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The ^{238}U to ^{235}U Fission Cross Section Ratio from 1 to 5 MeV

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ABSTRACT

Measurements of the $^{238}U:^{235}U$ fission cross section ratio were carried out in the 2 to 3 MeV region using two different methods to determine the relative masses. The energy dependence of the ratio was measured from 1 to 5 MeV and normalized to the results at 2.5 MeV. The measured value of the ratio at 2.5 MeV is 0.432 ± .004.

I. INTRODUCTION

Accurate values of the ^{238}U:^{235}U fission cross section ratio are important for fast reactor calculations, as a spectral index in the measurement of neutron spectra, and as a check on the reliability of cross section measurements. Accuracies of the order of 1% are now being requested. However two evaluations[1,2] based on essentially the same data differ by $\sim$ 5% at 4 MeV. There are some measurements which show fair agreement[3-5] in the 2 to 3 MeV region but the results of Stein[6] lie $\sim$ 3% lower while those of Lamphere[7] are $\sim$ 3% higher.

In this experiment samples of ^{238}U and ^{235}U were placed in and perpendicular to a neutron flux in a back-to-back position and fissions were detected with similar detectors. Between 2 and 3 MeV measurements were made using samples of known relative masses. The effects of neutrons scattered from the surroundings were reduced by placing the samples close to the neutron source and by using time-of-flight methods to select suitably time correlated events. Above 3 MeV longer flight paths were used to permit separation of the lower energy neutrons produced by secondary source reactions. The optimum sample requirements at other energies made relative mass determinations difficult so these measurements were made with samples of unknown masses and the final cross section ratio was obtained by normalizing the results to the absolute measurement at 2.5 MeV.

II. EXPERIMENTAL DETAILS

A. The Neutron Source

The $^{7}Li(p,n)^{7}Be$ reaction was used as a neutron source. Targets were produced by evaporating metallic lithium on a tantalum backing. Target thicknesses were $\sim$ 200 keV (measured at the (p,n) threshold) above 2 MeV neutron energy but were only 25 keV at lower energies where the cross section ratio was changing rapidly.

Although a lithium target provides a convenient and easily prepared neutron source it is not a monoenergetic one, particularly at higher energies, as there are a number of secondary reactions in the lithium and in the supporting plate which provide groups of lower energy neutrons. Above 2.38 MeV proton energy transitions to the first excited state of ^{7}Be produce a second neutron group. The $^{7}Li(p,^{3}He\ n)^{4}He$ reaction yields a broad neutron spectrum above its threshold at 3.68 MeV but the neutron contribution from this reaction is not significant below 5 MeV. Above 5.9 MeV a few additional neutrons may be produced by ^{6}Li reactions. At any energy neutrons may be scattered by the tantalum target support plate. Since the plate is only 0.25 mm thick most of these neutrons are initially emitted near 90 deg. so they effectively form a "scattered group" with a well defined energy. A previous experiment[8] at a 0 deg. neutron energy of 260 keV gave 4 ± 1% as the intensity of this group. It is generally less at higher energies where the neutron yield is more sharply peaked toward 0 deg. The Ta(p,n) reaction has a

low threshold but in this experiment the contribution from this reaction was negligible below 6 MeV.

A neutron time-of-flight spectrum at 7 MeV proton energy is shown in Fig. 1. The first, second and scattered neutron groups are not resolved but all others are separated. The broad peak near channel 180 is primarily due to the $^{7}Li(p,^{3}He\ n)^{4}He$ reaction.

The reaction is still useful as a neutron source even if the lower energy neutron groups cannot be separated providing the relative intensities of these groups are known. The intensity ratios for the first and second groups are shown in Fig. 2. The data of Bevington et al.[9] was used below 4 MeV and an auxillary experiment was performed, using a hydrogenous detector and a 6 meter flight path, to provide ratios at higher energies. The estimated error is ± 5%.

B. Detector and Electronics

The fission detector, shown schematically in Fig. 3, was a low-mass, double ionization chamber which used methane as a counter gas. When operated at 300 V and with a plate separation of 6 mm the pulse rise time was ∿ 30 nanosec. The pulse was sent to a clipping amplifier with a clipping time of 8 nanosec. The amplifier output triggered a fast discriminator which provided the start signal for the time to pulse height converter. The stop signal came from a pick-up loop in the proton beam tube. The time converter output provided one parameter for storage in

a two parameter array. The second parameter was obtained by stretching the output of the clipping amplifier. The array dimensions were generally 32 × 128 or 16 × 256.

One of the better time spectra is shown in Fig. 1. The time resolution of the system was ∿ 3 nanosec which is much less than the clipping time. This was obtained by setting the triggering level of the fast discriminator well down into the alphas. When the data was processed those pulses that were less than ∿ 1.5 times the maximum alpha pulse were discarded. Thus all pulses with amplitudes near the triggering level were discarded and the time spread was reduced.

The detector was usually operated with the fissile deposit on the negative electrode. Figure 4 shows that the separation between the alphas and the fission fragments was very good for this mode of operation. For some measurements it was necessary to put as much material as possible into the detector so deposits were placed on both electrodes. Under these conditions the separation was rather poor but the system was stable enough to permit reliable realtive measurements.

C. Sample Preparation

The mass anlayses of all the batches of fissile materials used are listed in Table I. Samples were deposited on 0.25 mm polished stainless steel or 0.12 mm molybdenum plates by electroplating or by vacuum evaporation of UF_4. Since the evaporation technique involved a large amount of waste it was used only with the more plentiful C and F materials. At low

energies where the ^{238}U fission cross section is low high purity materials were needed to reduce the correction for other isotopes. For this reason materials A and D were used below 2 MeV. The deposit diameter was 2.5 cm and the thicknesses were $\sim$ 150 $\mu g/cm^2$ for A and $\sim$ 500 $\mu g/cm^2$ for D. Materials A and E were used for the relative measurements above 3 MeV since they were fairly pure and available in the required amounts. These deposits had a 5 cm dia., a thickness of $\sim$ 500 $\mu g/cm^2$ and each detector contained $\sim$ 20 mg.

Measurements were made between 2 and 3 MeV using deposits of known mass ratios. These were determined by the following methods.

Method 1: This method is based on the measurement of specific alpha activities. Eighteen 2.5 cm dia. deposits of material C and of material F were prepared by vacuum evaporation of UF_4. Deposit thicknesses ranged from 70 to 1000 $\mu g/cm^2$. All samples of C were counted in a low geometry alpha counter. Half the deposits were dissolved from their backings and the resulting solution analyzed colormetrically. As the activity of material F was low it was counted in a 2π alpha counter before half the deposits were analyzed colormetrically. This data determined the specific activity of C and also the apparent specific activity of F as a function of the 2π count rate. The mass of uranium in the remaining deposits was then calculated from the measured alpha rates. Since only the ratio was needed the absolute accuracy of the analytical method did not influence the result.

Method 2: This method is based on the relative thermal fission rates of a pure ^{235}U sample and of a ^{238}U sample which contains several percent ^{235}U. Deposits were prepared in pairs from materials B and G by electroplating. The deposit thicknesses ranged from $\sim$ 150 to $\sim$ 250 $\mu g/cm^2$ and in each pair were equal to within 5%. The relative thermal fission rates of each pair were measured by comparing them to a reference sample in the thermal column of the Argonne Thermal Source Reactor. At the position used, the cadmium ratio was $\sim$ 500 to 1. The relative number of uranium atoms in each pair of deposits was calculated from the thermal fission ratios and the mass analysis.

D. Experimental Procedures

Measurements in the 2 to 3 MeV region were made with samples prepared by method 1 and method 2. The detector was placed 5 cm from the neutron source and time-of-flight methods were used to reduce and measure the time independent background. At energies above 3 MeV the detector contained 5 cm dia. deposits on both electrodes and was placed 52 cm from the neutron source. At this distance the neutrons from the Ta(p,n) and $^{7}Li(p,^{3}He\ n)^{4}He$ reactions could be separated by time-of-flight. Frequent measurements were made at 2.5 MeV to test for drift or instability in the system and to provide for normalization. The lithium target thicknesses used in these measurements were 200 keV which, combined with the subtended angle, gave a total energy spread of $\sim$ 150.

The physical arrangement used below 2 MeV was similar to that used in the 2-3 MeV range. However the energy spread due to angular resolution and target thicknesses was only 40 keV and a continuous proton beam was used so that the higher current could compensate for the thinner lithium target and the lower ^{238}U fission cross section. Under these conditions it was not possible to determine the time-independent background at the same time but other measurements showed that it was small, had little energy dependence and thus did not affect a relative measurement. Measurements were also made at 2.5 and 3.08 MeV for normalization.

All measurements except those using the continuous beam, were performed twice with each set of samples, once with the ^{238}U deposit facing the neutron source and then with the ^{235}U in that position. When the results were averaged effects due to momentum of the incident neutron, geometry, and elastic scattering in the sample support plates were eliminated.

III. CORRECTIONS

A. Other Neutron Groups and Isotopes

The measured fission ratio can be written as

$$\frac{F_1}{F_1} \backsimeq \frac{N_1}{N_2} \frac{\sum_g G^g \sum_i P_{1i} \sigma_i^g/\sigma_{235}^1}{\sum_g G^g \sum_i P_{2i} \sigma_i^g/\sigma_{235}^1} \qquad (1)$$

The F_1 and F_2 are the number of fissions from detector 1 and detector 2 respectively. N is the total number of uranium atoms in a sample, G^g is the fraction of neutrons in neutron group g, P_{1i} is the atomic fraction of isotope i in detector 1, σ_i^g is the fission cross section of isotope i at the energy of neutron group g. This equation can be readily solved for $\sigma^1_{238}/\sigma^1_{235}$.

All cross section ratios were initially taken from Davey's evaluation.[1] Then the correction was recalculated using the results of this experiment for the ^{238}U to ^{235}U cross section ratio.

The use of very pure materials below 2 MeV where the ^{238}U cross section is changing rapidly made the correction about equal to the percentage of low energy neutrons. At higher energies, where the fission cross sections have little energy dependence, the effect of the second and scattered neutron groups are very small. The correction for the samples prepared by method 2 was 7.1% at 2.5 MeV but this was largely due to the 10% ^{235}U in the ^{238}U sample.

B. Neutron Scattering

Elastic scattering does not affect the results unless it occurs in the sample support plates. Otherwise each deposit is affected about equally. In order to correct for scattering in the support plates, measurements were made with the ^{238}U deposit facing the source then with the ^{235}U in that position. An average of the two results eliminated

the elastic scattering. Below 2 MeV measurements were made with only one orientation. Here it was assumed that the elastic scattering correction was independent of energy and was eliminated by the normalization.

The principal source of inelastically scattered neutrons was the counter electrodes, particularly the sample backing plates because of their proximity to the uranium deposits. The correction factor, calculated using a multigroup cross section set, was 1.006 at 2.5 MeV.

C. Pulse Height Extrapolation

This was only necessary for the absolute measurements. The correction to each spectra was 1-2% depending on deposit thickness, with an estimated error of 10%.

D. Background

The time-independent background, produced by very low energy neutrons, was insignificant in the ^{238}U detector. For the ^{235}U detector the average correction was $\sim$ 1.5% at 5 cm. distance. At 52 cm. the correction was typically 10%. The background was measured in the interval to the right of the neutron peak in Fig. 1. The width of the interval was usually 30 channels and the statistical error due to the correction was usually < 0.2%.

E. Uranium Deposit Thickness

No corrections were made for this effect. All uranium deposits used were $\leq$ 350 $\mu g/cm^2$ and each pair of deposits were about the same thickness.

F. Sample Geometry

With the 5 cm target to deposit distance the small separation between the deposits caused their geometry factors to differ by $\sim$ 2%. This was eliminated when measurements were made with both detector orientations. At low energies where only one orientation was used the difference was eliminated by the normalization.

G. Momentum Effects

These were eliminated at higher energies by averaging measurements made with both detector orientations. At low energies the effects were eliminated by the normalization since they are < 1% and proportional to $E^{\frac{1}{2}}$.

IV. ERRORS

The identified sources of error for the 2.5 MeV point are given in Table II. The values listed are standard deviations in terms of percent of the final cross section ratio. The combined error in Table II and the total uncertainty in Table IV are the root-mean-square of all identified errors.

The small error assigned to the mass ratio determined by method 2 is largely due to the small number of individual measurements involved in each determination. Only the thermal fission ratio and the mass analyses were needed and since both the thermal fission ratios and those at higher energies were measured with the same detector system a number of possible systematic errors were eliminated. The errors in the thermal fission ratio was $\sim$ 0.4% while the mass analysis introduced an additional uncertainty of $\sim$ 0.2%.

The "relative σ" refers to errors in the $\sigma_i^g/\sigma^1_{235}$ terms in Eq. (1). Since the amounts of ^{234}U and ^{236}U are very small this error is determined by the uncertainties in the energy dependence of the ^{235}U fission cross section and the ^{238}U to ^{235}U fission cross section ratio. These uncertainties were set equal to 2% for error evaluation. The contribution of "relative σ" to the error was generally less than 0.5% except near 5 MeV where the second group neutron yield is large. There it amounted to 0.85%.

The remaining errors in Table II do not show much energy dependence although the error due to other neutron groups does increase to $\sim$ 0.6% at lower energies where the ^{238}U cross section changes rapidly. The relative consistency reflects the scatter in repeated measurements that cannot be attributed to the counting statistics. The statistical error includes the extrapolation to zero pulse height and the background subtraction.

V. RESULTS AND DISCUSSION

The results of the absolute measurements are listed in Table III. The errors include only those factors contributing to the relative errors

of the two methods. These are mass ratio, counting statistics and relative consistency. Table IV lists all the results. The relative measurements are normalized to the 2.5 MeV point and the errors listed are the estimated total standard deviation. The data in Table IV is also shown in Figs. 5 and 6 together with the results of some other measurements.[3-5,6,7]

From Fig. 5 it can be seen that the results of this experiment are in good agreement with the shape of the $^{238}U:^{235}U$ ratio found by Stein et al.[6] but are ∿ 3% higher. In the 2 to 3 MeV region they are in good agreement with Jarvis,[3] White and Warner[4] and Poenitz and Armani[5] but are ∿ 4.5% below that of Lamphere.[7]

The only extensive measurements at low energies are those of Lamphere.[7] In the 2 to 3 MeV region they are ∿ 4% above the results of this experiment so they have been multiplied by 0.96 and plotted in Fig. 6 for comparison. There is a large difference in the region between 1.3 and 1.9 MeV which approaches 15% near 1.5 MeV. Stein et al.[1] also have a point here which is lower by an even greater amount. Above and below this region the agreement is fairly good. The overall agreement could be improved by an energy shift of ∿ 20 keV.

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Table I: Isotopic Composition in At. %.

Isotope	Material						
	A	B	C	D	E	F	G
234	0.028	1.034	0.856	[	<0.0014		0.114
235	99.856	98.409	93.249	6ppm	0.0141	0.415	10.0842
236	0.062	0.455	0.332	]			0.050
238	0.054	0.102	5.526	100.000	99.986	99.585	88.993

Table II: Errors for the Absolute Measurements at 2.51 MeV.

	Method I	Method II
Mass Ratio	1.0 %	0.5 %
Counting Statistics	0.35	0.35
Relative Consistency	0.36	0.36
Extrapolation	0.15	0.15
Inelastic Scattering	0.20	0.20
Other Neutron Groups	0.20	0.21
Relative σ's	0.41	0.41
Combined Error	1.22	0.89

Table III: Summary of Absolute Measurements

E_n	Method I		Method II		Average	
	Ratio	% Err*	Ratio	% Err*	Ratio	% Err*
2.00	.4105	1.12			.4105	1.12
2.51	.4328	1.17	.4317	0.70	.4320	0.60
3.08	.4332	1.15	.4360	1.04	.4347	0.77

*Errors include relative mass, statistical, and relative consistency. Uncertainties that do not contribute to the relative error of the two methods are not included.

Table IV: The ^{238}U:^{235}U Fission Cross Section Ratio

E_n (MeV)	ΔE_n (MeV)[b]	$\frac{\sigma_f\ ^{238}U}{\sigma_f\ ^{235}U}$	Total Uncertainty (%)
0.898	.038	0.0104	3.86
1.005	.040	0.0132	2.56
1.108	.041	0.0220	2.16
1.205	.041	0.0302	2.18
1.306	.042	0.0525	1.99
1.401	.042	0.156	1.64
1.514	.042	0.276	1.49
1.617	.043	0.327	1.45
1.720	.043	0.344	1.45
1.821	.044	0.391	1.44
1.914	.045	0.412	1.46
1.995	.046	0.403	1.42
2.00	.15	0.410[a]	1.23
2.51	.14	0.432[a]	0.75
3.08	.13	0.435[a]	0.92
3.28	.09	0.448	0.97
3.58	.09	0.466	0.98
4.08	.08	0.486	0.99
4.47	.08	0.499	1.51
5.08	.07	0.508	1.31
5.33	.07	0.517	1.25

[a]Absolute Measurement. Other values are normalized to these.

[b]Energy resolution.

FIGURE CAPTIONS

Fig. 1 A time spectrum from the ^{235}U detector. The first group neutron energy is 5.34 MeV (7.00 MeV proton energy). The time scale is 1.05 nanosec/channel.

Fig. 2 The ratio of the intensities of the second to first neutron groups from the $^{7}Li(p,n)^{7}Be$ reaction at 0 degrees.

Fig. 3 A schematic drawing of the fission detector.

Fig. 4 A pulse-height distribution during a pulsed measurement.

Fig. 5 The fission cross section ratio $^{238}U:^{235}U$ above 2 MeV.

Fig. 6 The fission cross section ratio $^{238}U:^{235}U$ below 2 MeV.

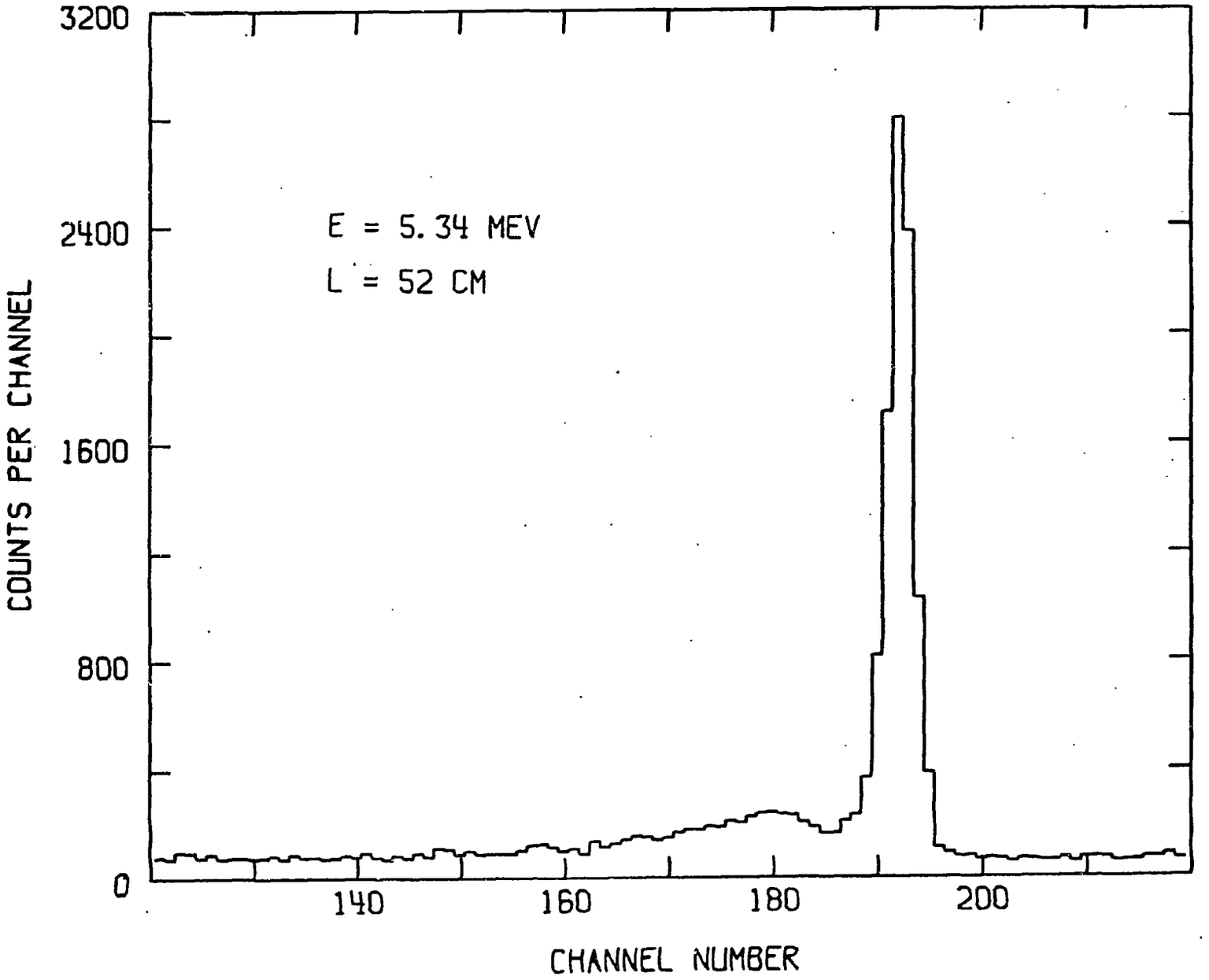
3200
2400
1600
800
0
COUNTS PER CHANNEL
E = 5.34 MEV
L = 52 CM
140
160
180
200
CHANNEL NUMBER

FIGURE - 1.

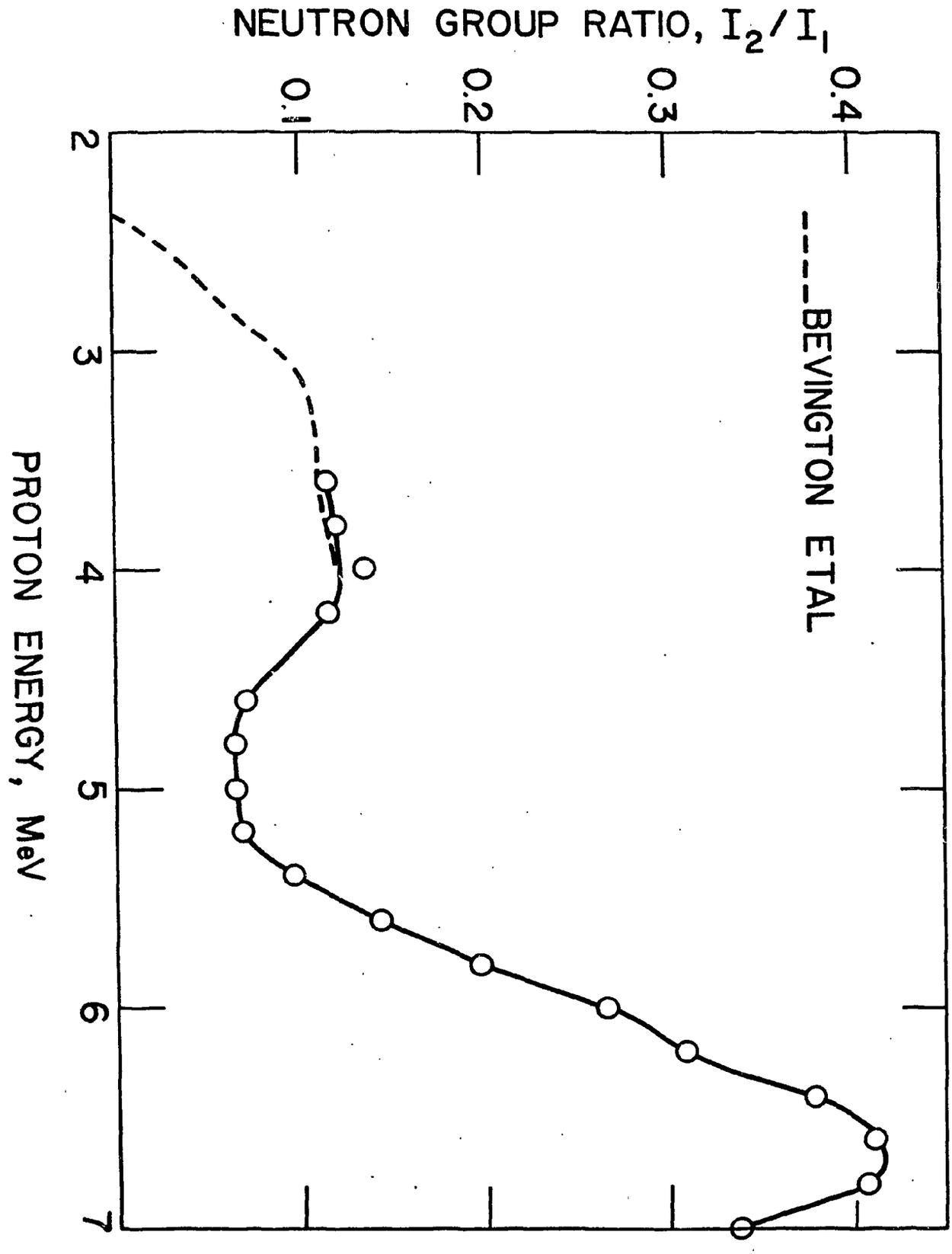
NEUTRON GROUP RATIO, I_2/I_1
0.1
0.2
0.3
0.4
2
3
4
5
6
7
PROTON ENERGY, MeV
----BEVINGTON ETAL

FIGURE - 2.

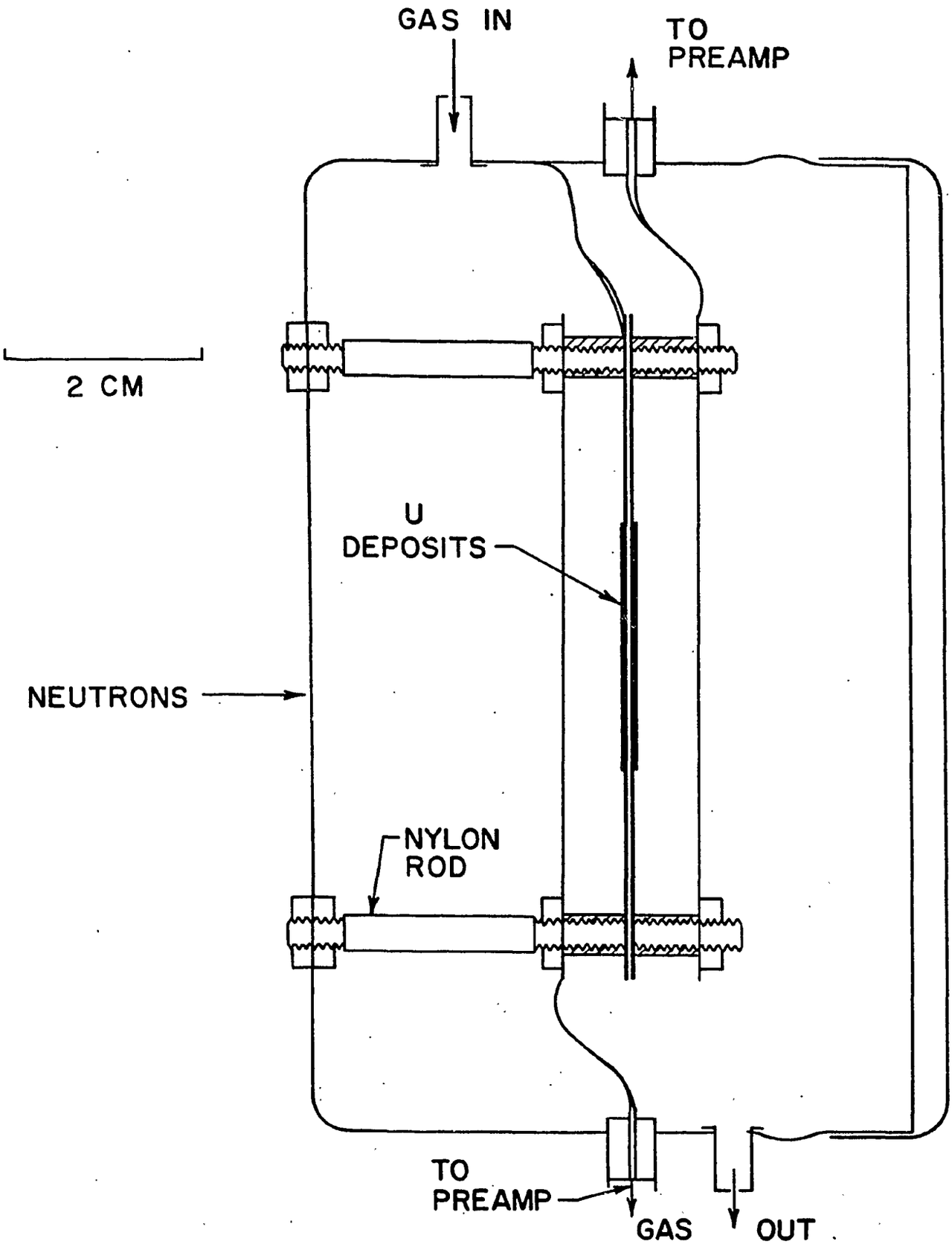
GAS IN
TO
PREAMP
2 CM
U
DEPOSITS
NEUTRONS
NYLON
ROD
TO
PREAMP
GAS
OUT

FIGURE - 3.

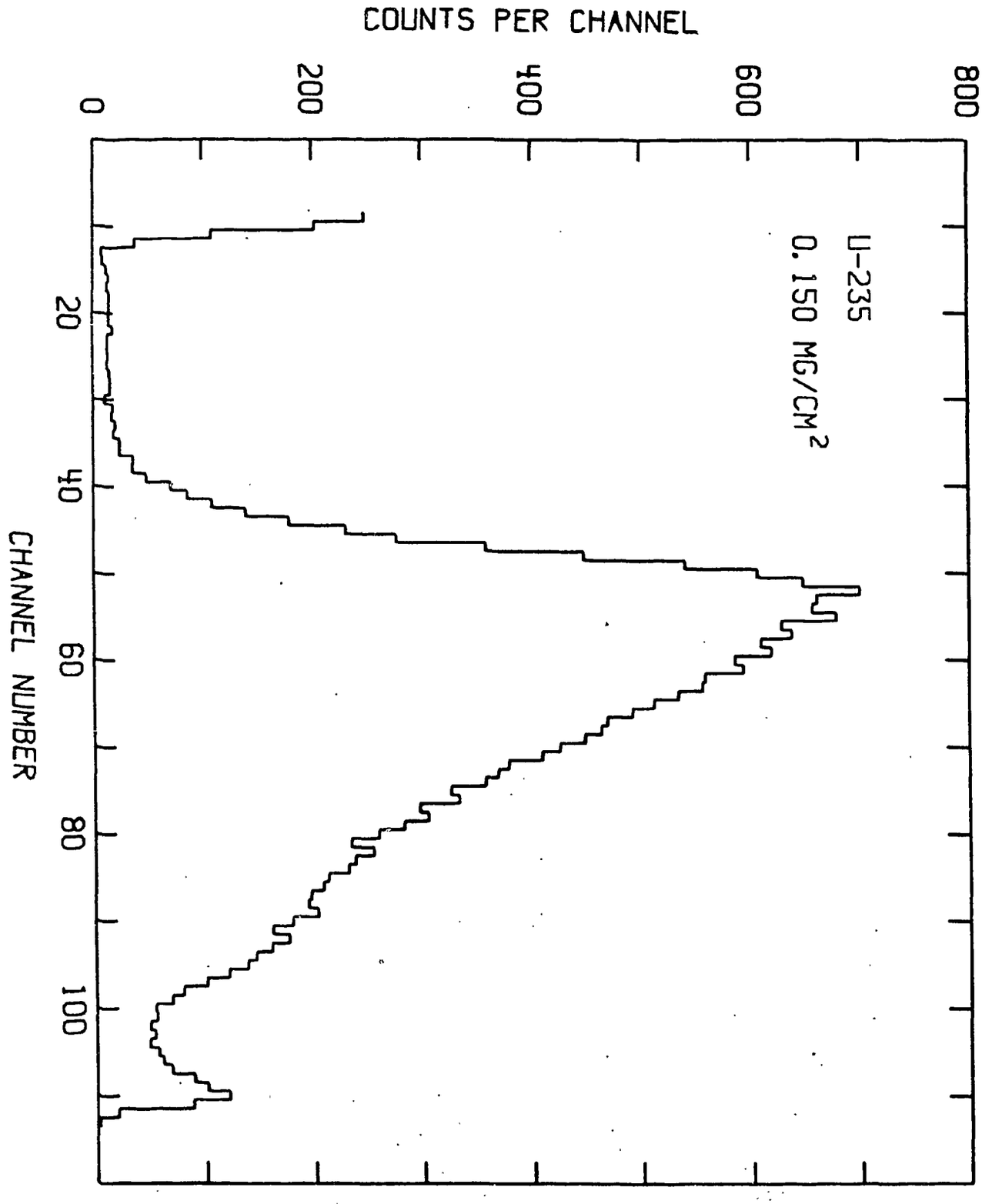
COUNTS PER CHANNEL
0
200
400
600
800
U-235
0.150 MG/CM2
CHANNEL NUMBER
20
40
60
80
100

FIGURE - 4.

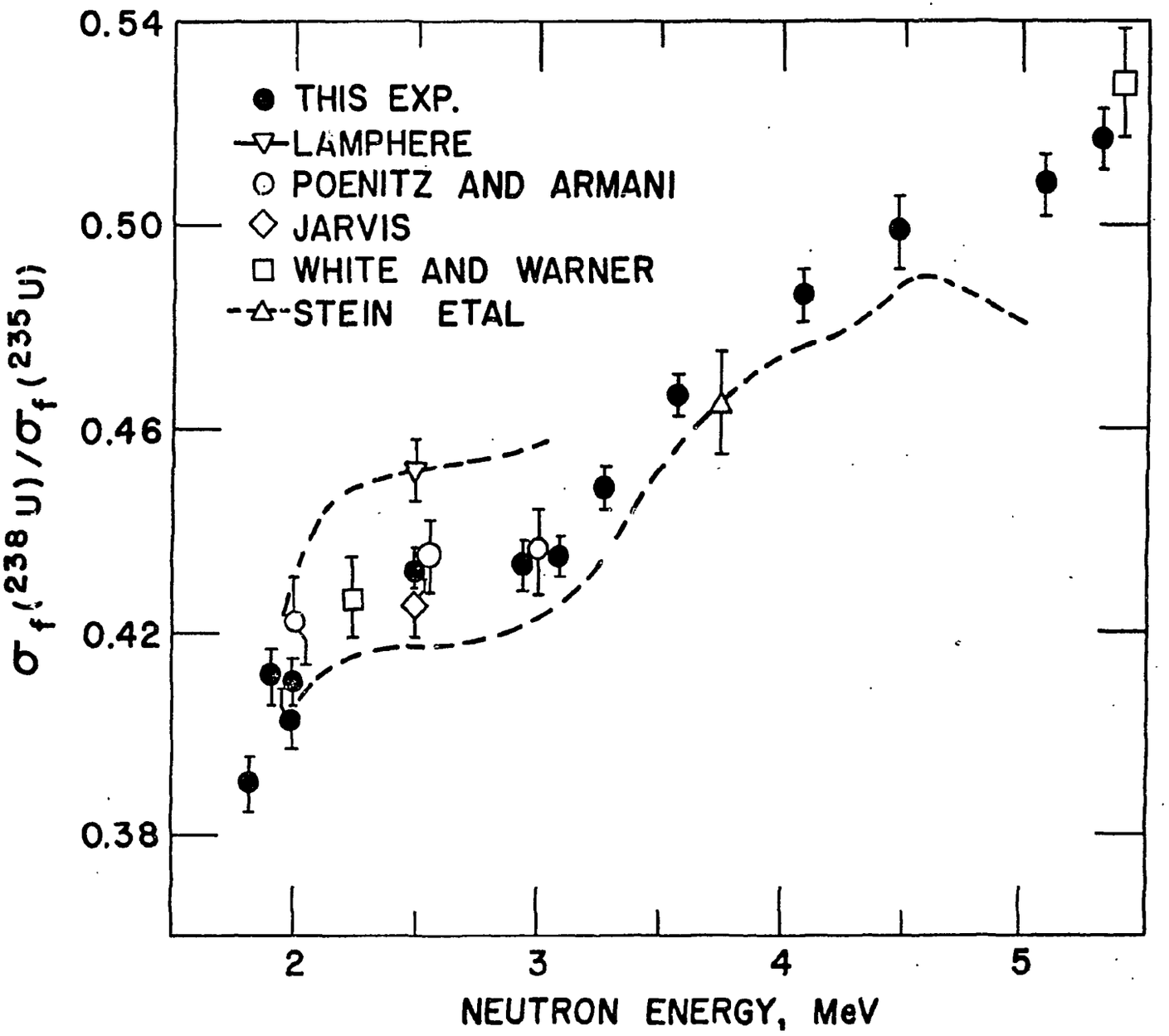
0.54
0.50
0.46
0.42
0.38
2
3
4
5
NEUTRON ENERGY, MeV
$\sigma_f(^{238}U)/\sigma_f(^{235}U)$
THIS EXP.
LAMPHERE
POENITZ AND ARMANI
JARVIS
WHITE AND WARNER
STEIN ETAL

FIGURE -5.

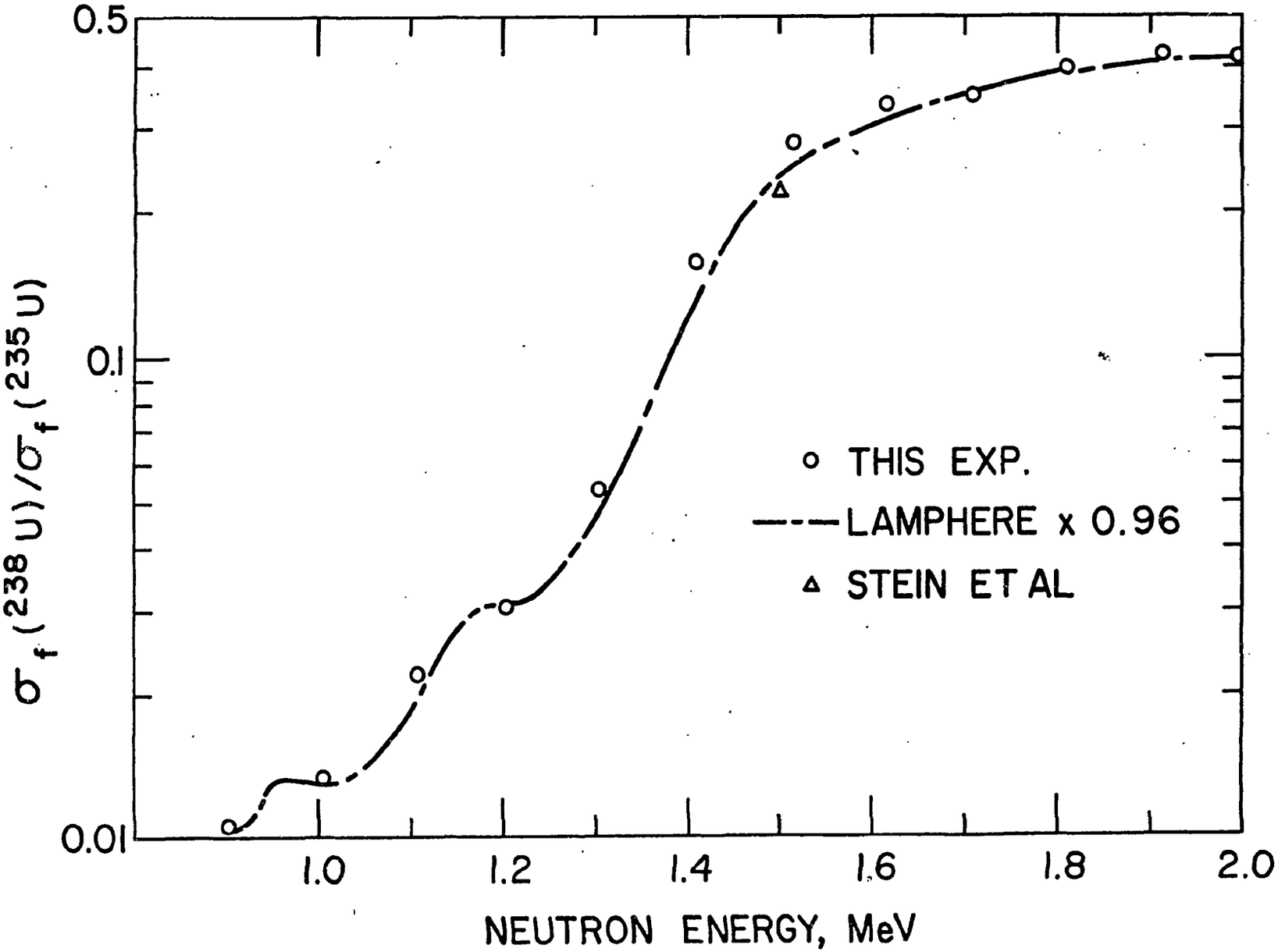
0.5
0.1
0.01
$\sigma_f(^{238}U)/\sigma_f(^{235}U)$
1.0
1.2
1.4
1.6
1.8
2.0
NEUTRON ENERGY, MeV
o THIS EXP.
---LAMPHERE x 0.96
△ STEIN ET AL

FIGURE -6.