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Note on the Prompt-Fission-Neutron Spectra of ^{235}U and ^{239}Pu*

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ABSTRACT

The prompt-fission-neutron spectra of ^{235}U and ^{239}Pu were experimentally studied using fast time-of-flight techniques. The ratio of the average-prompt-fission-neutron energy of ^{239}Pu to that of ^{235}U, deduced from measurements to 8.0 MeV, was 1.075 $\pm \lesssim$ 2.0%. The ^{235}U spectrum, measured to 1.6 MeV, was consistent with a Maxwellian distribution having the "accepted" temperature of $\sim$ 1.3 MeV. Both the ratio and spectral values were generally consistent with previously reported microscopic results.

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I. INTRODUCTION

Some recent macroscopic and spectrum-averaged measurements and some analyses of fast critical experiments indicate an uncertain knowledge of the prompt-fission-neutron spectrum.[1-6] It has been suggested that the average energy of the prompt-fission-neutron spectrum of ^{235}U is ten or more percent harder than previously deduced from a number of microscopic measurements[7-13] and that the average energy of the ^{239}Pu spectrum is appreciably nearer that of the ^{235}U spectrum than indicated by many microscopic measurements. The affect of such fission spectrum uncertainties on fast-reactor parameters has been extensively studied[4] and efforts made to identify possible uncertainties in either macroscopic or microscopic results.[14]

The microscopic experiments reported herein had the limited objectives of:

A. Testing the validity of the previously reported microscopic prompt-fission-neutron spectrum of ^{235}U and providing a basis for comparison with macroscopic results. For this purpose a measurement of the lower ($\leq$ 1.6 MeV) energy portion of the spectrum was carried out. This is an energy region where the microscopic and some of the macroscopic results differ by appreciable amounts. Furthermore, it is an energy region amenable to careful microscopic study.

B. Determining the ratio of the average prompt-fission-neutron energy of ^{239}Pu to that of ^{235}U with sufficient precision to well define discrepancies between the values deduced from macroscopic and microscopic measurements.

Generally, the experimental rationale was the definition or the resolution of discrepancy rather than definitive and comprehensive spectral study.

II. EXPERIMENTAL METHOD

All of the measurements employed pulsed-beam time-of-flight techniques assuming fission-neutron emission was prompt relative to the few nano-second resolution of the apparatus.[15] Measurements were concurrently made at eight angles with respect to the incident neutron beam distributed between 25 and 155 deg. Neutrons of energies 35 $\pm$ 12 or 400 $\pm$ 25 keV were incident on the fissile samples in bursts of $\sim$ 1 nsec duration. Proton-recoil scintillation detectors were placed about 200 cm. from the fission samples and biased at two levels; "high" (cut-off 250-400 keV) and "low" (cut-off $\lesssim$ 100 keV). The fissile samples were right cylinders 2 cm high and 2 cm in diameter fabricated for material of $\gtrsim$ 94% isotopic purity. The experimental fission-neutron velocity resolution was $\sim$ 1.5 nsec/m. Generally, the observed neutron time distributions were reasonably well defined as illustrated in Fig. 1.

The neutron sensitivity of each detector was calibrated to energies of 1.6 MeV relative to the differential elastic scattering of carbon.[16] The calibration procedure included corrections for neutron attenuation and multiple scattering within the samples and for the finite angular definition of the apparatus. The time scale was determined using calibrated time markers and was verified by observation of neutrons of known incident energy elastically and inelastically scattered from well understood nuclei (for example, the excitation of the 845 keV state in ^{56}Fe). Time-zero was referenced to the observed prompt-fission gamma-ray. Backgrounds were determined from time intervals forbidden to fission neutrons by time or bias considerations. Generally, backgrounds were small relative to the exstrema of the neutron distributions but became increasingly important as the energy increased and finally approached the fission neutron intensity at 7 to 8 MeV. The signal-to-background ratio thus limited the upper energy of the measurements.

Further details of the time-of-flight method and of the specific apparatus employed here are described elsewhere.[17]

III. EXPERIMENTAL RESULTS

A. Ratio of Average-Fission-Neutron Energy of ^{239}Pu to that of ^{235}U

The relative ratio of ^{239}Pu fission neutrons to those of ^{235}U was determined as a function of fission-neutron energy from ∿ 0.3 to ∿ 8.0 MeV. The measured time distributions were converted to energy distributions and averaged over intervals of ∿ 50 keV with results such as illustrated in Fig. 2. As in this example, some structure could be construed from the spectra. The structure was not consistent with respect to detector, incident-energy or spectral energy and was not judged statistically significant. The ratio of ^{239}Pu to ^{235}U neutrons was calculated from the energy spectra point-by-point. Uncertainties were calculated from the respective counting statistics.

The interpretation of the measured ratios was based upon the assumption that both ^{235}U and ^{239}Pu spectra are of the form

$$N(E) \sim E^n \exp(-E/T) \qquad (1)$$

where n is a single constant for ^{235}U and ^{239}Pu and T a "temperature" which can vary with isotope. With n = 1/2, Eq. (1) becomes the Maxwellian form often used to describe the prompt-fission-neutron spectra.[9]

It follows from Eq. (1) that

$$\ln R = A + B \cdot E \qquad (2)$$

where R is the energy dependent ratio of ^{239}Pu to ^{235}U fission neutrons and A and B constants. Further, B can be expressed as a function of the "temperature" (T of Eq. (1)) of the ^{235}U and the ^{239}Pu spectra by the form

$$B = \frac{1}{T(235)} \left(\frac{T(235)}{T(239)}\right) - 1 \qquad (3)$$

where T(235) and T(239) refer, respectively to ^{235}U and ^{239}Pu temperatures.

A and B of Eq. (2) were determined from a least-squares fit to the measured R distributions. Individual datum points were weighted inversely proportional to the square of their respective standard deviations. A typical fit to an experimental distribution is shown in Fig. 3. All R distributions obtained at differing incident neutron energies, with different detectors and at different biases were treated independently. Not all detectors responded identically with some giving what were judged to be "exceptional" (good or bad) distributions particularly with respect to backgrounds. These differing qualities are indicated by the subjective weighting factors (wt) assigned to each individual result in Table I. In one instance, a detector simply failed and its result was omitted. Using these weighting factors the average $\overline{B}$ values for a given incident energy and bias group were calculated from all eight detectors and an RMS deviation from the average determined with the results shown in Table I. Generally, the best estimated signal-to-background ratio was obtained at "high" biases (bias 1 of Table I) and an incident neutron energy of 35 keV. The poorest signal-to-background ratio was associated with "low" biases at a 35 keV incident energy. The quality of the "high" bias 400 keV incident energy results was intermediate between the above two. These subjective judgements of quality were apparently born out by the calculated RMS deviations. The above weighting procedure did not lead to results appreciably different from those obtained with a simple average.

The ratio of "temperatures," $\frac{T(239)}{T(235)}$, and average fission-neutron energies, $\frac{\overline{E}(239)}{\overline{E}(235)}$, follows directly from Eq. (3). Using a value of $T(235) = 1.297$ MeV[10] and the $\overline{B}$ values derived from the measurements the ratios $\overline{R} = \frac{\overline{E}(239)}{\overline{E}(235)}$ were calculated with the results given in Table I. Relatively large changes in the reference T(235) value had only a small effect on $\overline{R}$ (a 10% shift in T changes $\overline{R}$ by about 1%). The individual $\overline{B}$ values were further combined, weighting each by the inverse of its RMS deviation, to obtain a "Grand" average $\overline{B} = -0.0543$. This $\overline{B}$ value leads to $\overline{R} = \frac{\overline{E}(239)}{\overline{E}(235)} = 1.075$ with an estimated uncertainty of 1.5 - 2.0%. No significant dependence of $\overline{R}$ on incident neutron energy was noted over the limited incident-neutron-energy range of the present experiments.

B. ^{235}U Fission-Neutron-Spectrum to Energies of 1.6 MeV.

The calibrated response of the detectors was used to deduce the shape of the ^{235}U prompt-fission-neutron spectrum to fission-neutron energies of 1.6 MeV. Results typical of a number of measurements are indicated by the solid datum points shown in the upper portion of Fig. 4. A numerical measure of the spectral shape was determined assuming the distributions were of Maxwellian form. Each measured distribution was fitted, by the method of least-squares with the expression

$$\ln \frac{N(E)}{\sqrt{E}} \approx \alpha + \beta E \qquad (4)$$

where β is the inverse of the Maxwellian "temperature." One such fit is indicated by the curve in the upper portion of Fig. 4. The subjectively weighted average beta value obtained from all of the measurements was $\beta = -0.710 \pm 0.070$ MeV^{-1}. With no judgement of quality the uncertainty was increased by about a factor of two. There was no observable dependence of beta on incident neutron energy ($\sim$ 35 keV and $\sim$ 400 keV incident-neutron energies). The microscopic data of Barnard et al.[10] was interpreted in an identical manner and over the same energy interval as employed in the present work. Their results and the interpretation, indicated by the open datum points and curve in the center of Fig. 4, led to a value of $\beta = -0.737$ MeV^{-1}. The beta derived from the Barnard data and that of the present experiment imply "temperatures" somewhat larger than usually deduced from a wider energy range.[10] No significance is attached to this difference due to the small energy range employed (∿ 0.3 to 1.6 MeV). The present beta values and associated uncertainties are consistent with an "accepted" microscopic temperature of ∿ 1.297 MeV.[10]

The present results to 1.6 MeV are in contrast to those implied by McElroy[5,6] from fission-averaged cross-section measurements, primarily those of Grundl and of Fabry.[1,2,3] McElroy arrives at a prompt-fission-neutron spectra indicated by the shaded band in the lower portion of Fig. 4. Though relatively wide, this band appears qualitatively different from either of the microscopic spectra shown in the upper portion of the figure. The upper and lower limits of the McElroy band spectrum are reasonably, though not strictly, described by Maxwellian distributions as indicated by the curves in Fig. 4. These curves were obtained using fitting procedures identical to those employed in the interpretation of the present microscopic results. The deduced beta values corresponding to the maximum and minimum limits of the McElroy band were: $\beta = -0.493\ \text{MeV}^{-1}$ and $\beta = -0.338\ \text{MeV}^{-1}$, respectively. These macroscopic β values are not consistent with; those derived from the present work even accepting the largest uncertainties, the values derived from the results of Barnard et al.,[10] nor with trends in a number of other microscopic results.[9] The microscopic distributions, optimally normalized, meet or exceed alternate limits of the macroscopic band spectrum within the energy interval 0.0 to 1.6 MeV.

IV. CONCLUDING REMARKS

The ratio of the average prompt-fission-neutron energy of ^{239}Pu to that of ^{235}U ($\bar{E}(239)/\bar{E}(235) = 1.075$) deduced from the present work is in good agreement with values obtained from a number of other microscopic experiments, particularly those of Barnard et al.[10] The present ratio is not consistent with some reported macroscopic results nor with the expression

$$\bar{E} = 0.75 + 0.65 \sqrt{\bar{\nu} + 1} \qquad (5)$$

$\bar{E}$ = average-fission-neutron energy
$\bar{\nu}$ = average number of prompt neutrons emitted/fission

deduced by Terrell from kinetic and equation-of-state considerations and comparisons with experiment. Eq. (5) leads to $d\bar{E}/d\bar{\nu}$ values approximately half those implied by the ratio resulting from the present experiments. Eq. (5) is similiarly inadequate in describing $d\bar{E}/d\bar{\nu}$ values as determined from the recent $\bar{E}(239)$ measurements of Coppola and Knitter[18] and as calculated from a number of experimental studies of ^{252}Cf prompt-fission-neutron spectra[19-21] and $\bar{\nu}$ measurements.[22] Whether these discrepancies are due to the basic inappropriateness of Eq. (5) or to the lack of suitable experimental definition of the parameters is uncertain.

The prompt-fission-neutron spectrum of ^{235}U to 1.6 MeV, as determined from the present experiments, is consistent with a Maxwellian distribution with a temperature of ∿ 1.3 MeV and is very similar to that obtained by Barnard et al. and from a number of other microscopic studies.[7-13] The present results are not in reasonable agreement with spectra implied from fission-averaged-cross section measurements.[5,6] Some possible sources of this apparent discrepancy are:

A. Systematic errors in a number of microscopic measurements.

This would be despite the appreciably different methods employed and the care taken in work such as that of the present experiments. However, it should be emphasized that the accurate determination of continuum neutron spectra over a wide energy range is not an easy problem.

B. Uncertain fidelity of spectra employed in macroscopic work.

Macroscopic measurements tend to be carried out "in cavity" where the spectrum may not be equivalent to the "in vacuo" environment usually approached in microscopic studies.

C. Difficulties in deducing spectra from measured spectral-averaged cross sections.

There may be problems associated with the microscopic cross sections employed and/or with the weighting and calculational procedures utilized.

The present experiments are illustrative of the difficulties associated with accurate microscopic fission-spectra determinations and of the relative ease and accuracy of ratio measurements. Future emphasis should be given to the accurate determination of a standard fission-neutron-spectrum, such as that of ^{252}Cf, to which both microscopic and macroscopic results can be accurately related.

TABLE I: RATIO OF AVERAGE-PROMPT ..SSION-NEUTRON ENERGY OF ^{239}PU TO THAT OF ^{235}U

Det. No.[a]	E_{in} = 35 keV, Bias 1		E_{in} = 400 keV, Bias 1		E_{in} = 35 keV, Bias 0	
	B	wt	B	wt	B	wt
1	-0.054193	4	-0.061146	3	-0.006082	1
2	-0.033642	3	-0.033916	3	-0.017869	1
3	-0.069658	3	-0.021794	2	-0.089434	2
4	-0.068393	4	-0.060907	2	-0.016193	1
5	-0.079382	2	-0.081418	2	-0.064606	3
6	-0.063139	2	-0.027285	3	-0.041101	2
7	-0.049243	3	OMIT		-0.031288	3
8	-0.070019	3	-0.062669	2	-0.083037	2
	$\overline{B}$ = -0.0601		$\overline{B}$ = 0.0483		$\overline{B}$ = -0.0503	
	RMS Dev. = 0.0136		RMS Dev. = 0.0205		RMS Dev. = 0.0302	
	$\overline{R}^b = 1.084 \pm 0.020$		$\overline{R}^b = 1.066 \pm 0.028$		$\overline{R}^b = 1.069 \pm 0.045$	
			GRAND WEIGHTED AVE. $\overline{B}$ = -0.0543			
			$\overline{R}$ = 1.075			

[a]Detectors 1 through 8 at angles with respect to the incident neutron beam of 28.5, 38.5, 53.5, 69.5, 84.5, 114.5, 129.5 and 154.5 deg., respectively.

[b]$\overline{R}$ of this table is defined as the ratio $\frac{T(239)}{T(235)} = \frac{\overline{E}(239)}{\overline{E}(235)}$; T(235) = 1.297 was assumed for calculational purposes as described in the text.

FIGURE CAPTIONS

Fig. 1 Measured time distribution of fission neutrons. The time-per-channel number, indicated on the horizontal axis, was ∿ 1.01 nsec and flight path ∿ 200 cm. Vertical bars indicate the uncertainty in the datum points inclusive of background contributions.

Fig. 2 A measured fission-neutron energy distribution from ^{235}U. The incident neutron energy ∿ 35 keV. The primary velocity distribution has been converted to an energy scale. Vertical bars indicate the uncertainties in the datum points. No corrections have been made for detector sensitivity.

Fig. 3 Linear fit to the logarithum of the measured relative ratio of the ^{239}Pu fission spectrum to that of ^{235}U. Vertical bars indicate experimental uncertainties. The incident energy was 35 keV and the detector No. 6 as defined in Table I.

Fig. 4 ^{235}U prompt-fission-neutron spectra in the interval 0.0 to 1.6 MeV. The solid datum points are illustrative of a number of results of the present work. The open circles represent the microscopic results of Barnard et al.[10] The shaded band indicates the spectra implied by McElroy[6] from macroscopic fission-averaged cross-section measurements. Solid curves are the results of fitting Maxwellian distributions to the respective values as described in the text. Normalization of the three distributions is arbitrary.

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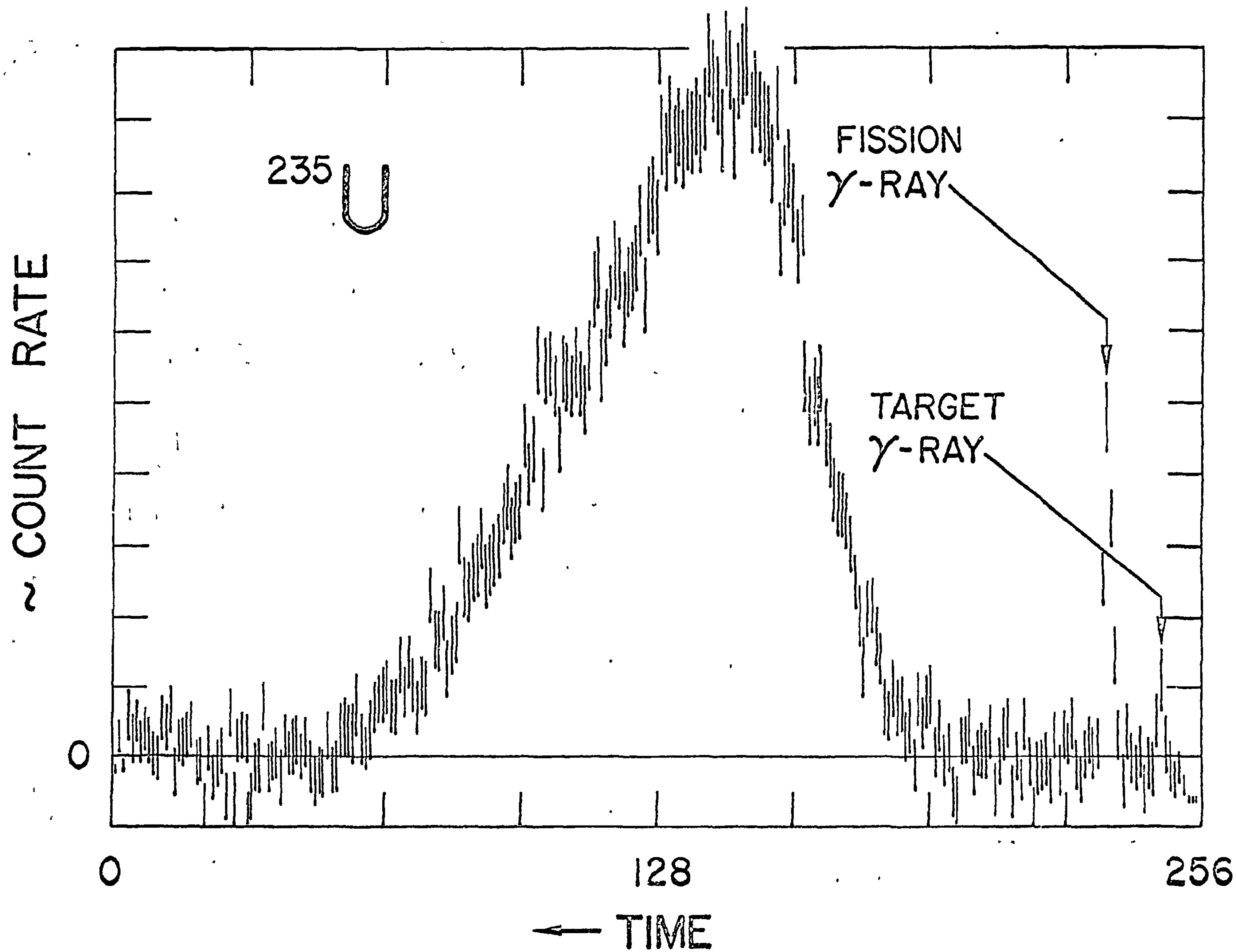
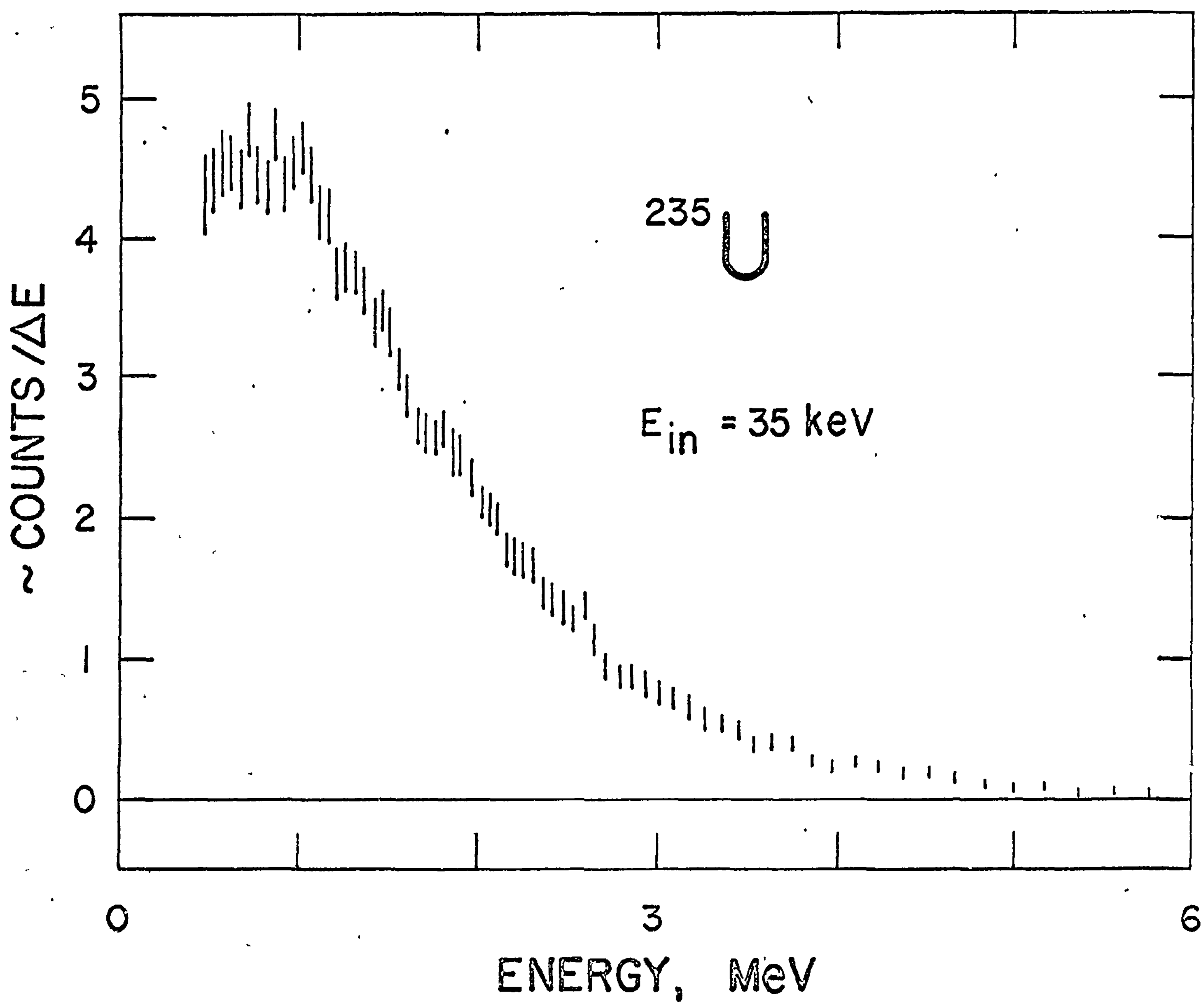
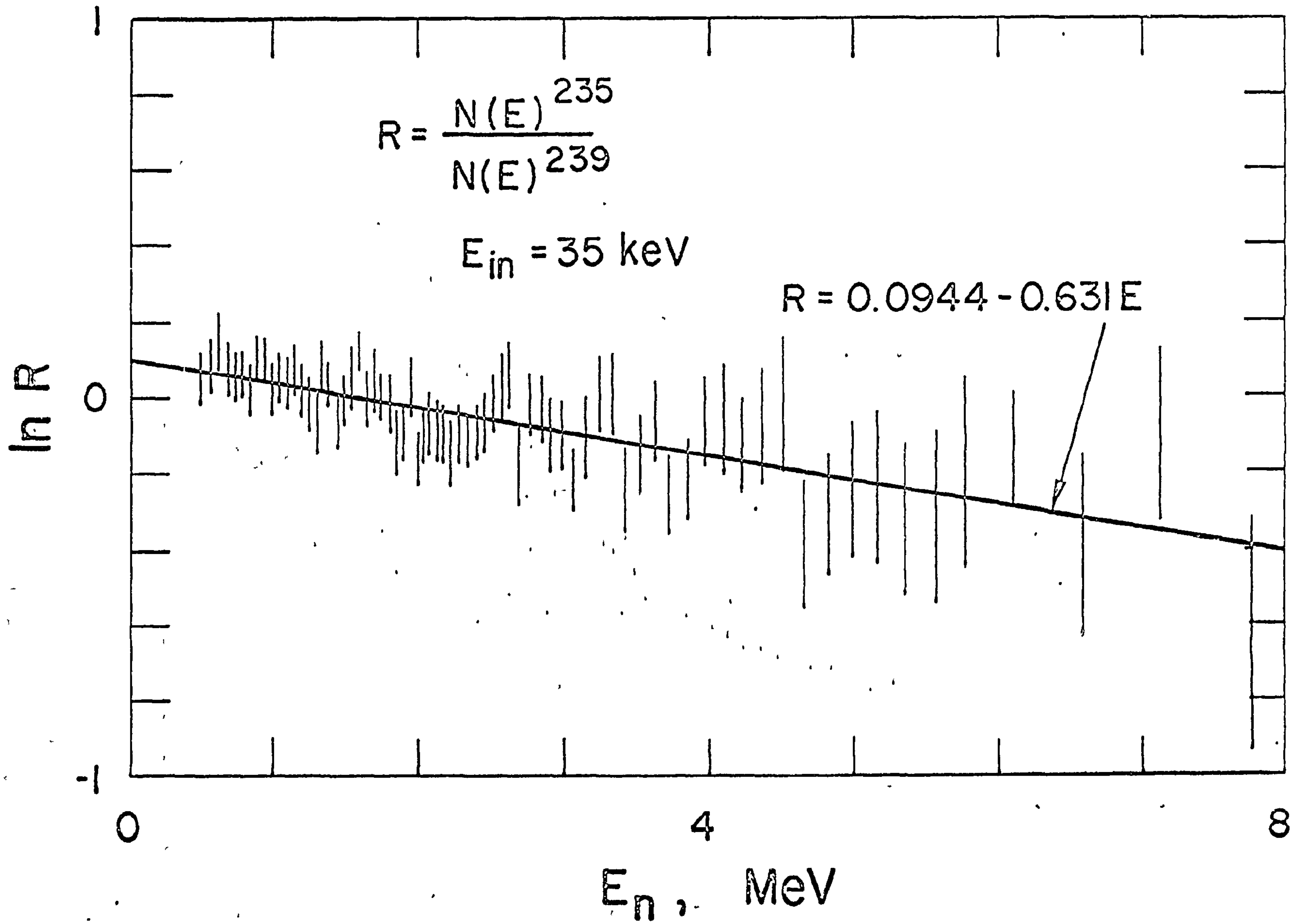
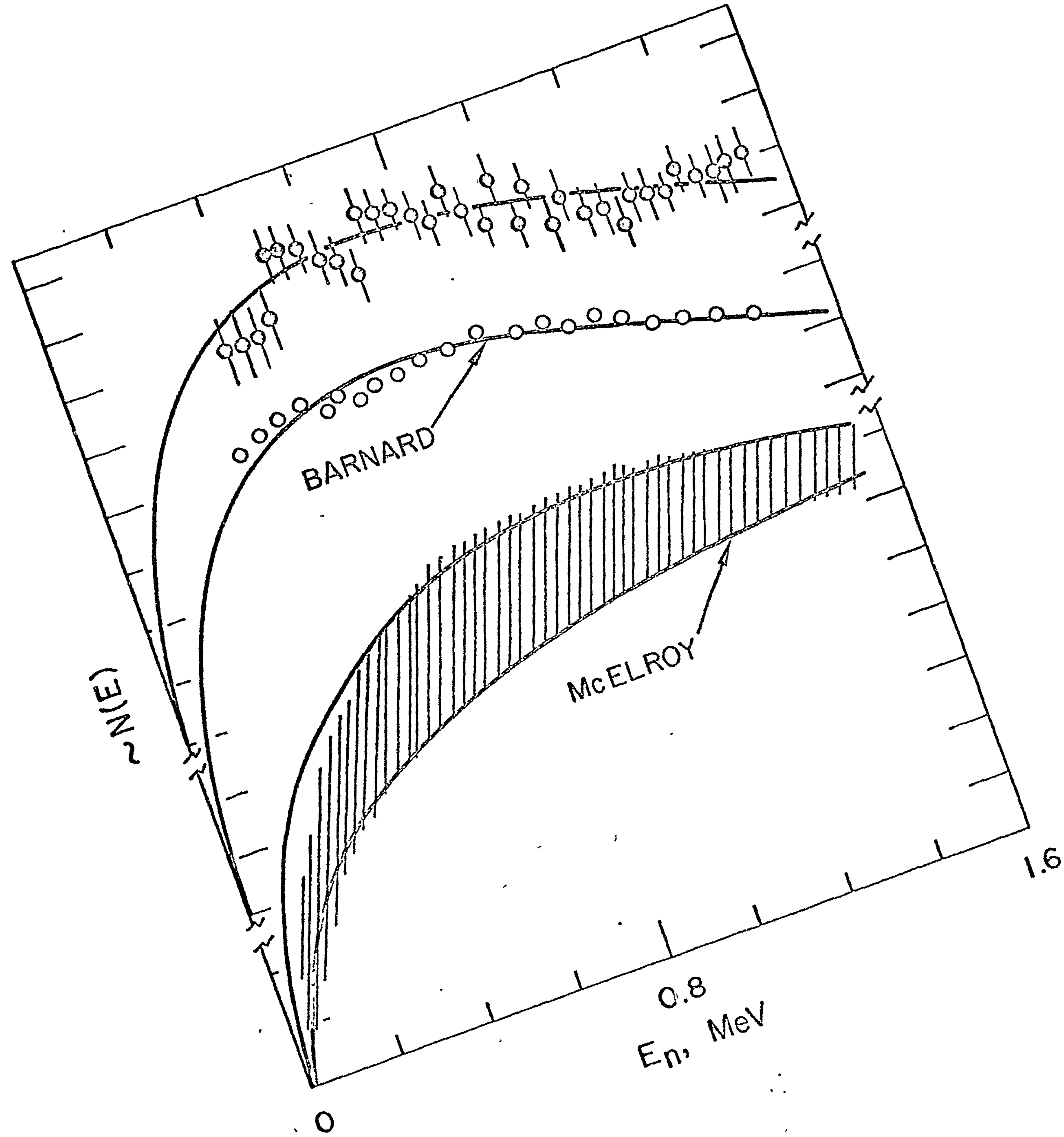


235 U
FISSION
γ-RAY
TARGET
γ-RAY
~ COUNT RATE
0
0
128
256
← TIME


~ COUNTS /ΔE
0
1
2
3
4
5
235 U
E in = 35 keV
0
3
6
ENERGY, MeV



$R = \frac{N(E)^{235}}{N(E)^{239}}$
$E_{in} = 35$ keV
R = 0.0944 - 0.631E
ln R
1
0
-1
0
4
8
E_n, MeV


~N(E)
BARNARD
McELROY
0
0.8
1.6
En, MeV