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NUCLEAR FISSION INDUCED BY RADIATIONLESS TRANSITIONS IN THE MU-MESONIC ATOMS Th^{232}, U^{235}, AND U^{238}

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ABSTRACT

The time distribution of fissions in Th^{232}, U^{235}, and U^{238} induced by μ^- mesons was measured with a multiplate gas-scintillation fission chamber.

A significant number of prompt fissions not associated with μ^- nuclear capture was observed. The results are:

Nucleus	Ratio of prompt fissions to fissions from nuclear capture
Th^{232}	0.064 ± .022
U^{238}	0.072 ± .014
U^{235}	0.111 ± .021

The work of Mukhin et al. shows that the intensities of μ-mesic K x-rays for these elements relative to Pb are 0.85 ± .0 (Th), 0.77 ± .04 (U^{238}), and 0.71 ± .05 (U^{235}). This intensity reduction is qualitatively consistent with earlier predictions that, for these elements, a direct excitation of the nucleus competes with electromagnetic radiation in the transition to the ground state of the mesic atom. Our results indicate such direct nuclear excitation.

The number of fissions observed may be consistent with the results of Mukhin et al. and with photofission data, if allowance is made for the effect on the fission barrier of the μ meson in the 1S state of the mesic atom.

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I. INTRODUCTION

In 1958 Zaretsky predicted that transitions between low-lying μ-mesic atomic states may induce nuclear excitation instead of x-ray emission.[1] His first calculation indicated that most and perhaps all of the observed nuclear fissions induced by μ stoppings in U-loaded emulsion[2] were due to nonradiative atomic processes.

The authors of this paper, stimulated by the above predictions, demonstrated in an earlier experiment that, at most, only a small fraction of μ-meson-induced fissions in U could be due to a process other than nuclear capture.[3] A similar conclusion was reached independently by Belovitskii et al. based on measurements made in nuclear emulsions.[4] Recent observations of μ-mesonic atoms have shown that for large Z, the μ-mesonic K x-ray yield is significantly less than 100% (see table 1).[5,6] This experimental fact has led the authors of this paper to repeat their previous experiment with greater statistical accuracy and to use U^{235},† and Th^{232} as targets, in addition to natural U. The observation of any nuclear excitation demonstrably unrelated to the nuclear capture of the meson would explain the missing x rays and confirm the predictions of Zaretsky as later modified by Zaretsky and Novikov.[7]

The time required for a μ^- meson to come to rest in condensed material is $< 10^{-9}$ sec. Within 10^{-12} sec after coming to rest, the meson is captured by an atom and, by means of Auger and radiative transitions, cascades into the 1S state

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† Natural U is 99.3% U^{238}, and the U^{235} was 93.3% enriched.

of the mesonic atom.[8,9] The mean lifetime for nuclear capture is long compared with these times ($>5\times10^{-8}$ sec) and easily measurable. Nuclear excitations induced by atomic transitions will occur promptly, as opposed to nuclear capture excitations, that occur with the characteristic nuclear capture lifetime.

The energy of the 2P–1S transition is approximately 6.4 to 6.5 MeV in the elements with the low x-ray yield.[10] This energy is greater than the neutron binding energies and the measured fission thresholds of these elements (see table 2).

In principle, one may detect the radiationless-transition nuclear excitation by the associated prompt neutron emission, γ radiation, or nuclear fission. The neutrons may have kinetic energy up to the difference between the 2P–1S transition energy and the neutron binding energy (this is ≤ 1 MeV for the target isotopes). It is necessary to separate these neutrons from neutrons due to μ^- nuclear capture excitation for which the average neutron emission is $\gtrsim 1.5$ neutrons per μ capture. (In Pb, for example, there are $1.60\pm.06$ neutrons per μ capture.[14]) Time-of-flight technique has been used by Johnson et al., in an unseccessful attempt to detect the prompt neutrons.[15] The identification of these low-average-energy prompt neutrons in the presence of higher-intensity, higher-average-energy neutrons from nuclear capture is a virtually impossible task. It would be comparably difficult to distinguish between prompt nuclear γ's and x rays. Nuclear fission is unique in giving a large, unambiguous energy release that can be precisely timed. For this reason, this experiment was directed toward the fission process.

II. EXPERIMENTAL PROCEDURE

The problem of detecting the prompt fissions required a counter with good time resolution (of the order of nsec). For this purpose we constructed a multiplate noble-gas scintillation chamber (fig. 1). The scintillator was a mixture of 80% argon and 20% nitrogen; the nitrogen acted as a wave length shifter.

The chamber was viewed by two RCA 6655A photomultiplier tubes. There were two outputs from each tube. The last dynode signals were fed into 10-Mc pulse-height discriminaotrs, and the discriminator outputs were set in coincidence. The anode signals were added to give a pulse proportional to the light produced by the fission fragments.

The targets each consisted of nine 3.25-in.-diam plates spaced 0.125 in. apart. The U plates were 0.010 in. thick and the Th plates were 0.025 in. thick; however, because of the short range of the fission fragments the effective target thickness was very small. The U^{235} target consisted of nine stainless steel plates 0.010 in. thick. Each plate had 1 mg/cm^2 of $U^{235}F_4$ evaporated on each side. The surfaces of all target plates were polished and aluminized to maximize their reflectivity.

Since all three targets were α-particle emitters, we had to discriminate against a large α background. At the operating pressure of the chamber, 45 psi above atmospheric, the ratio of fission to α-particle pulse height was greater than 3:1 for a thin Cf^{252} source (a few mg/cm^2). Figure 2 shows the α and fission pulse-height distribution from Cf^{252}. The α background was eliminated by the pulse-height discrimination mentioned above. The efficiency for detecting Cf^{252} fissions in the chamber was 100%, based on comparison with a continuous-flow methane ionization chamber. A full description of the scintillation chamber is in preparation.

A μ beam of high purity was necessary, since fissions from π^- stoppings in the targets have effectively the same time distribution as the fissions produced by radiationless transition excitation; for experimental purposes this is a δ function at zero time. There are techniques for increasing the μ/π ratio in a beam, but these also reduce the μ intensity. Unfortunately, owing to the small effective target thickness, a maximum μ yield was also necessary. Therefore we separated the π^- and μ^- by range only, using a 213±5-MeV/c beam from the Berkeley 184-inch cyclotron. The beam from the cyclotron was taken out through the meson wheel, collimated at the entrance and exit of the wheel to a 4×4-in. area, then bent 45 degrees and collimated again to a 4×4-in. area before entering the experimental area. The magnet, with 28-deg wedge pole pieces, gave a momentum focus at the center of the absorber between counters S_2 and C with sufficient dispersion to have a momentum spread of 2.5%. Figure 3 shows the experimental arrangement.

The counter telescope is shown in Fig. 4. All counters were made of plastic scintillator, with the exception of the fission chamber described above, and counter C, which was a 5×5×1-in. water Cerenkov counter used to discriminate out electrons in the beam. Counters S_1, S_2, S_3, and S_4 were viewed with RCA 6655A photomultiplier tubes, and counters C and A with RCA 6810A phototubes.

The beam monitor comprised counters S_1 and S_2 in coincidence. The mu-stopping signal was obtained from a coincidence between S_3 and S_4 in anticoincidence with the sum of A and C. The coincidence pulse from the fission counter triggered a 0.5-μsec gate that was set in coincidence with the delayed μ^--stopping signal. The μ^--stopping signal was delayed in such a way that, for prompt fissions, it arrived at the middle of the gate.

The output of the coincidence between the fission gate and μ^--stopping signal was used to trigger a multiple-beam oscilloscope. The output of counters

S_2, C, S_3, S_4, and A was fed into an adder, timed so that when the output of the adder was displayed on one of the scope sweeps there was at least 30 nsec between the pulses. On another scope sweep, counter S_3 and the sum of the anode signals from the fission phototubes were displayed, and these were timed in such a way that 100 nsec elapsed between S_3 and a prompt fission. This display, together with the gate timing, allowed us to measure the accidental fission rate by looking at "negative-time" fissions and to study the effect of our time resolution on the time distribution of zero-time fissions. Both scope sweeps were time-calibrated with a 50-Mc Tektronix crystal oscillator. The scope pictures were recorded on 35-mm TRIX film. A block diagram of the electronics is shown in Fig. 5.

Figure 6 shows a differential range curve taken with a stainless steel "dummy" target. The data runs were made at 12.5-in. of CH_2, since the average thickness of the actual targets was a little greater than that of the stainless steel. The μ^- stopping rate in a 3.4-g/cm^2 target in an area defined by a 1.5-in. diameter counter was 20 000/min.

We measured the prompt fission yield as a function of absorber thickness, assuming that in the neighborhood of the π peak all prompt fissions were produced by pions. The prompt-fission yield vs absorber thickness is given in Fig. 7. We assumed as a first approximation that the range distribution is Gaussian and therefore fitted the fission yield with such a curve, and obtained a $P(\chi^2) \approx 0.5$; this showed that the data are well described by such a function.

It can be seen, by comparing Figs. 6 and 7, that the π^- stopping rate at 10 in. of CH_2 is already down to 1% of the μ^- stopping rate at 12.5 in. (because of the extremely low yield it was not practical to extend the measurement beyond 10 in.). If the calculated Gaussian properly describes the π distribution up to the μ range, the μ/π ratio would be $\approx 10^9$; if the π stoppings continued to fall off at the same rate as the end of the curve in Fig. 7, the μ/π ratio would

be $\geq 10^4$. A μ/π ratio $\geq 10^3$ at the μ range seems a certainty and, as we show below, this is sufficient for our purposes, so that the contribution of π contamination is negligible. It is also shown by an independent argument that the observed prompt fissions cannot possibly be explained by π contamination.

Differential range curves, time calibrations, discriminator-stability checks, and zero-time determinations were made frequently. The original values remained constant throughout the run.

The actual data runs were between an hour and an hour and a half long. Four to six runs were taken consecutively. The Th^{232}, U^{238}, and U^{235} targets were alternated every six to nine hours. The background was measured simultaneously at negative times.

For each event, the fission pulse height and its time relative to S_3 were measured. The criteria for an acceptable event were: (a) that no pulse from either counter C or A was present, (b) that the fission pulse height was larger than or equal to the minimum pulse height from the thin Cf^{252} source on the targets; and (c) that the relative timing of S_3 and S_4 was correct and that there were no spurious telescope pulses. Eighty percent of the scope pictures satisfied the above criteria.

The zero time was obtained from fissions produced by stopping pions. Because of reading errors and electronic rise times, the time distribution (Fig. 8) appears as a Gaussian with a half-width of 1.5 nsec, and introduces a significant modification to the exponential distribution of fission from mu capture. Figure 9 shows the effect of a Gaussian distribution with $\sigma = 1.5$ nsec on an exponential with a mean life of $\tau = 75$ nsec. To correct for this effect, the fissions were grouped into intervals (or bins) 10 nsec wide, a time that is long compared with the Gaussian perturbation. The fissions in the first 5 nsec of negative time were included in the first interval.

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III. DATA REDUCTION AND RESULTS

A least-squares fit of the data to a lifetime curve was made with an IBM 704 computer, with the first channel omitted.[16] The program output gave the zero-time fission rate I_0, and the mean life, τ of the bound muon, together with their standard errors. In the calculation, we varied the background by a standard deviation in each direction in order to study the sensitivity of our results to fluctuations in the background. These variations produced an average change of ±0.6% in the mean lives, and negligible changes in the zero-time intercepts. A χ^2 test was made on each fit as a measure of its validity. Figure 10 shows a plot of the events vs time and the best fits for the data. Table III is a summary of the least-squares results. The U^{238} lifetime is in good agreement with our earlier measurement of 75.4±5.5 nsec[3] but not with the other reported value which was 88±4 nsec.[17]

The contribution of nuclear-capture fissions in the first channel is given by $N_{0(cap)} = I_0\, \tau(1-\exp[-t_0/\tau])$, where t_0 is the width of the first channel. The errors in the determinations of time zero and the channel widths were small compared with the statistical error in I_0. The difference between the number of events in the first channel and the calculated contribution from nuclear capture fissions gives the fission events associated with radiationless transition.

We did not determine the absolute stopping rate in the targets because of problems connected with measuring the effective target thickness; therefore we were unable to obtain the fission probability associated with a stopping μ meson. We express our results as the ratio of fissions associated with radiationless transition to fissions due to μ^- capture. From our data we also obtained the ratio of μ^- to π^- fission probabilities for all three targets. Table 4 gives the results of the experiment together with their statistical standard errors.

A μ^-/π^- fission ratio of 0.15±.03 for U^{238} can be obtained by comparing the radiochemical results of Russell and Turkevich[21] with the emulsion measurements by Mikhul and Petrashku.[22] This ratio is in good agreement with our value.

IV. DISCUSSION

No correction was made for π^- contamination in our beam, which was estimated in sect. II above to be much less than 0.1%. A further check can be obtained by calculating the contamination that would be necessary to account for our results. If we assume no contribution from radiationless transitions, the π^- contamination would be given by

$$\frac{\mu^- \text{ fission probability}}{\pi^- \text{ fission probability}} \times \frac{\text{prompt fissions}}{\mu^- \text{ capture fissions}}$$

$$= \frac{\pi^-}{\mu^-} \text{ stoppings in the target.}$$

The targets were run at the same absorber thickness, no one would expect the same π^- contamination in all three targets, but we find that the ratio is <u>different</u> for each target (see table 5), indicating that an anomalously large π contamination is not sufficient to explain our results.

The probability that prompt fissions will be produced by radiationless transitions is equal to the product of the radiationless-transition probability and the probability that the excited nucleus will fission. A rough estimate may be made by assuming the radiationless-transition probability to be equal to the fraction of missing x rays, and the fission probability to be equal to the fission branching ratio for 6.5-MeV γ rays, which is $\sigma_f/\sigma_f + \sigma_n$, where σ_f is the photofission cross section and σ_n is the photoneutron cross section. The 2P–1S radiationless-transition excitation results from electric dipole interaction and there is experimental evidence that the photoexcitation cross section at this

energy is also electric dipole.[23,24]

The results of the calculation are given in table 6 (column c). We also include in column d the total μ^- fission probability as measured with emulsions.

A comparision of columns (c) and (d) of table 6 with our experimental results (table 4) shows a striking inconsistency; namely, the estimated prompt-fission probability is much larger than experimentally observed.

We believe this discrepancy can be explained by the following consideration: The μ^- meson in a 1S state spends approximately 50% of its time inside the nucleus in elements near U. This introduction of negative charge into the nucleus reduces the Coulomb energy of the nucleus, thereby increasing the fission barrier. One might estimate the order of magnitude of the change in fission probability due to changes in the height of the fission barrier by using a relationship derived by Frankel and Metropolis[28] for the spontaneous fission lifetime:

$$T = 10^{-21} \times 10^{7.85\,E_{th}} \text{ sec}, \qquad (1)$$

where E_{th} is the height of the fission barrier in MeV. This expression gives a change by a factor of 10 in the fission time for every 0.13 MeV of change in the fission barrier.

If the perturbation due to the presence of a μ^- meson in the 1S state of U^{238} were sufficient to raise the fission barrier approximately 0.1 MeV above the 2P-1S transition energy, then the fission probability would be decreased by approximately a factor of 10 and make our results consistent with the estimated prompt fission probability. Although neither present numerical data nor theory are sufficiently precise for accurate calculations, estimates by Zaretsky and Novikov indicate that an increase in the fission barrier of roughly the necessary amount does occur.

In summary, we conclude that the prompt nuclear fissions we have observed result from the nonradiative atomic process proposed by Zaretsky, and that they

may be consistent with x-ray results if allowance is made for the modification of the fission barrier by the muon.

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FOOTNOTE AND REFERENCES

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Table 1. Measured 2P–1S x-ray yields relative to Pb in high-Z μ-mesic atoms

Nucleus	2P–1S transition x-ray yield	
	Mukin et al.[a]	Hincks et al.[b]
W		1.05 ± .1
Pb	1	1
Bi	1± .06	0.9± .1
Th^{232}	0.85± .0 (?)	
U^{235}	0.71± .05	
U^{238}	0.77± .04	0.7± .1

[a] See ref. 5.

[b] See ref. 6.

Table 2. Neutron binding energies and fission thresholds in Th^{232}, U^{235}, and U^{238}

Nucleus	Neutron binding energy[a]	Measured fission threshold[b]	Estimated fission barrier[c]
Th^{232}	6.20± .04	5.40± .22	5.95
U^{235}	5.37± .15	5.31± .25	5.75
U^{238}	6.03± .13	5.08± .15	5.80

[a] See ref. 11.

[b] See ref. 12.

[c] See ref. 13.

Table 3. Least-squares results for the Th^{232}, U^{235}, and U^{238} data

Nucleus	No. of events	τ(nsec)	I_0, Fissions per 10^{-8} sec at t = 0	Background per 10^{-8} sec	$P(\chi^2)$
Th^{232}	592	74.2±5.6	74±6	1.87±.56	0.65
U^{235}	1130	66.5±4.2 [a]	147±11	2.64±.64	0.15
U^{238}	1328	75.6±2.9 [a]	179±7	.32±.32	0.90

[a] The difference in lifetime between U^{235} and U^{238}, while not significantly large, is very likely real and a consequence of the "isotope effect" calculated by Primakoff (ref. 18). Lifetime differences have also been observed between isotopes of Cl and of Ca (refs. 19, 20). Our lifetime difference gives Primakoff's δ a value of $3.08^{+0.07}_{-0.22}$.

Table 4. Results of the experiment.

Nucleus	Radiationless transition fissions / Nuclear capture fissions	μ^- Fission probability / π^- fission probability
Th^{232}	0.064±.022	0.085±.004
U^{235}	0.111±.021	0.281±.015
U^{238}	0.072±.014	0.174±.007

Table 5. π^- contamination.

Nucleus	π^- contamination necessary to produce the experimental results	Estimated upper limit to π^- contamination
Th^{232}	0.0068	0.001
U^{235}	0.0318	0.001
U^{238}	0.0125	0.001

Table 6. Estimate of radiationless-transition-induced fissions.

Nucleus	(a) Photofission branching ratio at 6.5 MeV	(b) Fraction of x-rays missing [a]	(c) = (a)(b) Fraction of μ^- expected to produce prompt fission	(d) Total μ^- fission probability
Th^{232}	0.18 [b]	0.15±.0(?)	0.27	0.018±0.012 [d]
U^{235}	0.4 [c]	0.29±.05	0.1	<0.28 [e]
U^{238}	0.24 [b]	0.23±.04	.055	0.070±.008 [f]

[a] See ref. 4.

[b] See ref. 24.

[c] Estimated from Table 2 and Fig. 2 of ref. 26.

[d] See ref. 27

[e] The μ^-/π^- fission ratio of 0.28 (Table 4) sets this upper limit.

[f] See ref. 22.

FIGURE CAPTIONS

Fig. 1. Photograph of the fission chamber showing a nine-plate target and a thin window for the μ^- beam.

Fig. 2. Cf^{252} alpha and spontaneous-fission pulse-height distribution.

Fig. 3. Plan view of the experimental arrangement of the 184-inch cyclotron "meson cave."

Fig. 4. The counter telescope.

Fig. 5. Block diagram of the electronics.

Fig. 6. Differential range curve taken with a target of nine stainless steel plates, each 0.010 in. thick.

Fig. 7. Prompt fission yield from the U^{238} target vs absorber thickness.

Fig. 8. An example of the time distribution of π^- fissions of U^{238}.

Fig. 9. The effect of the time distribution on the exponential time distribution The modified curve is described by the function

$$F(E) = \frac{\tau}{N_0} \frac{dN}{dt} = \exp[-t/\tau]\left(\frac{1}{2} + \frac{1}{\sqrt{2\pi\sigma^2}} \int_{-t}^{0} \exp[-y^2/2\sigma^2]\,dy\right).$$

Fig. 10. Time distributions of the fissions of Th^{232}, U^{235}, and U^{238} produced by stopped μ^- mesons (background included). The solid lines are least-square fits to the data, excluding the first time interval.

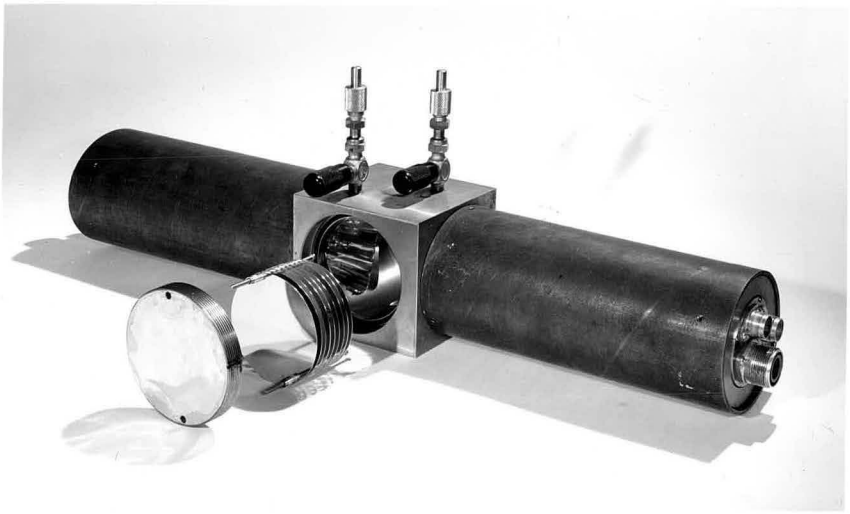


Fig. 1

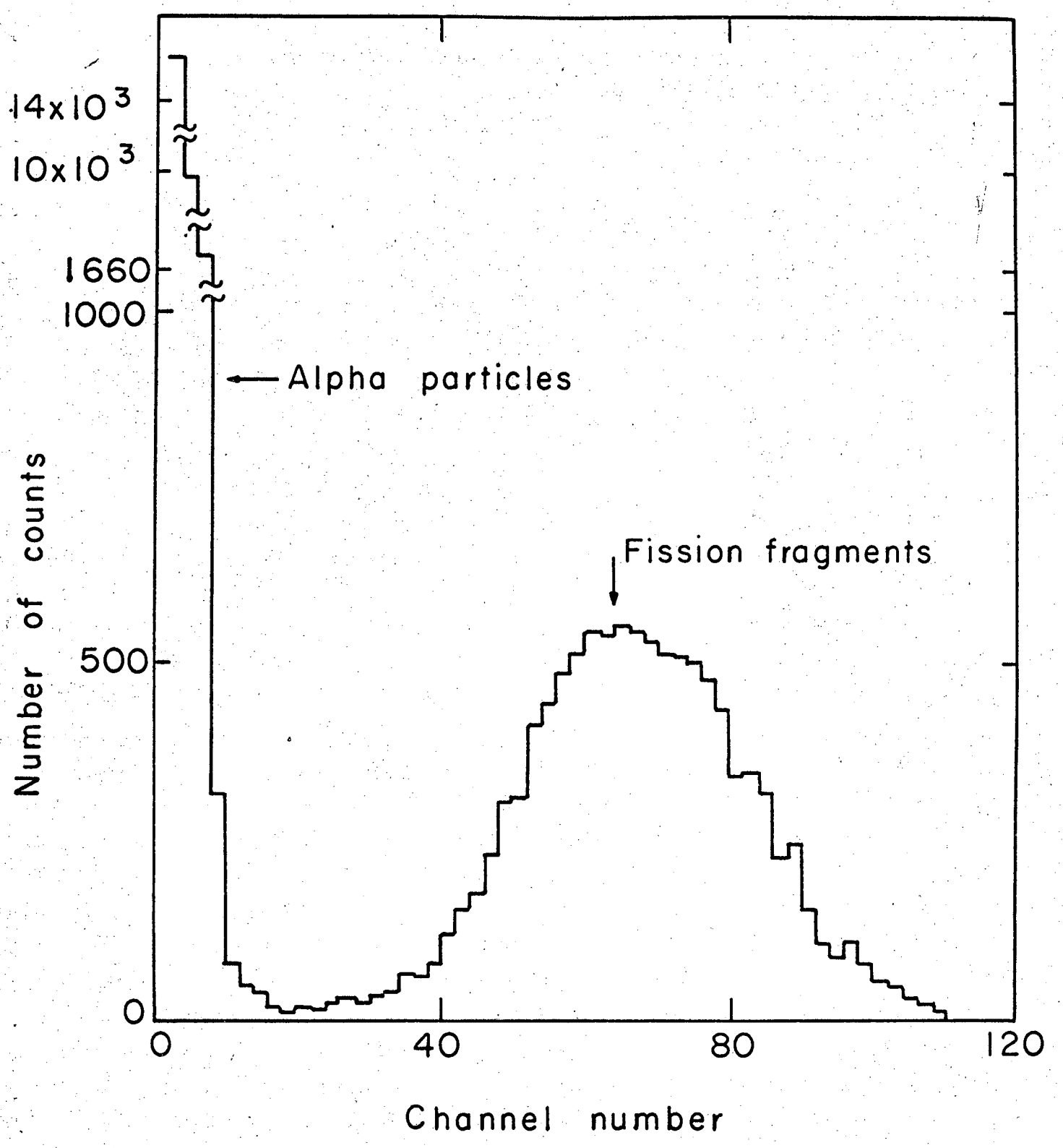


Fig. 2

MU-27146

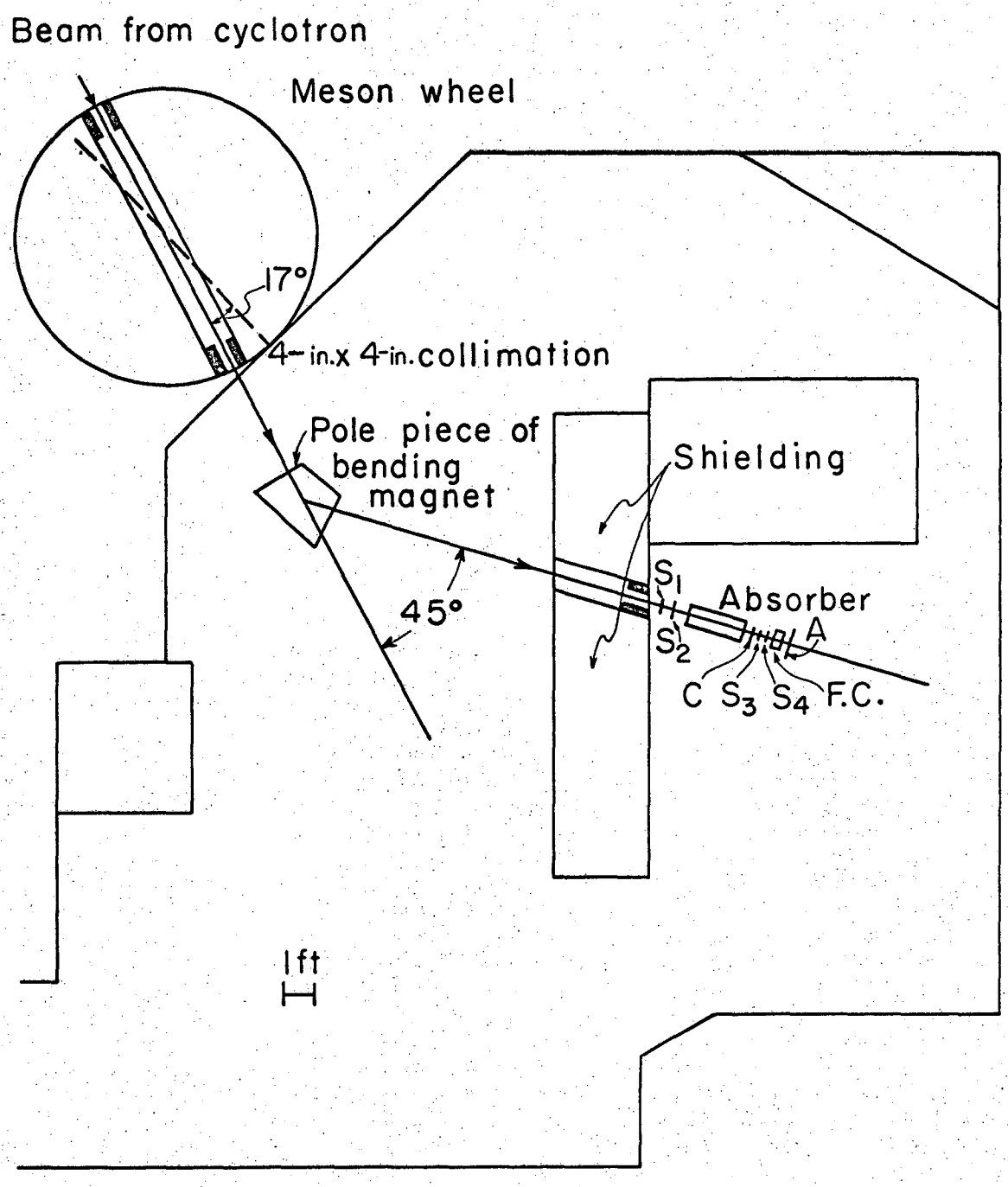
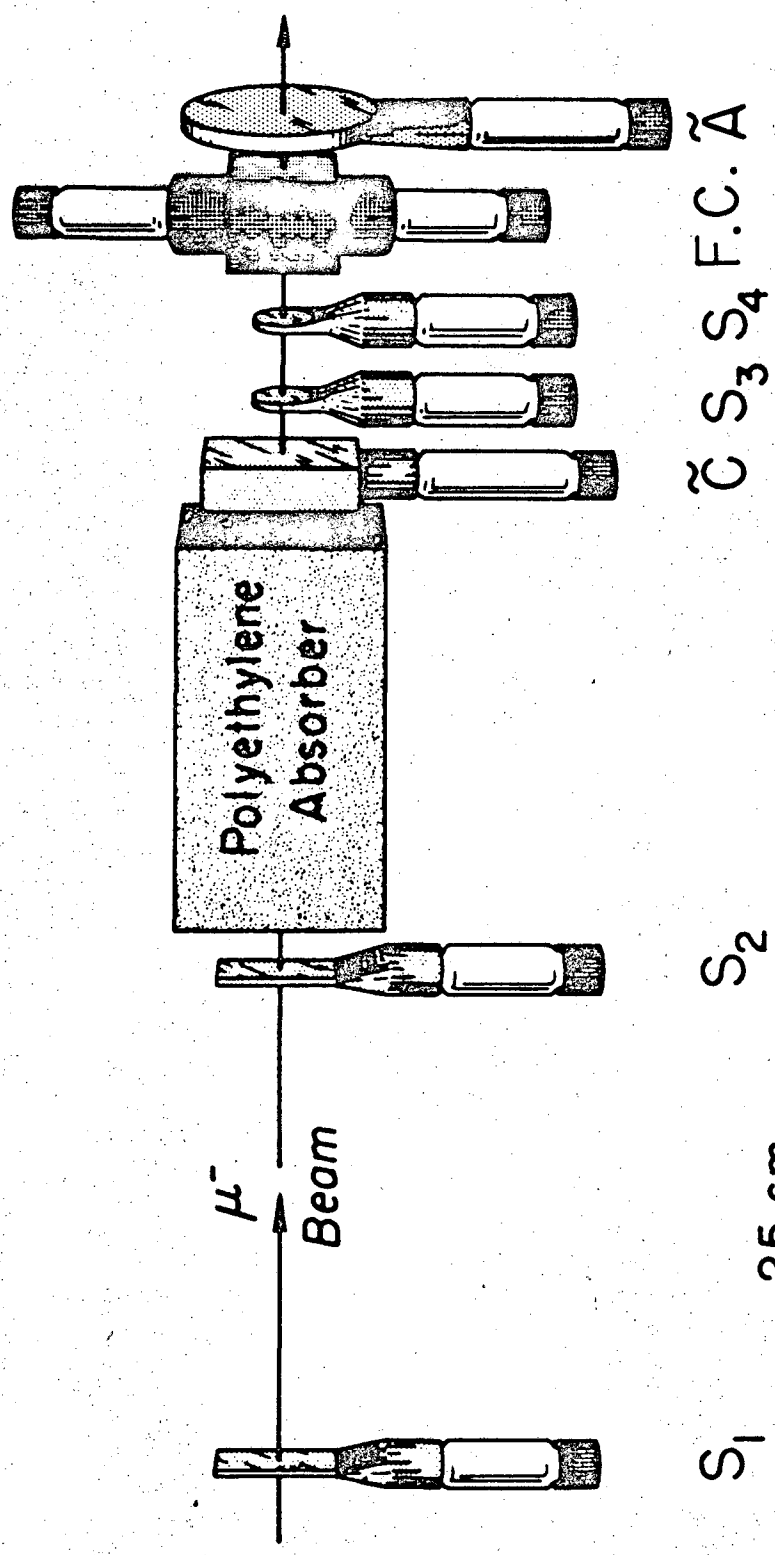


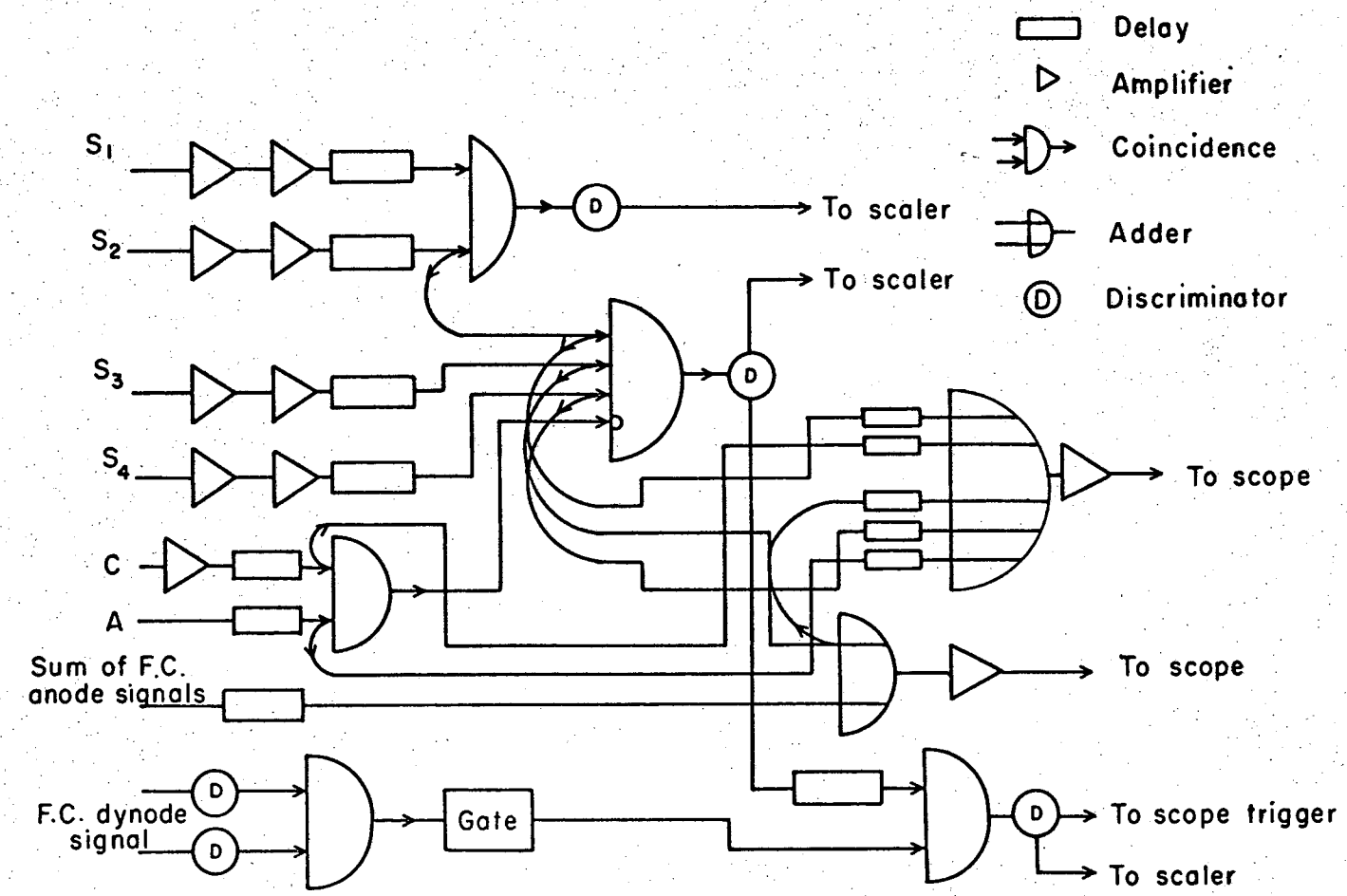
Fig. 3

MU-27151



MU-17654-A

Fig. 4



MU-27206

Fig. 5

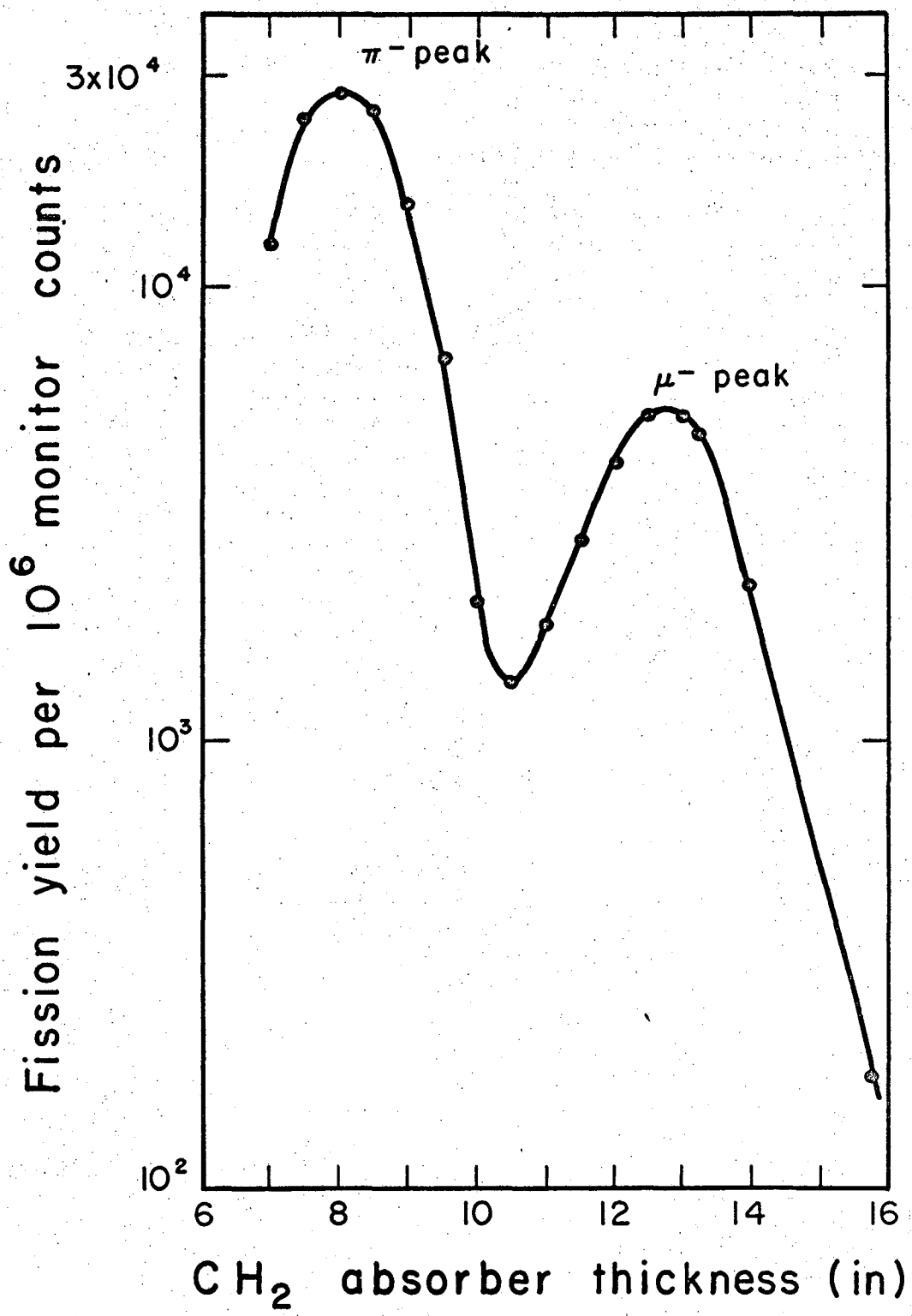


Fig. 6

MU-27205

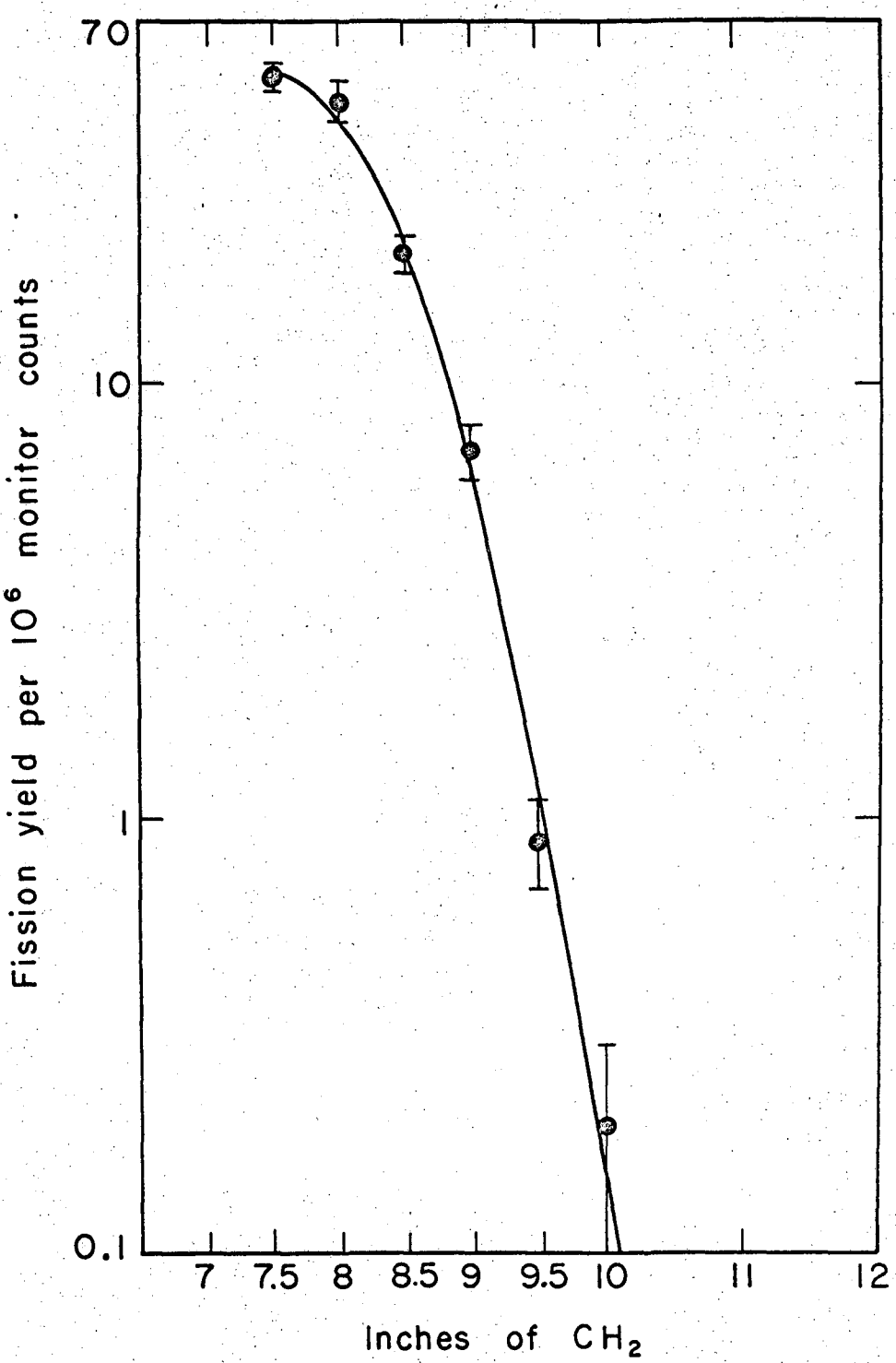


Fig. 7 MU-27208

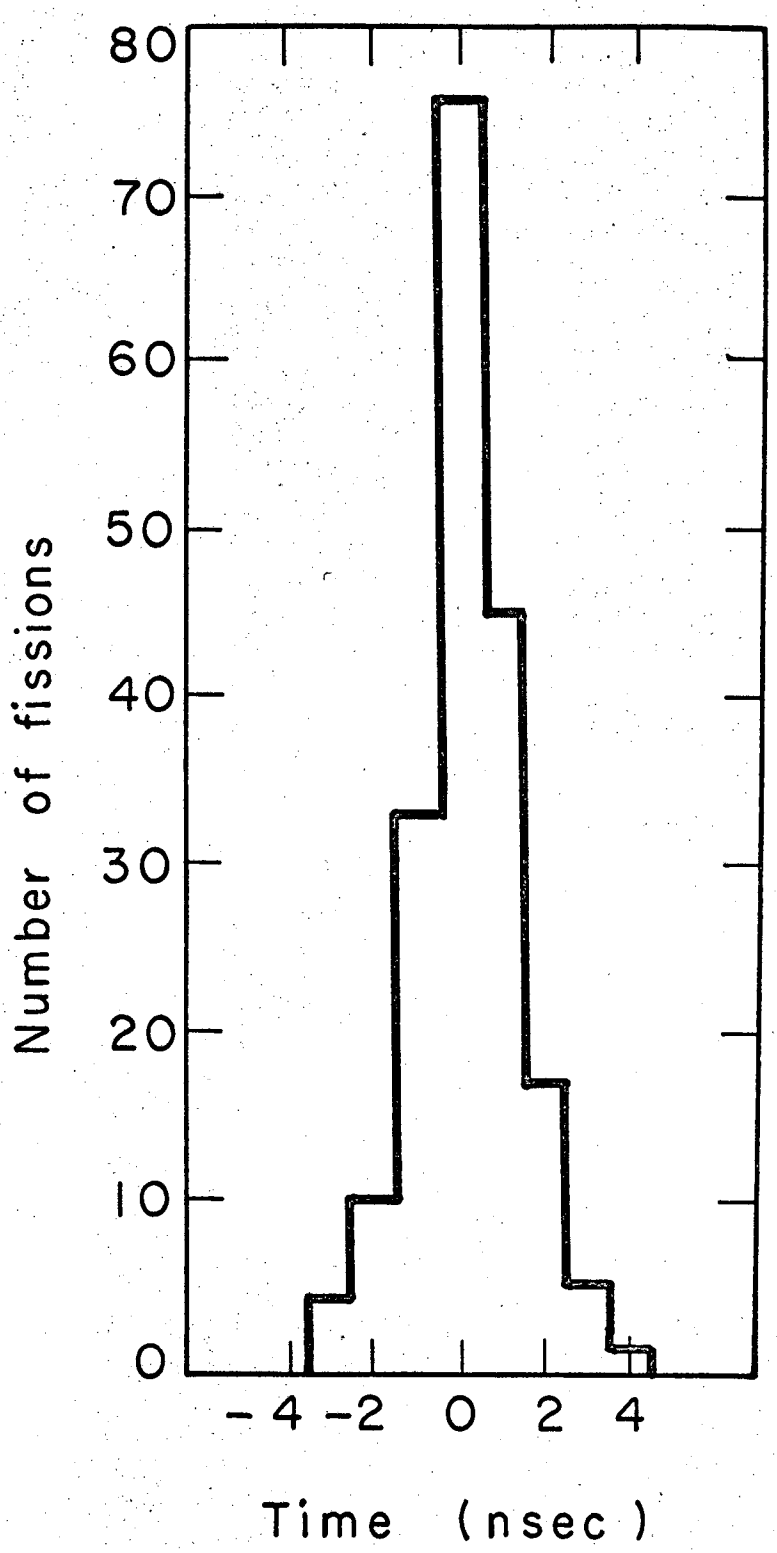
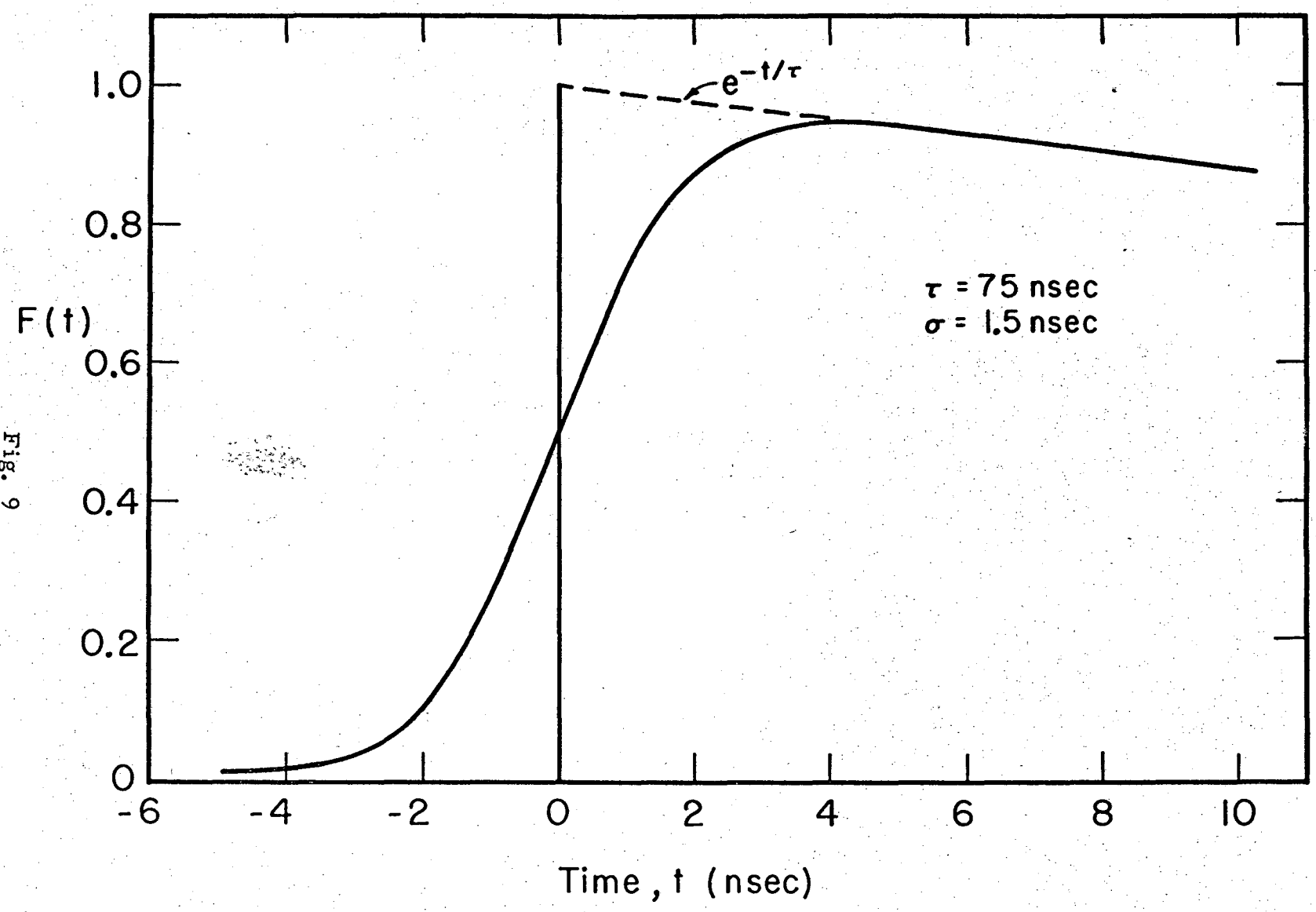


Fig. 8 MU-27207



MU-25251

Fig. 9

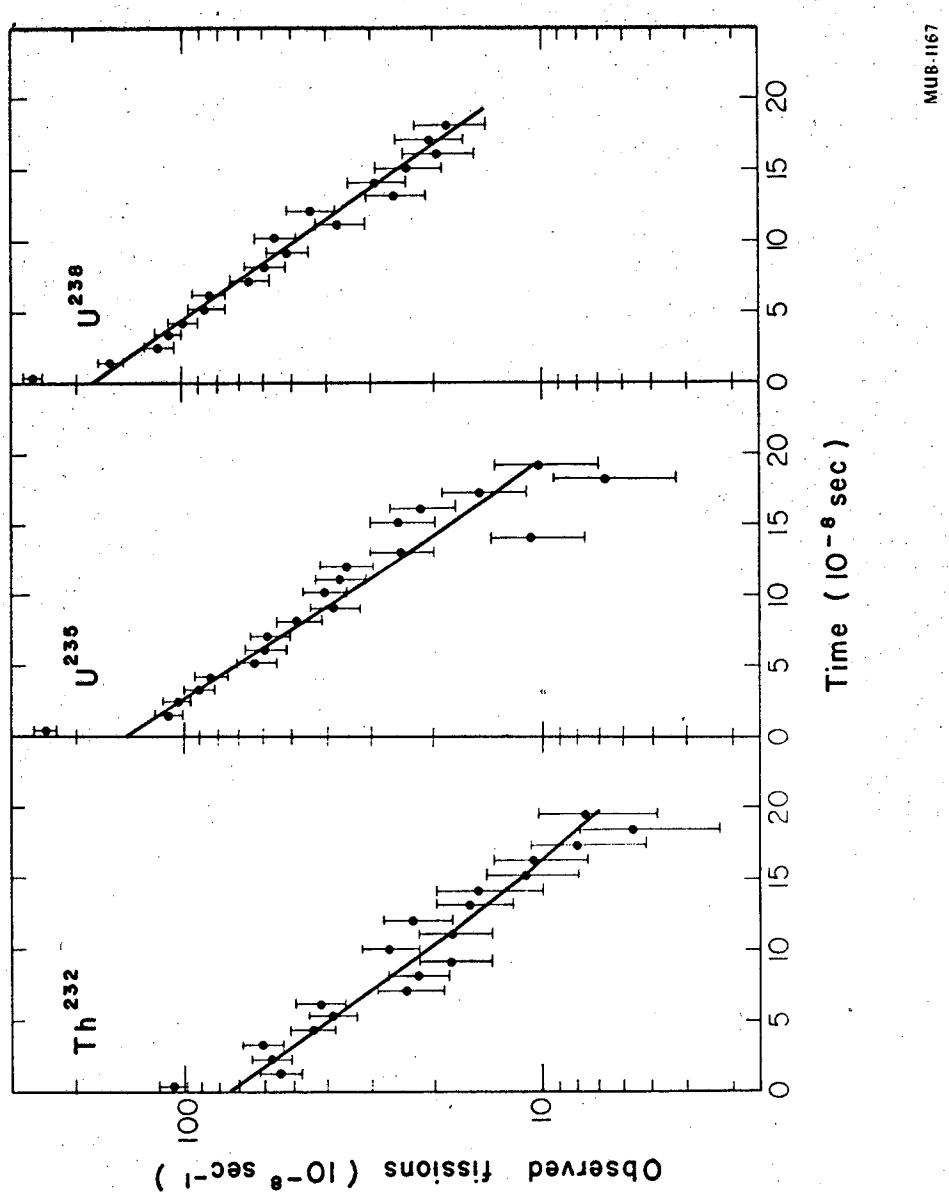

Th 232
U 235
U 238
100
10
0
5
10
15
20
Observed fissions (10⁻⁸ sec⁻¹)
Time (10⁻⁸ sec)
MUB-1167

Fig. 10

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