Fragment Mass Distributions for Thermal-Neutron-Induced Fission of Pu^{239} and Pu^{241}

F. J. WALTER, H. W. SCHMITT, AND J. H. NEILER*
Oak Ridge National Laboratory, Oak Ridge, Tennessee
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The fragment mass distributions for thermal-neutron-induced fission of Pu^{239} and Pu^{241} have been determined from correlated energy measurements of fragment pairs. These mass distributions are compared with each other and with the post-neutron-emission mass yield for Pu^{239} obtained from radiochemical and mass spectrometric measurements. It is shown that these comparisons are consistent with the saw-tooth character of the function $\nu(M)$ as well as with the cluster model of fission in which the stability of the closed-shell structures fundamentally influences the fragment mass and energy distribution for fission at low excitation energy.

THE fragment mass distributions for thermal-neutron-induced fission of Pu^{239} and Pu^{241} have been determined from correlated energy measurements of fragment pairs. The method was generally the same as that employed in earlier experiments.[1,2] Surface barrier detectors were used in conjunction with standard low noise, charge-sensitive amplifier systems, and the resulting correlated pulse heights were recorded by a 128×128-channel punched-paper-tape correlation recorder. Special fast coincidence and inspection circuits, which are described elsewhere,[3] were used to minimize spectrum distortion by pile-up of fission fragments or natural alpha particles on fission fragments. A narrow, highly collimated beam of unfiltered neutrons from the Oak Ridge Research Reactor was incident on a 50-μg/cm^2 deposit of Pu^{239} on a 70-μg/cm^2 self-supporting nickel foil or on a 50-μg/cm^2 Pu^{241} deposit on a 30-μg/cm^2 self-supporting carbon foil. The silicon surface barrier detectors, made from 500-ohm-cm silicon, were located outside the neutron beam and were collimated with round-edge aluminum collimators. These detectors were 4 cm^2 in area and were of the same type which have exhibited resolution widths of <1.5 MeV full width at half-maximum for high-energy Br^{79}, Br^{81}, and I^{127} ions as determined in an auxiliary experiment.[4,5]

A typical single-side energy spectrum for Pu^{239} is shown in Fig. 1. The energy calibrations for the detectors were obtained for Pu^{239} by normalization of the average pulse height for each fragment group (light and heavy) to the appropriate average fragment energy obtained from time-of-flight data,[6] corrected for neutron emission. The fragment pulse height versus energy relation thus derived for each detector was assumed to apply also to fragments from the fission of Pu^{241}.

For each experiment the correlated pulse heights, serially recorded on punched paper tape, were sorted and summed into a 128×128-channel matrix. Energy calibrations were applied and appropriate transformations of the data (assuming momentum and mass conservation; $M_1E_1=M_2E_2$, $M_1+M_2=240$ or 242) were made in order to derive the fragment mass distribution. The mass distribution thus obtained (without corrections for neutron emission) is essentially the pre-neutron-emission mass distribution. The results for Pu^{239} are shown in Fig. 2. The mass yields obtained from radiochemical and mass spectrometric measurements, as compiled by Katcoff,[7] are plotted for comparison. These are post-neutron-emission mass yields and are expected to produce a mass distribution curve shifted to slightly lower masses, as indeed is observed. The shift indicated in Fig. 2 is greater for the light fragment than for the heavy fragment in near-symmetric fission, and is greater for the heavy fragment than for the light fragment in very asymmetric fission.

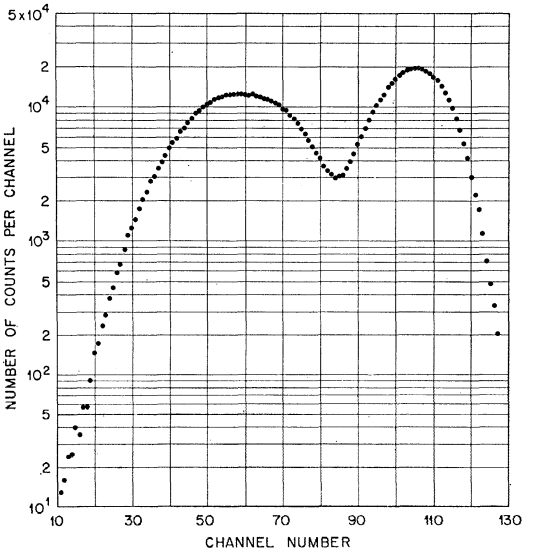

FIG. 1. Pulse-height spectrum for fission fragments from Pu^{239} thermal-neutron-induced fission.

* Present address: Oak Ridge Technical Enterprises Corporation, Oak Ridge, Tennessee.

[1] H. W. Schmitt, J. H. Neiler, F. J. Walter, and A. Chetham-Strode, Phys. Rev. Letters **9**, 427 (1962).

[2] W. M. Gibson, T. D. Thomas, and G. L. Miller, Phys. Rev. Letters **7**, 65 (1961).

[3] C. W. Williams, H. W. Schmitt, F. J. Walter, and J. H. Neiler, Nucl. Instr. Methods (to be published).

[4] C. D. Moak, J. H. Neiler, H. W. Schmitt, F. J. Walter, and G. F. Wells, Rev. Sci. Instr. **34**, 853 (1963).

[5] F. J. Walter, C. D. Moak, J. H. Neiler, H. W. Schmitt, W. M. Gibson, and T. D. Thomas, Bull. Am. Phys. Soc. **8**, 39 (1963).

[6] J. C. D. Milton and J. S. Fraser, Can. J. Phys. **40**, 1626 (1962).

[7] S. Katcoff, Nucleonics **18**, 201 (1960).

This result is consistent with the saw-tooth character of the function $\nu(M)$, the number of neutrons emitted as a function of fragment mass, as discussed by Terrell.[8]

In Fig. 3, the Pu^{239} mass distribution from this experiment is compared to the time-of-flight results of Milton and Fraser.[6] The two yields are normalized at the peaks of the distribution in order to allow comparison of shapes.

The mass distribution for Pu^{241}, obtained in the present experiment, is shown in Fig. 4. In addition, the observed mass distribution of Pu^{239}, normalized at the peak of the distribution for comparison of shapes, is plotted. It is of interest that the shapes of the mass distributions are similar, although the observed peak-to-valley ratios for Pu^{239} and Pu^{241} are 130 and 270, respectively. Perhaps of greater importance is the

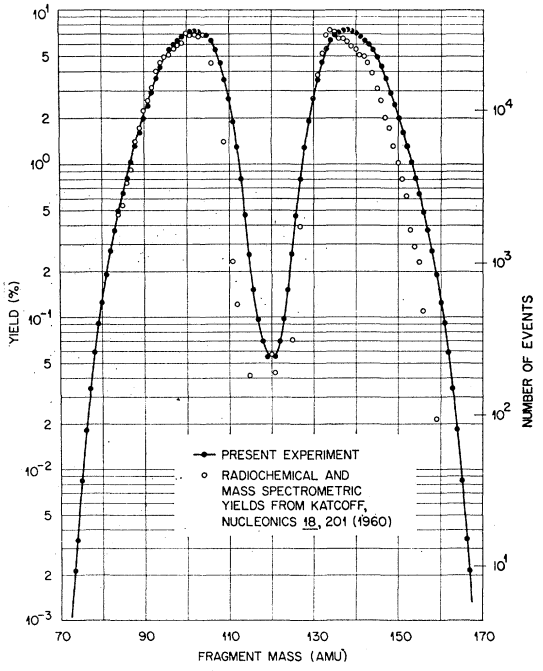

FIG. 2. Fission fragment mass yield for Pu^{239} thermal-neutron-induced fission.

observation (from Fig. 4) that the two-neutron difference (242−240) in the compound system appears predominantly in the light fragment for near-symmetric fission, and appears to be shared between the light and heavy fragments for more asymmetric fission. As with the 3-particle fission result,[1] this result is quite consistent with the cluster model of fission,[9] in which the stability of the closed-shell clusters fundamentally influences the fragment mass and energy distributions in fission at low excitation energy. The formation of

[8] J. Terrell, Phys. Rev. **127**, 880 (1962).

[9] K. Wildermuth and H. Faissner, *Proceedings of the International Conference on Nuclear Structure, Kingston,* edited by D. A. Bromley and E. W. Vogt (The University of Toronto Press, Toronto, Canada, 1960), p. 972; Phys. Letters **2**, 212 (1962).

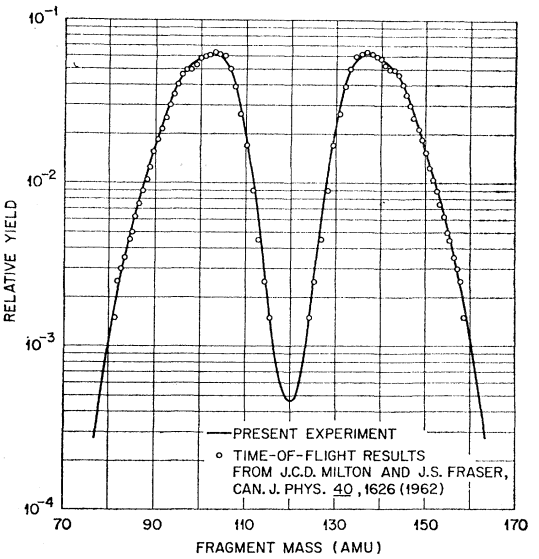

FIG. 3. Comparison of time-of-flight and correlated energy measurements of the fragment mass yield for Pu^{239} thermal-neutron-induced fission.

heavy fragments containing a minimum of 50 protons and/or 82 neutrons is energetically favored; hence, it is reasonable that the low-mass edge of the heavy-fragment peak occurs in the mass range 128–135 amu, where $Z\cong 50$ and/or $N\cong 82$. This occurs both for Pu^{239} and for Pu^{241} thermal-neutron-induced fission, as shown in Fig. 4.

In the case of very asymmetric fission, for both plutonium isotopes, both fragments contain more neutrons than are contained in the next-lowest closed neutron shell ($N=50$ for the light fragment, $N=82$ for

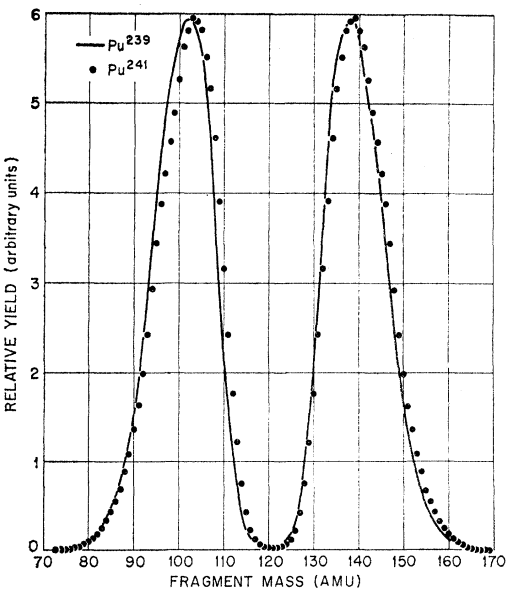

FIG. 4. Comparison of fission fragment mass distributions for thermal-neutron-induced fission of Pu^{239} and Pu^{241}.

the heavy fragment). On this basis it is reasonable that the two-neutron difference in the compound system be shared between the very asymmetric fragments, as is observed.

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Q Value for the $B^{10}(He^3,N^{12})n$ Reaction by Magnetic Analysis*

T. R. Fisher† and W. Whaling

California Institute of Technology, Pasadena, California

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The Q value for the reaction $B^{10}(He^3,N^{12})n$ has been measured by observing the $(N^{12})^{5+}$ recoil ions in a magnetic spectrometer at 10-MeV bombarding energy. Observations at 1.7° and 7.0° laboratory angle yield $Q=1.570\pm0.025$ MeV, based on the ThC′ alpha line at 8.7841 MeV. The N^{12}-C^{12} mass difference computed from this Q value corresponds to an endpoint energy of 16.320 MeV for the N^{12} beta decay and a ft value of $(1.29\pm0.02)\times10^4$. The ratio $ft(N^{12})/ft(B^{12})$ becomes 1.10 ± 0.02.

I. INTRODUCTION

RECENT interest in the beta spectra of B^{12} and N^{12} has stimulated remeasurements of the ft values for these decays.[1,2] Our knowledge of the ft value for N^{12} is limited by the uncertainty in the end point energy of this decay, which can be calculated from the N^{12}-C^{12} mass difference. This mass difference is determined from a cycle of nuclear reaction energies, in which the least certain link is the $B^{10}(He^3,n)N^{12}$ Q value determined by neutron energy measurements in emulsions.[3] We have remeasured this Q value using a magnetic spectrometer to measure the energy of the recoil N^{12} ions.

II. EXPERIMENTAL METHOD

The ONR-CIT tandem accelerator provided a $(He^3)^{++}$ beam of energy 10.009 ± 0.006 MeV. The uncertainty in the beam energy arises from the width of the entrance and exit slits, both of 0.2032-cm full width, on the 90° beam analyzing magnet of 86.36-cm radius. The beam from the tandem was passed through crossed electric and magnetic fields to filter out a weak heavy ion contaminant in the He^3 beam.

The B^{10} target was an unsupported foil of metal enriched to 94% B^{10}. To prepare these foils, a 400-Å layer of $BaCl_2$ was evaporated onto a clean glass microscope slide, and a layer of boron was deposited on this substrate by vacuum evaporation from a W boat. The boron foils were floated off on water and picked up on tantalum frames to expose an unsupported area of 0.6350-cm diameter. Analysis of the target composition by elastic proton scattering shows these foils to contain about 75% B atoms by number, with carbon and oxygen the principal contaminants.

Only trace amounts of tungsten, less than 0.5% by number of atoms, were evaporated along with the boron. The 5.3-MeV N^{12} ions were observed to lose 114 keV in passing through the foil used in the Q-value measurement, from which we estimate the energy loss of the 10-MeV He^3 beam in the foil to be 4.4 keV.

The 180° double-focusing magnetic spectrometer of 60.96-cm radius was set at 0° to observe the recoil N^{12} ions. Horizontal and vertical slits at the entrance of the spectrometer defined a square aperture with sides displaced 2.0° from the beam axis, and a square beam catcher with sides 1.1° off the beam axis prevented the He^3 beam from entering the spectrometer and permitted the usual integration of the beam current. The average angle of observation computed for this entrance aperture between the square at 1.1° and square at 2.0° is 1.73°.

The particle groups emerging from the spectrometer were detected in a Au-Si surface barrier counter located behind a 0.3175-cm slit in the focal plane. The detector output was fed into a 100-channel pulse-height analyzer, and a typical pulse spectrum is shown in Fig. 1. The energy of a particle group, determined to better than 4% from the pulse height in Fig. 1, together with the magnetic rigidity fixed by the spectrometer, provides

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† Present address: Physics Department, Stanford University, Stanford, California.

[1] R. W. Peterson and N. W. Glass, Phys. Rev. **130**, 292 (1963).

[2] T. R. Fisher, Phys. Rev. **130**, 2388 (1963).

[3] F. Ajzenberg-Selove, M. L. Bullock, and E. Almqvist, Phys. Rev. **108**, 1284 (1957).