milton and Fraser.<sup>1</sup> The two third-particle detectors, made of 3000 ohm-cm silicon, were operated with sensitive depths greater than the range of a 30-MeV alpha particle and were located at about  $80^\circ$  with respect to the axis of the fragment detectors.<sup>2</sup> All four detectors were  $\sim 4\text{ cm}^2$  in area and exhibited resolutions better than 40 keV full width at half maximum for natural alpha particles. The fission fragment detectors exhibited resolutions better than 1.2 MeV for 95-MeV  $Br^{79}$  and  $Br^{81}$  ions, as determined in an auxiliary experiment using the Oak Ridge tandem Van de Graaff generator.<sup>3</sup>

In the analysis of the data, the simple momentum and mass conservation relations were used:

$M_1E_1 = M_2E_2$  and  $M_1 + M_2 = 235 + 1 - 4$ . For binary fission  $M_1 + M_2 = 235 + 1$ . It can be shown that recoil from prompt neutron emission by the fragments introduces a dispersion in mass (for any individual mass split) no larger than  $\sim 2$  amu. For ternary fission the detector geometry is such that recoil from the alpha particles introduces an average dispersion (for any mass split) less than  $\sim 2$  amu. The dispersion introduced by alpha-particle recoil is thus expected to result in a slightly broadened fragment mass distribution for ternary fission.

Among the interesting results obtained from these measurements is the integral mass distribution (summed over all third-particle energies) of the fission products. This distribution is shown in Fig. 1, compared with the mass distribution of fragments from binary fission as obtained during

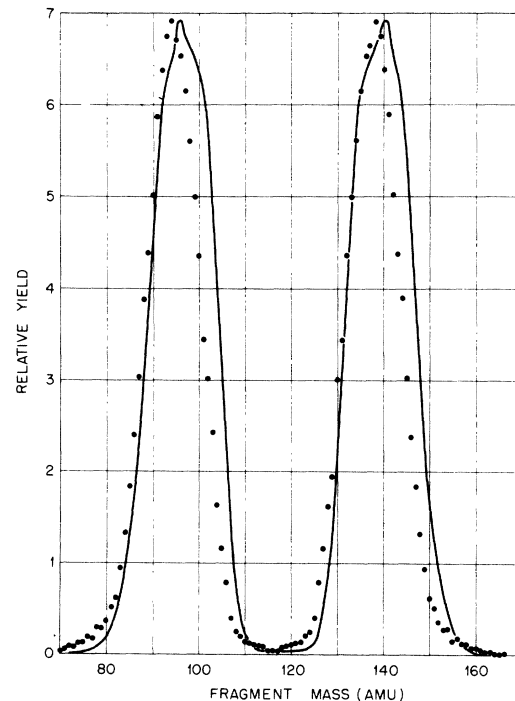


FIG. 1. Relative mass yields for fragments from  $U^{235}$  thermal-neutron-induced binary and ternary fission. Yields are normalized at the peaks. (Solid circles represent ternary fission; solid curves represent binary fission.)

the same experiment with identical source and detector conditions. The important feature of this comparison is that the low-mass sides of the two peaks are identical, within statistical uncertainties and within the dispersion mentioned above, for binary and ternary fission. The high-mass sides of the peaks in ternary fission are displaced downward with respect to the high-mass sides of the peaks in binary fission. It thus appears that the alpha particle is emitted principally at the expense of nucleons in the light fragment for near-symmetric fission and at the expense of nucleons in the heavy fragment for more asymmetric fission.

Much has been written about possible shell effects in fission.<sup>4</sup> The  $N=50$  shell occurs for masses in the 84- to 90-amu region, on the low-mass side of the light-fragment peak; the  $N=82$  shell occurs for masses in the 132- to 140-amu region, on the low-mass side of the heavy-fragment peak. The comparison shown in Fig. 1 suggests rather strongly that the nucleon clusters containing the closed shells  $N=50$  or  $N=82$  remain intact and are important both in three-particle fission and in binary fission, for  $U^{235}+n$  (thermal). In the development of the cluster model, where the principle of indistinguishability of nucleons is a key point, it has been suggested<sup>5</sup> that the formation of these clusters during the fission process (prior to scission) accounts for the asymmetry of fission in the case of low excitation energies of the compound system. This argument is perhaps better justified in the case of the  $N=82$  shell than in the case of the  $N=50$  shell; i.e., a decrease in yield is already expected for more asymmetric fission on the basis that the energy available for fission decreases rapidly as the light-fragment mass decreases beyond the  $N=50$  region.

The point of view that the  $N=50$  and  $N=82$  closed-shell nuclei play a fundamental role is consistent with recent determinations of  $\nu$ , the number of neutrons emitted from a fission product, as a function of the mass  $M$  of the product. Measurements have been made by Apalin *et al.*,<sup>6</sup> and Terrell<sup>7</sup> has calculated  $\nu(M)$  from a comparison of double time-of-flight data<sup>1,8</sup> and the radiochemical and mass-spectrometric yields tabulated by Walker.<sup>9</sup> It is found that  $\nu$  increases from 0.1 to 2.15 in the range  $85 \leq M \leq 107$ , and increases again from 0.3 to 2.3 in the range  $130 \leq M \leq 151$ . Because of the rather large statistical uncertainties, it was not possible for these authors to determine where in the range  $107 \leq M \leq 130$  the de-

crease of  $\nu$  from 2.15 to 0.3 occurs. If the above point of view is valid and shell effects are important in these considerations, the decrease should be associated with masses in the region below 132 amu, i.e., just before the  $N=82$  shell is filled, rather than with symmetric fission.

A result of further interest from the present measurements is that the mass distribution of the heavy fission products is essentially independent of long-range alpha-particle energy. The alpha-particle energy spectrum<sup>10</sup> was divided into four groups: 6 to 12 MeV, 12 to 18 MeV, 18 to 24 MeV, and 24 to 30 MeV. The fragment mass distribution for each group is shown in Fig. 2. Statistical uncertainties preclude a precise or detailed comparison of these spectra; it is evident, however, that no gross differences exist.

The average total fragment kinetic energy,  $\langle E_K \rangle = \langle E_1 + E_2 \rangle$ , as a function of mass ratio was computed for each of three alpha groups:  $6 \leq E_\alpha \leq 12$  MeV,  $12 \leq E_\alpha \leq 18$  MeV, and  $18 \leq E_\alpha \leq 24$  MeV. These are shown together with similar results for binary fission in Fig. 3. It is significant that all of the maxima in  $\langle E_K \rangle$  occur in the neighborhood of mass ratios for which the heavy fragment contains the closed  $N=82$  shell. The small rise in the binary curve at  $M_H/M_L = 1.6$  corre-

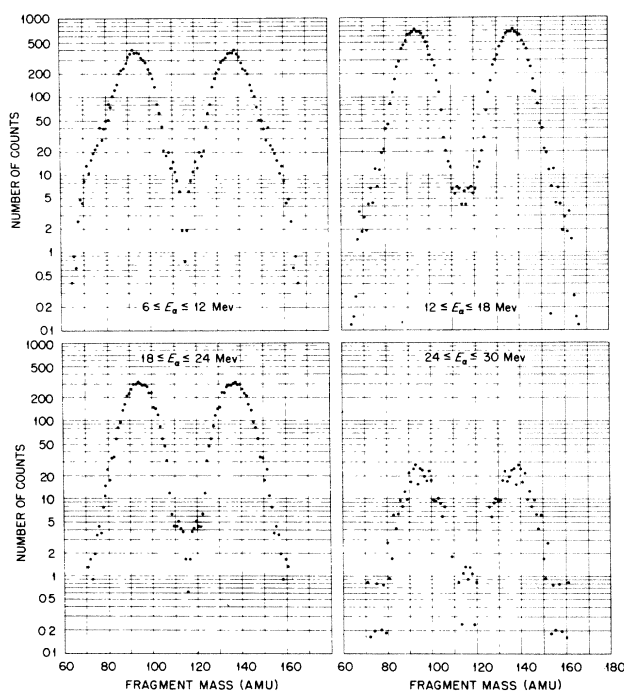


FIG. 2. Fission fragment mass yield as a function of third-particle energy for  $U^{235}$  thermal-neutron-induced ternary fission.

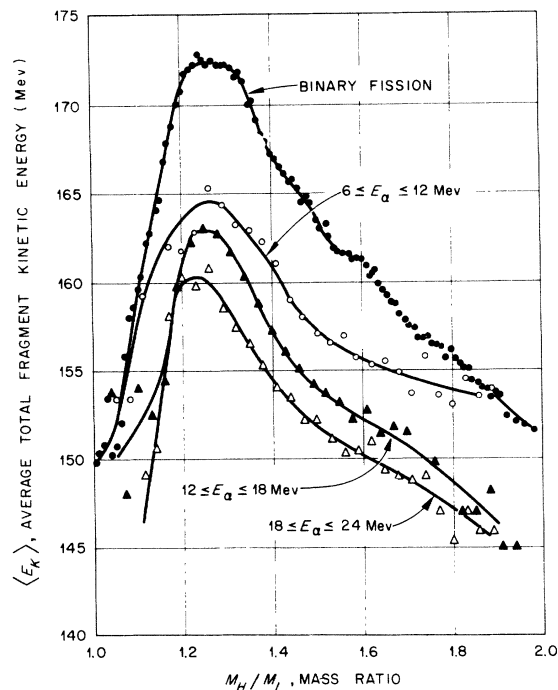


FIG. 3. Average total fragment kinetic energy as a function of mass ratio. Results for three groups of alpha-particle energies in ternary fission are compared with those for binary fission. Note that  $E_K$  is the sum of the fragment energies only, and does not include  $E_\alpha$ .

sponds to a light-fragment mass of 90 amu, which contains the closed  $N=50$  shell. The total fragment kinetic energy averaged over all mass ra-

tios and over all alpha-particle energies was found to be  $155 \pm 2$  MeV, in slight disagreement with the value of 150 MeV reported for ternary fission by Schröder *et al.*<sup>11</sup>

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<sup>1</sup>J. C. D. Milton and J. S. Fraser (to be published).

<sup>2</sup>For a discussion of the angular distribution of the long-range alpha particles, see E. W. Titterton, *Nature* **168**, 590 (1951).

<sup>3</sup>The authors very gratefully acknowledge the collaboration of C. D. Moak in the bromine-ion experiment.

<sup>4</sup>See, for example, I. Halpern, *Ann. Rev. Nuclear Sci.* **9**, 245 (1959); M. G. Mayer and J. H. D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley & Sons, Inc., New York, 1955).

<sup>5</sup>K. Wildermuth and H. Faissner, *Proceedings of the International Conference on Nuclear Structure, Kingston, 1960*, edited by D. A. Bromley and E. W. Vogt (University of Toronto Press, Toronto, Canada, 1960), p. 972.

<sup>6</sup>V. F. Apalin, Yu. P. Dobrynin, V. P. Zakharova, I. E. Kutikov, and L. A. Mikaelyan, *Atomn. Energ.* **8**, 15 (1960) [translation: *J. Atomic Energy (U.S.S.R.)* **8**, 10 (1961)].

<sup>7</sup>J. Terrell, *Phys. Rev.* **127**, 880 (1962).

<sup>8</sup>W. E. Stein, *Phys. Rev.* **108**, 94 (1957).

<sup>9</sup>W. H. Walker, Chalk River Report CRRP-913, Atomic Energy of Canada Limited, Chalk River, Ontario, 1960 (unpublished).

<sup>10</sup>See, for example, R. A. Nobles, *Phys. Rev.* **126**, 1508 (1962).

<sup>11</sup>I. G. Schröder, J. A. Moore, and A. J. Deruytter, *Bull. Am. Phys. Soc.* **7**, 304 (1962).

## CROSS-SECTION PROBABILITY DISTRIBUTIONS\*

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The purpose of this note is to introduce the notion of cross-section probability distributions and to give an example.

Energy-averaged cross sections are well known and form the basis of the optical model concept. Cross-section dispersions were first considered under another guise<sup>1</sup> and were subsequently emphasized by Thomas<sup>2</sup> when he derived a simple connection between the dispersion of the total cross section and the fluctuation cross section. Cross-section correlation coefficients were introduced at a later date.<sup>3</sup> It is, of course, in principle, possible to compute higher and higher

moments and in this way to define a cross-section probability distribution.

In order to clarify what we mean, we present a simple example. The example we choose is that of the completely soluble "picket fence" model in which the spacings between and widths of fine structure levels are assumed to be constant. We confine our discussion to  $S$  waves; in this case the relevant logarithmic derivative can be exhibited exactly by

$$f(E) = \frac{1}{\pi s} \cot \left[ \frac{\pi}{D} (E + i \frac{1}{2} \Gamma_r) \right], \quad (1)$$