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SPONTANEOUS FISSION *

Emilio Segrè

Department of Physics, Radiation Laboratory
University of California, Berkeley, California

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1. Historical

The first attempt to discover spontaneous fission in uranium was made by Libby,[1] who, however, failed to detect it on account of the smallness of effect. In 1940, Petrzhak and Flerov,[2] using more sensitive methods, discovered spontaneous fission in uranium and gave some rough estimates of the spontaneous fission decay constant of this substance.

Subsequently, extensive experimental work on the subject has been performed by several investigators and will be quoted in the various sections.

Bohr and Wheeler[3] have given a theory of the effect based on the usual ideas of penetration of potential barriers.

On this project spontaneous fission has been studied for the past several years in an effort to obtain a complete picture of the phenomenon. For this purpose the spontaneous fission decay constants λ have been measured for separated isotopes of the heavy elements wherever possible. Moreover, the number ν of neutrons emitted per fission has been measured wherever feasible, and other characteristics of the spontaneous fission process have been studied. This report summarizes the spontaneous fission work done at Los Alamos up to January 1, 1946. A chronological record of the work is contained in the Los Alamos monthly reports.[4]

2. Experimental Techniques

The experiments directed to the measurement of λ consisted in principle of putting a certain amount of the material to be investigated in ionization chambers

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* This is a partial reproduction of a report written in Los Alamos in 1945.

connected to linear amplifiers, and counting the fission pulses. The material was deposited on platinum discs as a thin layer.(5)

In all these experiments one of the main difficulties is offered by the alpha activity of the samples. As a matter of fact, this often limits the amount of a substance that can be studied at one time in an ionization chamber. The reason for this is that the fissions are recognized from the large size pulses that they give in the ionization chambers. Now the pulses generated by single alphas are from 10 to 20 times smaller; however, if the alpha emission is very strong, fluctuations in the alpha activity background may simulate large pulses and cause spurious fission counts to be recorded.

Qualitatively, these fluctuations will be roughly proportional to the square root of the number of alphas emitted during the "resolving time" of the apparatus. Attempts have been made to obtain a more quantitative picture of this effect by developing a suitable theory, but the phenomena that give rise to spurious pulses are too complex to be analyzed in a really satisfactory way and we shall limit ourselves to the statement above and to some experimental results to be given later.

It is clear, however, that it is desirable to have a high resolving power in the apparatus. The limitations to this may come from the collection time of electrons in the chamber and from the frequency response or rise time of the amplifier. For the chamber, electron collection with its high velocity is imperative. The chambers were filled with tank argon,(6) and special precautions had to be taken to avoid the presence of traces of organic vapors with their poisoning effects on the electron collection.

Two models of chambers were used. They are drawn in Figures 1 and 2. The large chambers (Figure 2) were used for material of low specific activity (U-235, U-238, Th) for which it is possible to use many milligrams of a substance without troubles due to the alpha activity. This requires large surfaces for the samples in order to preserve their thinness. The small chambers were used

for the more active substances (Figure 1).

The amplifiers used must have high resolving power and good stability in [redacted]. They must be absolutely free from disturbances such as high tension sparks, surges in the power supplies, etc; for this reason we have used battery operated units shielded in large metal boxes. The wiring diagram of one of these amplifiers is given in Figure 3. Its layout can be seen in Figure 4.

The pulses of the amplifier were registered on an impulse meter and could also be fed through a pulse lengthener to an Esterline-Angus recorder (Figure 5). The recording affords a useful check on the behaviour of the apparatus and was made periodically on all units.

Figure 6 shows a picture of one of the complete units, including the amplifier, ionization chamber, B batteries, high tansion supply, and Esterline-Angus. The chamber on the right is covered by a sheet metal can containing B_2O_3 for cosmic ray neutron shielding purposes.

The Esterline-Angus recording was also used to check that the fission pulses obey the Poisson distribution law. How well this occurs is shown in Figure 7 where we have reported the distribution of 141 uranium fissions.

Special precautions have to be taken also to shield substances that undergo neutron fission from neutrons due to cosmic rays. How important this effect may be, is shown by the following numbers concerning U-235. At sea level (Berkeley) this substance, observed in a wooden building, showed $(10.3\pm3.6)\times10^{-3}$ fissions per gram per second. Near Los Alamos (1900 meters above sea level), in a light wood building, we found $(15.5\pm2.2)\times10^{-3}$ fissions per gram per second, but shielding with 1.3 grams per square centimeter of cadmium reduced the counting rate to $(1.1\pm0.4)\times10^{-3}$ fissions per gram per second.

Starting from this last datum, it is possible to plan adequate boron shielding.

In the Los Alamos experiments, a shield of B_2O_3 7.4 grams per square centimeter thick was used. This means 2.7 grams per square centimeter of boron, which should cut down all the effects due to slow neutrons originating in cosmic rays, by

more than a factor of 10. It may be added that such a shield cuts off substantially all neutrons below 200 electron volts of energy.

In order to determine the spontaneous fission decay constant λ for the various substances, it is essential to know the amount effectively counted in each sample. This is done for most substances by subjecting the chamber containing the sample to be calibrated to a constant neutron flux produced by a Ra+Be source and counting the fissions obtained. Without changing the source or the geometry, we then replace the sample to be calibrated with a standard sample containing a known amount of substance and deposited in such a way as to be sure that it is thin for fission fragments. The amount of substance in the standard multiplied by the ratio of the fission rates of the unknown to the standard then gives the effective amount contained in the sample. After this calibration a curve giving the observed fission rate versus the gain of the amplifier may be taken in order to estimate the size of the errors that may be introduced by small gain changes. Figure 8 shows one of these plateau curves.

During operation the gain of each unit was checked every one or two days with a pulse generator (wiring diagram Figure 9). Also, for long periods polonium samples having alpha activities larger than the samples investigated, were substituted for the latter in the ionization chambers in order to check that no spurious counts would be registered. The samples were also periodically rotated among the units available.

3. Spontaneous Fission of the Various Nuclei

The single substances investigated will now be discussed.

3-1. Radium 226

This substance was investigated by D. West.[7] He found an upper limit of 0.6 fissions per gram per second for its spontaneous fission decay constant.

3-2. Ionium (Th-230)

A sample of ionium extracted by Dr. Fontana from uranium ores, was kindly put at our disposal by Dr. Hamilton. In this sample the Th/Io ratio is 3.4. The

plates were prepared as thin layers by evaporation on platinum discs and the amount of ionium was calculated from the alpha activity assuming a half life of ionium of 8.3×10^4 years. The plates had a diameter of 4 centimeters and contained approximately 1 milligram of ionium each. They were observed for about 1300 hours total, corresponding to 1.45 gram hours of observation. Two fissions occurred in that time; however, they may well be due to the thorium in the sample. Indeed, one would expect (see below) in 5 gram hours of observation on thorium about 0.8 fissions.

We conclude that 3.8×10^{-4} fissions per gram per second is the upper limit for the spontaneous fission of ionium.

3-3. Thorium (Th-232)

The large chambers were used for the investigation of this substance. The material used was thorium nitrate (C. P. Baker) which was ignited to ThO_2 and deposited as a thick layer using a small amount of collodion binder. The effective amount was determined by comparing the fast neutron fission of the sample with that of a thin layer of ThO_2 in the same chamber and source geometry. The thorium did not undergo any special treatment and hence was not radioactively pure. It is clear, however, that the concentration of thorium family products present in the sample is exceedingly small. In each pair of discs used in the large chambers there were approximately 0.25 grams of thorium effective. In the Los Alamos experiments 178 fissions in 1202 gram hours of observation were counted. This gives 4.1×10^{-5} fissions per gram per second.

There is a small cosmic ray effect even for fast neutron fissions in thorium, but this is practically negligible. As a matter of fact, we can estimate that the cosmic ray effect will be less than 10^{-6} fissions per gram per second.

Since the thorium spontaneous fission is so small, the question of its possible causation by impurities has to be considered. Of these only ordinary uranium can be of importance and a special experiment was made on this point by subjecting the thorium samples to a slow neutron bombardment. It showed an apparent

slow neutron cross section of less than 10^{-27} cm^2. If this were all due to natural uranium impurity the corresponding spontaneous fission would still be less than 0.1 of the effect observed in thorium.

Maurer and Pose[9] have reported some measurements on neutron emission by thorium, attributed to spontaneous fission. We shall discuss them in a subsequent section.

3-4. Protactinium 231

The material used for the samples was obtained from Dr. Agruss.

The samples were electrolytically deposited on platinum, and the amount was determined by alpha counting assuming a half life of 3.2×10^4 years.

Two samples were used: one of 140×10^{-6} grams, which was observed for 1119 hours and gave one fission; and one of 490×10^{-6} grams, which gave 10 fissions in 1129 hours.

From these data we conclude that protactinium gives 5×10^{-3} fissions per gram per second.

Again, this is probably an upper limit. From data on thermal irradiations of this same material, we know that it contains less than 2 percent uranium. The effect of this impurity on the spontaneous fission rate is negligible.

3-5. Uranium-232

This substance is formed by a (d-2n) reaction on Th-232 followed by a beta decay. The material used was prepared by the Berkeley group. It was evaporated on a platinum disc, and it had an alpha-activity of 6×10^6 disintegrations per minute due to U-232. Assuming a half life of 30 years for U-232, the sample contains 5.3×10^{-8} grams of U-232. The sample was observed for 950 hours, during which time three fissions occurred. The sample was also irradiated with slow neutrons and fission counted in order to find its content of ordinary uranium. This was found to be such that during the time of observation only 0.2 fissions were to be expected. In spite of this, in view of the long period of observation

during which a spurious fission may conceivably occur, we consider the apparent decay constant of 16 fissions per gram per second as an upper limit.

3-6. Uranium-233

This substance is formed by a (d-p) or an (n, γ) reaction on Th-232 followed by 2 beta emissions. Its half life is 1.63×10^5 years.(9) Several samples were examined in the small chambers. The material was obtained by (n, γ) on Th in the Clinton pile and prepared in Chicago by Seaborg's group. It was electroplated on platinum foils. It was observed so as to accumulate 1.35 gram hours during which one fission occurred. This fission may be explained by the U-238 content of the sample. This result gives a decay constant smaller than 2×10^{-4} fissions per gram per second.(10)

3-7. Uranium-234

A sample of U-234 was obtained on load from Dr. Latimer in Berkeley. The material was prepared by extraction of UX_1 from uranium and subsequent decay of this substance.

It contains a little over 10^{-5} grams effective of U-234 as measured by its alpha-activity. We observed it for 3300 hours without observing any spontaneous fissions, from which we conclude that the spontaneous decay constant is smaller than 9×10^{-3} fissions per gram per second.

3-8. Uranium-235 and 238

These isotopes, which occur in natural uranium, could not be separated quantitatively, from each other and the observations were always performed on mixtures containing all three of the natural uranium isotopes. However, the compositions of the mixtures could be changed by using materials enriched by the electromagnetic method.(11)

In these experiments large chambers of the second type were used.

If we call λx, λy, λz, the spontaneous fission decay constants of U-238, U-235, U-234 in fissions per gram per second, we find the counting rate c_i in a given sample is

$$c_i = x_i \lambda_x + y_i \lambda_y + z_i \lambda_z \qquad (1)$$

where x_i y_i z_i are the grams of U-238, U-235, U-234 in that sample. Practically, the term $z_i \lambda_z$ turns out always to be negligible compared with the other two because, as stated above, λ_z is small and also z_i is generally small. By observing the counting rate in samples of different known isotopic composition, we can solve the equations for λ_x and λ_y.

We shall now describe in detail an example of these measurements.

In this run three samples were used; one of ordinary uranium and two of enriched material. The isotopic composition of these materials is U-238: U-235: U-234 = 141 : 1 : 0.00725 in mass and U-238: U-235: U-234 = 0.334: 1: 0.00588 in mass, respectively.

The isotopic analysis was checked for the enriched material by mass spectrograph and by the Berkeley method of analysis.(12)

The samples were electroplated on platinum discs 13 centimeters in diameter and 0.01 centimeters thick and ignited to U_3O_8. The total mass of uranium was determined by direct weighing; and the mass of 25, by measuring the fissions occurring in a slow neutron flux. From these measurements we find that the ordinary uranium sample contains (in two plates) 38.50 milligrams of U and the two enriched samples used contain 42.95 milligrams and 36.60 milligrams of enriched material, respectively.

The normal sample gave 310 fissions in 381 hours. The enriched samples gave 101 fissions in 395 hours and 133 fissions in 558 hours. These raw data have to be corrected for the efficiency of the chamber. This is done by taking a curve of the fission rate with a constant neutron source versus bias of the amplifier and extrapolating to zero bias and then correcting further this result to take into account, theoretically, that some fission fragments cannot escape from the layer because of the finite thickness of the same. The first correction is 6 percent, the second is 3 percent. With these corrections we find, e.g., that

normal material gives $\frac{310 \text{ fissions}}{13.39 \text{ g x hr}} = 23.2$ or 6.43×10^{-3} fissions per gram per second.

Similarly, enriched material gives

$$\frac{234 \text{ fissions}}{34.08 \text{ g x hr}} = 6.87 \text{ or } 1.91 \times 10^{-3} \text{ f/g sec}$$

Introducing these numbers into equation (1) and solving we obtain $\lambda_x = 6.48 \times 10^{-3}$ f/g sec and $\lambda_y = 0.40 \times 10^{-3}$ f/g sec $\lambda_z \Sigma$ can be neglected.

The errors in these values come from statistical error in counting for which we use the square root of the number of counts; error in the absolute mass of the foil (2 percent) and in the counting efficiency (2 percent); and error in the isotopic composition (2 percent in the ratio of 25 to 28).

Calling R the ratio of U-238 to U-235 in the enriched sample and using standard formulae of the theory of errors, one finds, neglecting some small terms:

$$(\Delta \lambda_x)^2 = \left(\frac{1}{m_1}\right)^2 (\Delta c_1)^2 + \left(\frac{c_1}{m_1^2}\right)^2 (\Delta m_1)^2 \qquad (2)$$

$$(\Delta \lambda_y)^2 = \left(\frac{c_1 R}{m_1^2}\right)^2 (\Delta m_1)^2 + \left(\frac{c_2 (R+1)}{m_2^2}\right)^2 (\Delta m_2)^2 + \left(\frac{R}{m_1}\right)^2 (\Delta c_1)^2 \qquad (3)$$

$$+ \left(\frac{R+1}{m_2}\right)^2 (\Delta c_2)^2 + \left(\frac{c_1}{m_1} - \frac{c_2}{m_2}\right)^2 (\Delta R)^2$$

In which m_1 is the effective mass of the sample of normal material, m_2 is the effective mass of the sample of enriched material, and c_1 and c_2 their counting rates in counts per hour.

In the run we are considering we had:

$m_1 = 0.0350$ g	$\Delta m_1 = 0.0010$
$m_2 = 0.0350$ g	$\Delta m_2 = 0.0010$
$c_1 = 0.814$ c/hr	$\Delta c_1 = 0.046$
$c_2 = 0.240$ c/hr	$\Delta c_2 = 0.016$
$R = 0.334$	$\Delta R = 0.007$

With these data we obtain, substituting in (2) and (3),

$$\frac{1}{m_1} \Delta c_1 = 1.31 \qquad (1.31)^2 = 1.72$$

$$\frac{c_1}{m_1^2} \Delta m_1 = 0.66 \qquad (0.66)^2 = 0.44$$

$$(\Delta\lambda_x)^2 = 2.16 \quad \Delta\lambda_x = 1.47$$

and similarly for

$$\frac{c_1 R}{m_1^2} \Delta m_1 = 0.22 \qquad (0.22)^2 = 0.05$$

$$\frac{c_2(R+1)}{m_2^2} \Delta m_2 = 0.26 \qquad (0.26)^2 = 0.07$$

$$\frac{R}{m_1} \Delta c_1 = 0.44 \qquad (0.44)^2 = 0.19$$

$$\frac{R+1}{m_2} \Delta c_2 = 0.61 \qquad (0.61)^2 = 0.37$$

$$\left\{\frac{c_1}{m_1} - \frac{c_2}{m_2}\right\} \Delta R = 0.11 \qquad (0.11)^2 = 0.01$$

$$(\Delta\lambda_y)^2 = 0.69 \quad \Delta\lambda_y = 0.83$$

The errors have been expressed in fissions per gram per hour. Expressed in more conventional units the results become:

$$\lambda_x = (6.48 \pm 0.41) \times 10^{-3} \text{ f/g sec}$$

$$\lambda_y = (0.40 \pm 0.23) \times 10^{-3} \text{ f/g sec}$$

Several series of measurements were made and the results are summarized in the following table:

Table 3-7

Date	$\lambda_x \times 10^3$	$\Delta\lambda_x \times 10^3$	$\lambda_y \times 10^3$	$\Delta\lambda_y \times 10^3$
1943-44	6.67	0.41	0.3	1.3
Dec. '44 and Jan. '45	6.48	0.41	0.40	0.23
Feb. - March '45	7.55	0.41	0.38	0.23
Average	6.90	0.24	0.38	0.17

In the final result we have to make another correction to take into account a residual effect of cosmic rays on the apparent spontaneous fission of U^{235}, and our present best figures are

$$\lambda_x = (6.90 \pm 0.24) \times 10^{-3} \text{ f/g sec}$$

$$\lambda_y = (0.30 \pm 0.17) \times 10^{-3} \text{ f/g sec}$$

It would be possible to improve these measurements for U-235 using almost pure U-235 for the samples.

3-8 Uranium 236

This isotope of uranium is formed by an (n,γ) reaction on U-235. The isotopic composition of the sample was determined by mass spectrograph[13] and also by irradiation data.[14]

The samples were plated on platinum discs as were the other uranium isotopes and were observed in the large chambers. The spontaneous fission attributable to U^{236} was ~~observed~~ looked for but not found.

3-9 Neptunium 237

This isotope is formed by beta decay of the 7-day U-237 which in turn is obtained by an n-2n reaction on U-238. Its half life is 2.2×10^6 years.

The samples investigated were prepared in the pile and were supplied to Los Alamos by the Metallurgical Laboratory in Chicago.

An early investigation with a sample of 8×10^{-5} grams protracted so as to accumulate 127×10^{-3} gram hours of observation gave only 1 fission.

Later with 3 stronger samples (about 8×10^{-4} grams/each) we accumulated 1.113 gram hours of observation with 6 fissions recorded. This would give 1.4×10^{-3} fissions per gram per second.

The effect of a possible small contamination of plutonium or uranium in the sample is negligible as tested by slow neutron irradiation; however, it is better to consider 1.4×10^{-3} fissions per gram per second rather as an upper limit because it is difficult to be absolutely sure of the genuineness of the few fission pulses observed over 1482 hours of counting.

3-10 Neptunium 239

A sample of this material was prepared by J. Miskel from depleted uranium irradiated in the water boiler. The sample was purified from fission products, uranium, and plutonium and deposited on a platinum disc by evaporation. Its mass was determined by the growth of the alpha activity of plutonium 239 in an aliquot. Approximately 0.45×10^{-6} grams were present initially and the sample was observed for 146 hours during which time no spontaneous fissions occurred. Taking into account the decay of the Np-239, we find that the gram hours of obeervation were approximately 2.5×10^{-5}. From this we conclude that 11 fissions per gram per second is the upper limit for the spontaneous fission of Np-239.[15]

3-11 Plutonium 238

This substance is prepared by a (d,2n) reaction on U-238[16] and it has a half life of about 60 years.

The sample used was kindly supplied by G. T. Seaborg, and had been accidentally contaminated with Pu-239 in such a way that the ratio of the alpha activity of Pu-238 to that of Pu-239 was 0.895. This was determined with differential alpha range apparatus.

The Pu-238 was mounted by evaporation on platinum discs and the effective amount present was measured by observing the slow neutron fission of the contaminant plutonium 239, and using the ratio of the alpha activities quoted above.

From this last number we have

$$\frac{\text{Pu-238}}{\text{Pu-239}} = \text{ratio of alpha activities} \times \frac{T_{\frac{1}{2}}(238)}{T_{\frac{1}{2}}(239)} = 0.895 \times \frac{60}{24400} = 0.0022.$$

Hence the effective amount of Pu-238 present is 0.0022 times the effective amount of Pu-239 present.

In each of our three samples we had approximately 10^{-5} grams of Pu-239 and 2×10^{-8} grams of Pu-238 effective. The samples were observed for 830 hours total, corresponding to 18.8×10^{-6} gram hours for Pu-238 and 8.6×10^{-3} gram hours for Pu-239. 144 fissions were counted.

From this we deduce a spontaneous fission decay constant of 2.1×10^{3} fissions per gram per second. The possible contribution of other Pu isotopes to spontaneous fission is negligible, being at the most of the order of 1 per cent of the total observed.

3-12 Pu 239 and Pu 240

Plutonium 239 was investigated for spontaneous fission soon after its discovery and no fissions were detected during about 5×10^{-4} gram hours of observation.(17)

This study was pursued with the increasing amounts of plutonium that became available at successive dates.

The samples were deposited on platinum discs by evaporation, electrolytically, or by painting.

The effective amount present was generally determined by comparison with a thin standard, in a constant slow neutron flux. The thin standard was in turn alpha counted. A half life for Pu-239 of 24,300 years was used.

Material produced in the Berkeley cyclotron gave on observations extending over about 10,000 hours, 12 fissions, and this corresponds to 0.010 fissions per gram per second.

The spontaneous fission of Pu^{240} was also measured.

3-13 95^{241}

A sample of this material borrowed from Mr. Seaborg was examined. In this sample 95^{241} was mixed with a relatively large amount of lanthanum. Three plates were made each containing about 7×10^{-9} grams of 95^{241}. This weight is deduced from the alpha activity assuming a half life of 40 years. The samples were observed for a total of 2700 hours corresponding to 1.8×10^{-5} gram hours. Three fissions were registered.

For reasons stated several times we consider the resulting number, 46 fissions per gram per second, an upper limit for the spontaneous fission constant of this material.

3-14 Summary

The following table summarizes all the data accumulated up to the present time on spontaneous fission decay constants.

In column 1 the chemical symbol of the element is given; in column 2, its atomic mass A; in column 3, the total number of fissions observed in all samples; in column 4, the total number of hours over which the observations have extended. This information is important because it is clear that the possibility of spurious fissions is proportional to the duration of the observations. Column 5 gives the gram hours of observation summed over all samples. Column 6 gives the spontaneous fission constant λ in fissions per gram per second, whenever it is known, with its probable error.

The expression $<t$ means that if one fission had been observed instead of none, that would be the calculated spontaneous fission constant; this means that one has the probability $1/e$ that the spontaneous fission decay constant is larger than t. More generally it can be shown that if there have been no spontaneous fissions in a time t the probability that the decay constant is smaller than $1/\tau$ is $e^{-t/\tau}$.

Sometimes it is convenient to use the "half life for spontaneous fission", i.e. the half life of a nuclear species that would obtain if the only decay possibility were spontaneous fission. This quantity is connected with the fissions per gram per second by the relation:

$$T = \frac{1.32 \times 10^{16}}{\lambda A}$$

where T is given in years, λ in fissions per gram per second, and A is the atomic mass.

The probability λ' that a given atom undergoes spontaneous fission in one second is

$$\lambda' = 1.66 \times 10^{-24} \; A \lambda \quad (\lambda \text{ in f/g sec})$$

Table 3-14

Element	A	Fissions Observed	Hours of Observation	G Hr of Observation	λ in f/g sec
Ra	226	2			(<0.6)
Th (Io)	230	2	1326	1.45	$< 3.8 \times 10^{-4}$
Th	232	178	6300	1202	4.2×10^{-5}
Pa	231	11	2200	0.62	5×10^{-3}
U	232	3	950	5×10^{-5}	16
U	233	1	1050	1.35	$< 2 \times 10^{-4}$
U	234	0	3370	3.4×10^{-2}	$< 9 \times 10^{-3}$
U	235				$(3.0 \pm 1.7) \times 10^{-4}$
U	236				not observed
U	238				$(6.90 \pm 0.24) \times 10^{-3}$
Np	237	6	1480	1.11	$\leq 1.4 \times 10^{-3}$
Np	239	0	146	2.5×10^{-5}	< 11
Pu	238	144	833	19×10^{-6}	2.14×10^{3}
Pu	239	12	~10,000	0.33	1.0×10^{-2}
Pu	240				observed
95	241	3	2700	1.6×10^{-5}	(46)

In addition to the data given above, it is clear from our blank runs that brass undergoes spontaneous fission at a rate $<10^{-9}$ fissions per gram per second.

4. NEUTRON EMISSION IN SPONTANEOUS FISSION

A most interesting question is to find the number ν of neutrons emitted, on the average, per fission. This number is well known for slow neutron induced fission in U-235 and Pu-239 and it is clearly desirable to know it also for spontaneous fission. As a matter of fact the datum of primary interest to the project was not so much the spontaneous fission decay constant, as the number of neutrons spontaneously emitted per unit time, per unit mass, by the substance studied.

Once a pile with an appreciable multiplication was set up, it was observed that even with all sources removed, the neutron density in the pile was quite appreciable. This density was attributed to the spontaneous emission of neutrons by the uranium and by measuring it with indium detectors and by comparing it with the density produced by sources emitting a known number of neutrons, it was possible to determine the number of neutrons emitted spontaneously by uranium. In this way it was found that about 1.5×10^{-2} neutrons per gram per second were emitted by ordinary uranium.(18)

The spontaneously emitted neutrons were also detected by G. Scharff Goldhaber and G. S. Klaiber.(19) These authors obtained about 1.75×10^{-2} neutrons per gram per second of an energy above 800 Kev.

H. Pose(20) measured the neutron emission of ordinary uranium and of thorium. His results have to be corrected because he assumes that 1 millicurie Rn + Be emits 15,000 neutrons per second. If one uses the figure 11,000 which is the best estimate available, based upon 13,000 neutrons per second for 1 millicurie Ra + Be, one obtains for uranium an emission of 1.54×10^{-2} neutrons per gram per second and for thorium 0.24×10^{-3} neutrons per gram per second. The latter figure is in all probability in error because combined with the known

spontaneous fission constant of thorium, it would give ν (Th) = 5.7, which seems higher than is likely.

Hanson[21] has also measured the neutron emission from a uranium sphere with a long boron counter and found $(1.6 \pm 0.1) \times 10^{-2}$ neutrons per gram per second. Other measurements made at the Clinton pile gave a value of 1.5×10^{-2} and we think that the present best value for the spontaneous neutron emission of uranium is 1.5×10^{-2} neutrons per gram per second. It is believed that this value is accurate to about 10 per cent not including possible errors in the calibration of primary neutron standards. Since the calibration of such standards, however, enters in practically all neutron measurements in the same way, errors in the calibration cancel in relative measurements.

From this value of the neutron emission and from the spontaneous fission constant given previously, we find ν (28) = 2.2 ± 0.3.

5. THE ENERGY SPECTRUM OF SPONTANEOUS FISSION FRAGMENTS

The high spontaneous fission rate of Pu-240 makes possible the investigation of the energy spectrum of fission fragments for spontaneous fission.

An experiment to this effect was performed by Segrè and Wiegand.[22] Figure 10 shows a schematic drawing of the chamber used. The chamber was filled with argon and electrons were collected. The electrons collected gave to the grid of the preamplifier a pulse proportional to the ionization, because of the presence of the screen grid. The sample was at -1700 v and the screen grid at -900 v with respect to the collecting electrode. The positive ions did not contribute to the pulse because the decay time constant (5 microseconds) of the amplifier was too short. The time of collection of the electrons was about 1 microsecond and the time of rise of the amplifier was 0.2 microsecond. The pulse was passed from the linear amplifier to an oscilloscope and recorded photographically.

A histogram of the pulses recorded is given in Figure 11. For comparison we recorded also the pulses produced on the same sample, in the same apparatus,

by a strong source of Po + Be neutrons slowed down in water.

It is clear that the two histograms are very similar, the one of spontaneous fission pulses being perhaps slightly shifted towards lower energies.

This experiment is interesting because one may suspect that in the spontaneous fission of Pu-240, which is certainly a rare process, only very few possibilities of fragmentation exist. The slow neutron fission of Pu-239 gives an excited Pu-240 with a mean life for fission estimated to be of the order of 10^{-15} seconds, a huge factor shorter than that of the fundamental state. In spite of this, one does not see a very great change in the modes of fragmentation.

It is perhaps possible that this may be due to some re-shuffling of the nuclear matter occurring after the barrier for fission has been passed, but before the two fragments come completely apart.

This experiment could be improved by chemical investigation of the yields of the various fission chains in spontaneous fission of Pu-240 and comparison with the yields of the same chains in slow neutron fission of Pu-239.

6. THEORY OF SPONTANEOUS FISSION

Several attempts have been made to explain spontaneous fission by a mechanism similar to that invoked by Gamow in the theory of alpha disintegration.

Bohr and Wheeler[23] in their fundamental paper on fission give a calculation of the spontaneous fission probability by assuming that a nucleus comes about 10^{21} times per second into the optimum configuration for fission and that the transparency of the barrier is given by

$$\exp\left(-\frac{2\pi}{h}\sqrt{2 M E_f}\, a\right) \qquad (4)$$

in which E_f is the photo fission threshold for the nucleus in question, M the mass of the nucleus, and a a length of the order of magnitude of the nuclear radius.

They emphasize, however, that this estimate can give only the order of

magnitude of the transparency, i.e. of the exponent in the expression of the lifetime.

Attempts have been made by S. Flügge[24] and L. Turner[25] to make more precise evaluations of the spontaneous fission constant using the formula of Bohr and Wheeler and an expression for E_f given by the same authors; but they have not been successful, as shown by the following table in which we report the spontaneous fission probabilities in sec^{-1} calculated by these authors, and the experimental results.

In the case of the numbers given by Turner the probabilities have been so normalized as to give the correct value for U-238.

The weakness of the expression (4) given above for a precise calculation of the spontaneous fission constant is borne out even more by the experimental values for the photo fission threshold reported by Kock, McEllinney, Gasteiger[26] and given in column 4 of the same table.

How much the nuclear spin present in the odd isotopes may affect the transparency of the barrier is an open question.

An attempt to measure directly the transparency of the fission barrier, a little below the top, has been made along the following lines: Np-237 and Pa-231 have a neutron fission threshold of about 400 Kev. A slow neutron capture hence excites the compound nucleus to about 400 Kev below the threshold. Preliminary experiments by G. Farwell and M. Kahn give 0.1 barns as an upper limit for the fission cross section at thermal energy for both substances.

Table 6

		Flügge	Turner	Photo Fission Threshold
	$\lambda 1_{exp}$	$\lambda 1$ sec^{-1}	$\lambda 1$ sec^{-1}	CF 3292 MEV
Io	$< 1.5 \times 10^{-25}$	1.2×10^{-24}		
Th-232	1.6×10^{-26}	1.2×10^{-26}		5.40
Pa-231	2.0×10^{-24}	1.1×10^{-21}		
U-233	$< 10^{-25}$		8×10^{-20}	5.18
U-234	$< 3.3 \times 10^{-24}$	1.1×10^{-20}	6.6×10^{-21}	
U-235	$(1.1 \pm 0.7) \times 10^{-25}$	1.1×10^{-21}	1.1×10^{-21}	5.31
U-238	2.7×10^{-24}	1.2×10^{-24}	2.7×10^{-24}	5.08
Np-237	$< 5.4 \times 10^{-25}$		1.2×10^{-19}	
Pu-238	8.4×10^{-19}		6.8×10^{-17}	
Pu-239	4.0×10^{-24}	1.2×10^{-17}	8.2×10^{-18}	5.31
95241	$(< 2 \times 10^{-20})$		7.4×10^{-16}	

Now $\sigma_f / \sigma_r = \frac{1}{a}$, the ratio of the fission cross section to the capture cross section, can be expressed as

$$\frac{\sigma_f}{\sigma_r} = \frac{\nu\tau}{\Gamma_r}$$

in which ν is the number of times per second in which the nucleus comes into a configuration most favorable to fission, τ is the transparency of the barrier, and Γ_r is the probability per unit time that the compound nucleus lose its excitation by gamma ray emission.

For slow neutron fissioners like U-235 or Pu-239 we know that $1/a$ is a few units. On the other hand for these nuclei τ is supposed to be about one. We conclude from this that ν / Γ_r is of the order of a few units, say 5.

For Np-237, since the capture cross section for thermal neutrons is about 100 barns, we find that $1/a$ is at least 1000, hence

$$\frac{\bar{\Gamma}_f}{\bar{\Gamma}_r} 10^{-3} = 5 \quad \text{or} \quad = 2 \times 10^{-4}$$

where we have assumed for $\frac{\bar{\Gamma}_f}{\bar{\Gamma}_r}$ the value 5 as for other heavy nuclei. Protactinium, with a capture cross section of about 300 barns, gives a similar result.

It must be remembered that this value of the transparency is very crude and probably represents an upper limit because all experimental errors in the determination of the slow neutron fission cross sections of Np-237 and Pa-231 tend to make them appear too large.

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(2) Petrzhak, K. A. and Flerov, G.N., C.R. Acad. Sci. U.S.S.R. 28, 500 (1940); Journal General Physics 3, 275 (1940)

(3) Bohr, N. and Wheeler, A., Phys. Rev. 56, 426 (1940)

(4) The experiments were conducted mainly by O. Chamberlain, G. Farwell, J. Jungerman, E. Segrè, and C. Wiegand.

(5) Most of the samples were prepared by D. Hufford, Mary Miller, J. Miskel, and R. Potter.

(6) Linde Co., incandescent lamp grade. The purity of this argon is 99.5 per cent or better, the remainder being nitrogen. Special precautions are taken to remove all oxygen.

(7) West, D., BM 731

(8) Maurer, W. and Pose, H., ZSf. Phys. 121, 285 (1943)

(9) See e.g., Linenberger, G. A., LAMS-256

(10) West, D., BM-1028, also reports observations of the spontaneous fission of U-233 which are, however, less protracted than ours.

(11) The first work of this type was done by J. W. Kennedy and E. Segrè, A 159. See Also O. Chamberlain, G. Farwell, E. Segrè, LA-86; and O. Chamberlain, LA-517.

(12) Kennedy, J. W., and Segrè, E., UCRL-9.

(13) Williams, D., and Yuster, P., LAMS-286 and LA-512

(14) Group R-4, LAMS-256

(15) Jungerman, J., LAMS-340

(16) Seaborg, G. T. and Wahl, A. C., A-126; for the mass assignment see Kennedy, J. W., Perlman, M., Segrè, E., Wahl, A.C., A-207.

(17) Kennedy, J. W., Wahl, A. C., A-69.

(18) See e.g., E. Fermi, CP-26.

(19) G. Scharff Goldhaber and G. S. Klaiber. CN-1463.

(20) H. Pose, ZSf. Physik, 121, 293 (1943)

(21) A. O. Hanson, LA-276.

(22) C. Wiegand, LA-491

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(23) N. Bohr and A. Wheeler, Phys. Rev. 56, 426, (1939).

(24) S. Flügge, ZSf. Phys. 121, 294 (1943).

(25) L. Turner, Rev. Mod. Phys. 17, 292 (1945).

(26) H. W. Kock, J. McEllinney, E. L. Gasteiger, CF-3292.

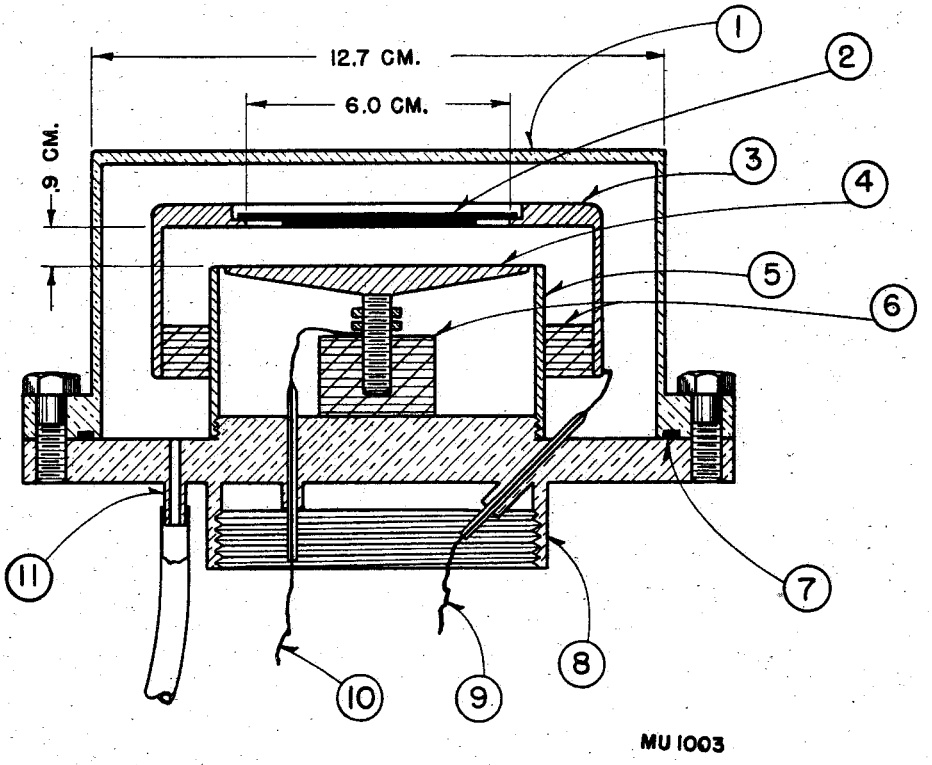

Figure 1
Argon Chamber

1. Brass Cones
2. Mounting Sample
3. Sample Holder and High Tension Electrode
4. Collecting Electrode
5. Guard Ring
6. Polystyrene Insulating Supports
7. Rubber Gaskets
8. Threaded Collar Fastens Chamber to Amplifier Chassis
9. High Tension Lead (Platinum Glass Seal Waxed in Place)
10. Grid Lead
11. Gas Outlet

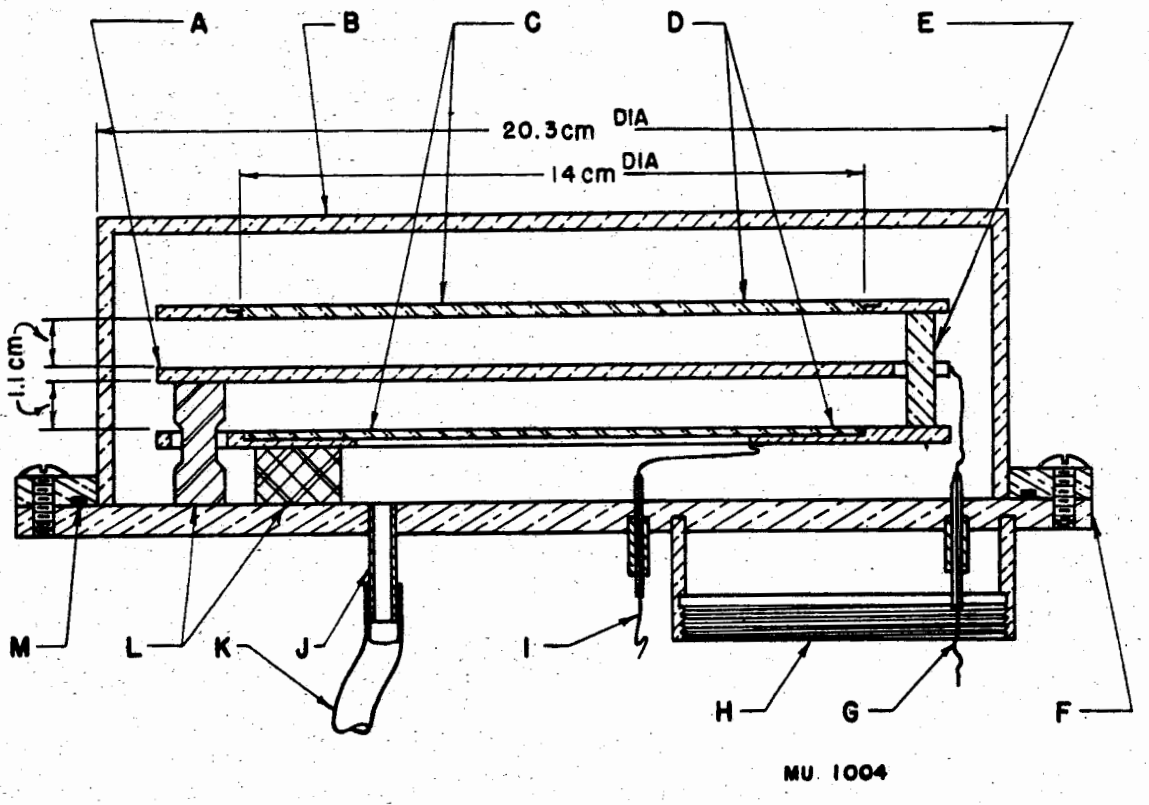

Figure 2

Double Desk Nitrogen Chamber

A. Collecting Electrode
B. Brass Cover
C. Sample Holders and High Tension Electrodes
D. Mounted Samples
E. Brass Supports
F. Brass Base Plate
G. Grid Lead
H. Threaded Collar Fastens Chamber to Amplifier Chassis
I. High Tension Lead (Platinum Glass Seal Waxed in Place)
J. Gas Outlet
K. Rubber Hose and Clamp
L. Polystyrene Insulating Supports
M. Rubber Gasket

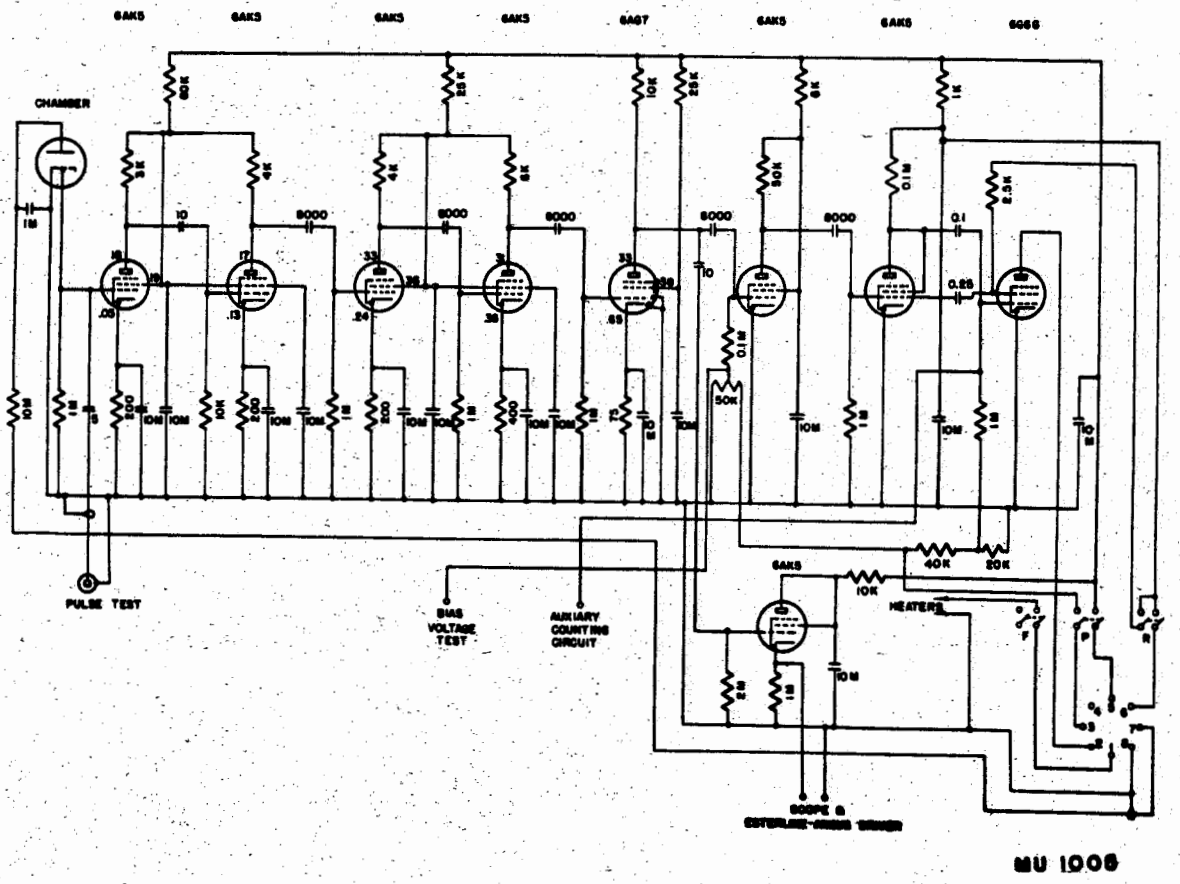

Figure 3

Fast Rise Time Battery Amplifier

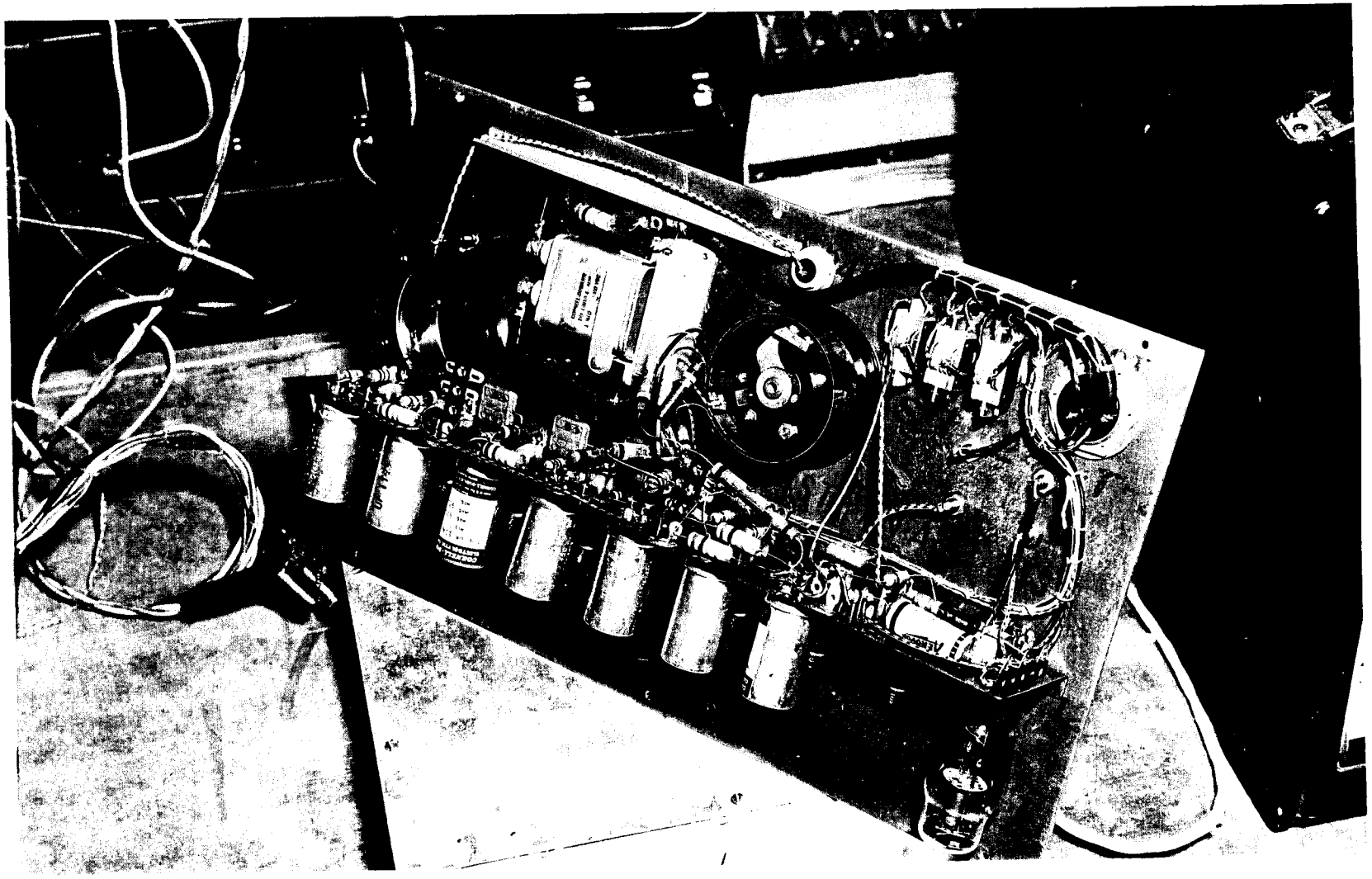

FIG. 4

PHOTOGRAPH OF BATTERY AMPLIFIER

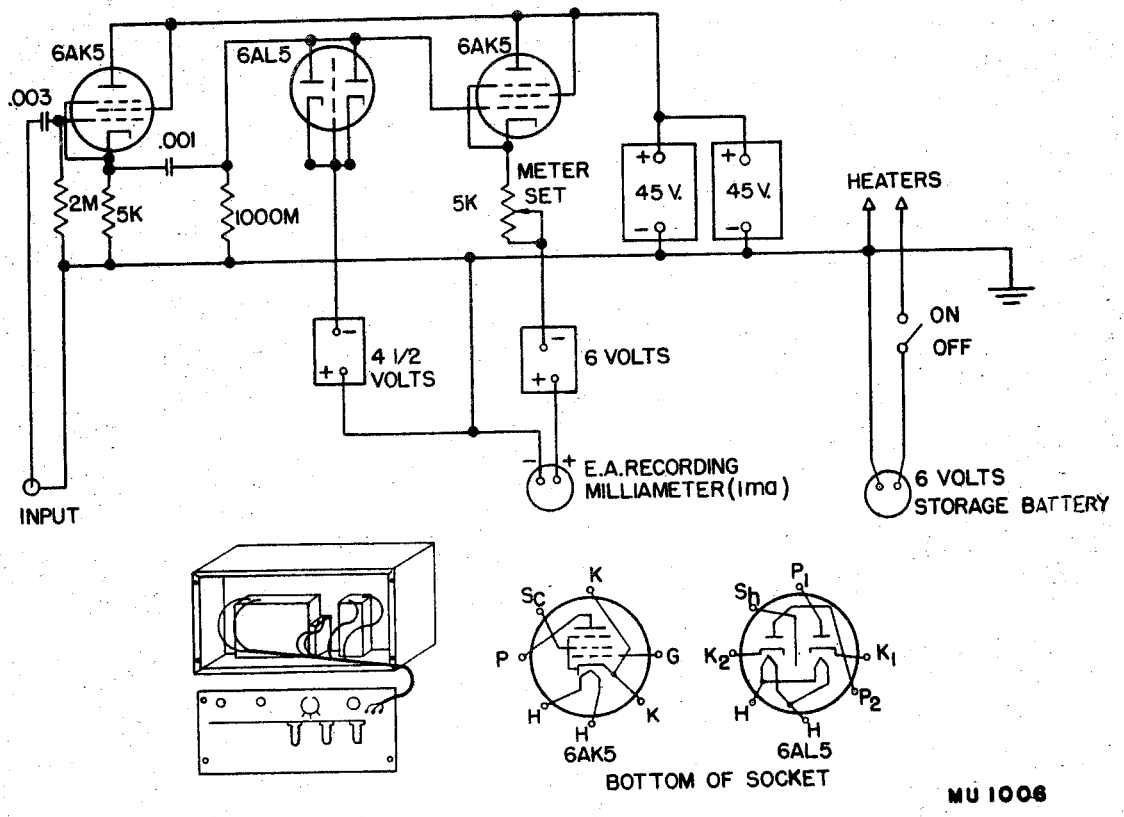
6AK5
.003
6AL5
6AK5
.001
2M
5K
1000M
5K
METER SET
45 V.
45 V.
HEATERS
ON
OFF
4 1/2 VOLTS
6 VOLTS
E.A.RECORDING MILLIAMETER(1ma)
6 VOLTS STORAGE BATTERY
INPUT
K
Sc
P
G
H
H
K
6AK5
P1
Sh
K2
K1
H
P2
H
6AL5
BOTTOM OF SOCKET
MU 1006

Figure 5

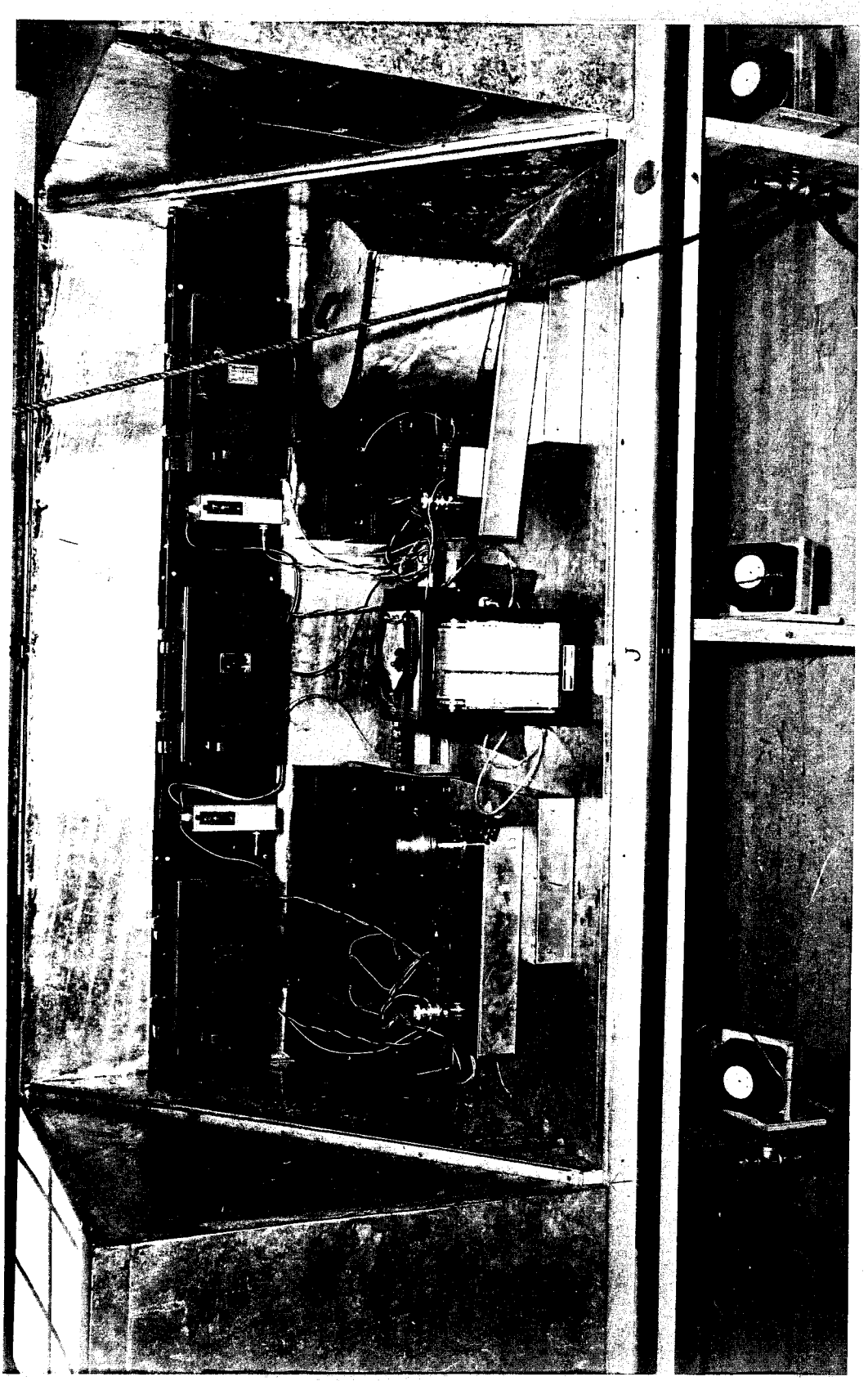

FIG. 6

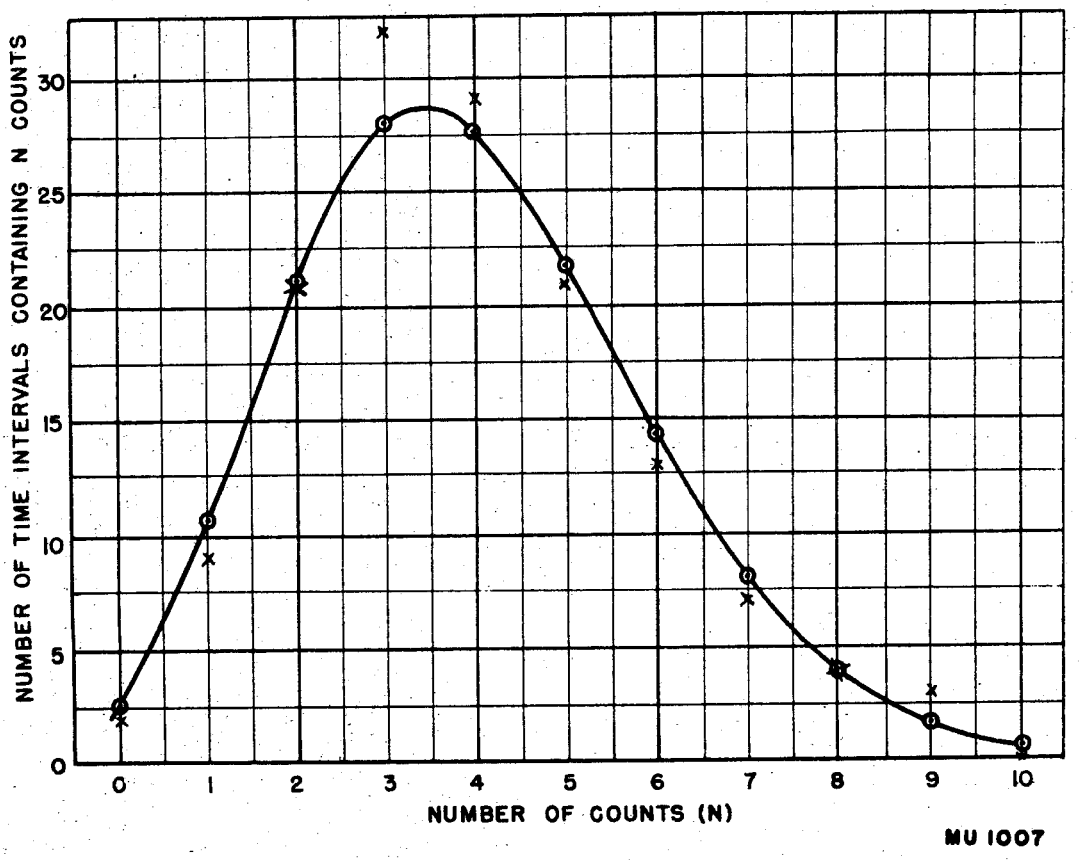

Figure 7

Time distribution of Uranium
Spontaneous Fissions

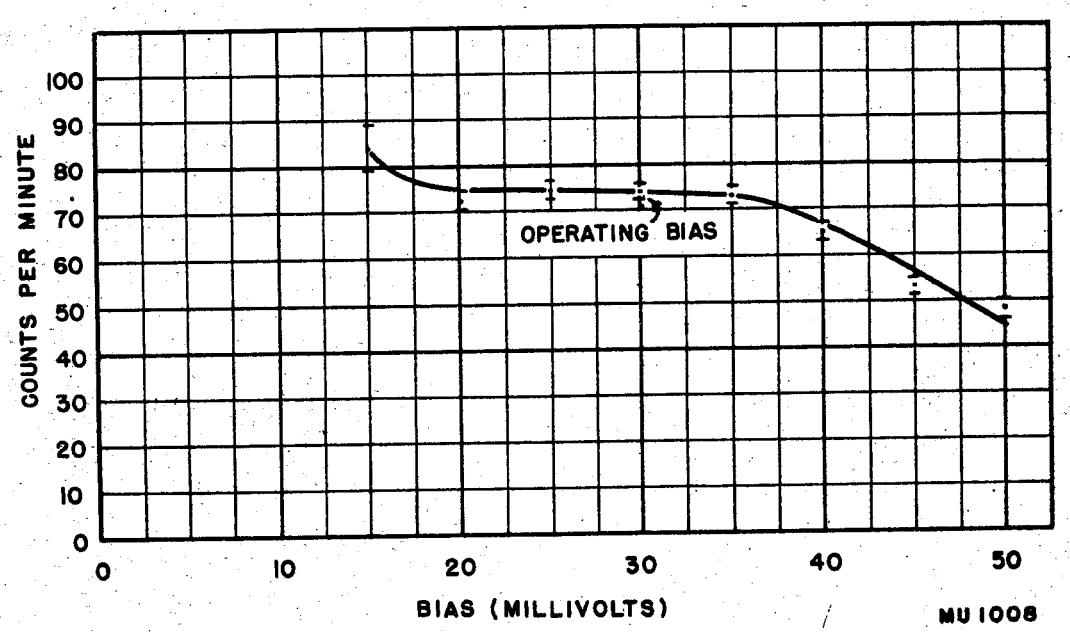

Figure 8

Fission Bias Curve

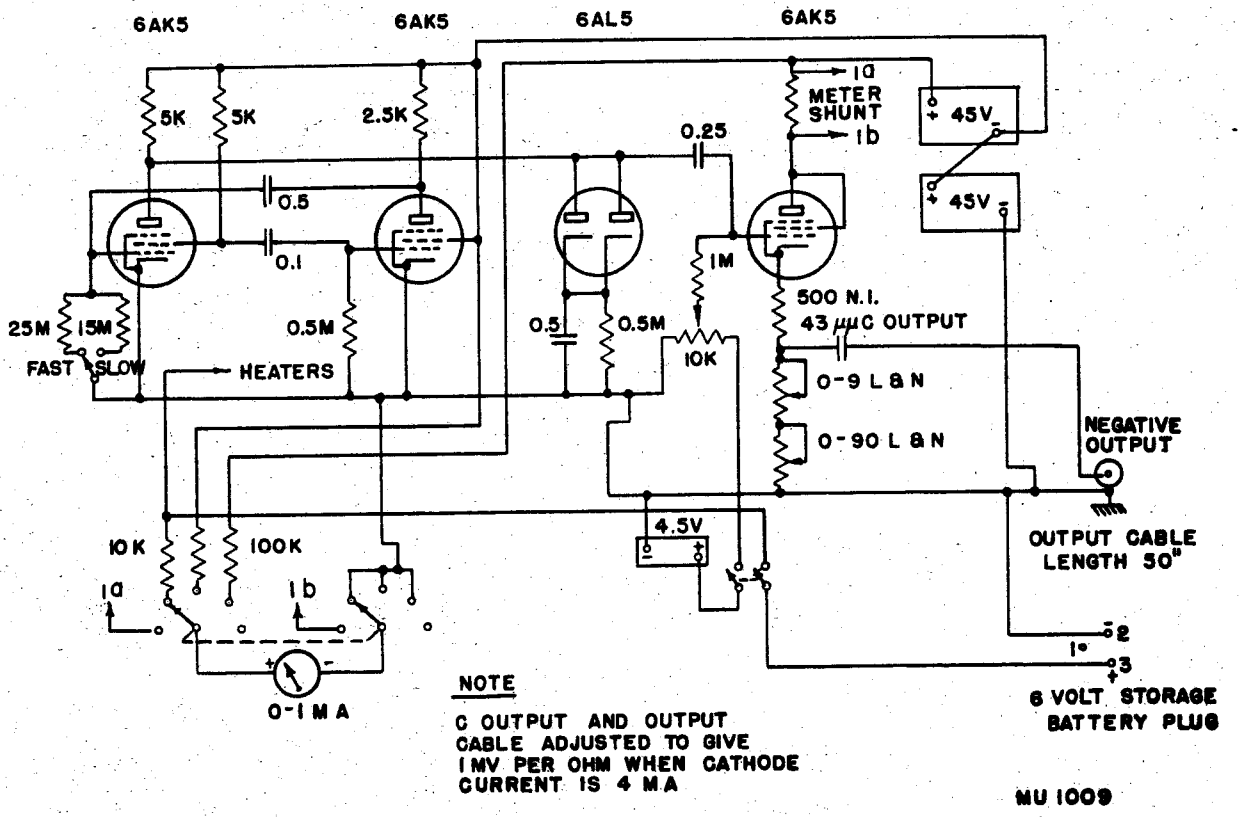

Figure 9

Fast Rise Time Pulse Generator

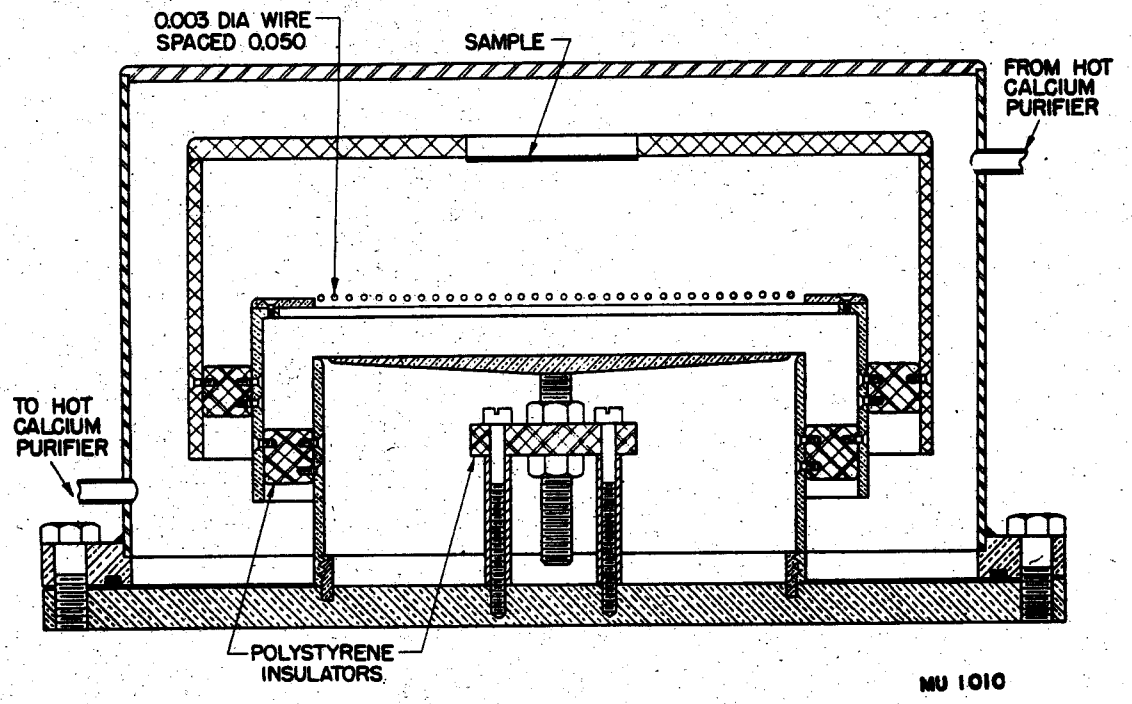

Figure 10
Chamber for Measuring Fission
Fragment Energies

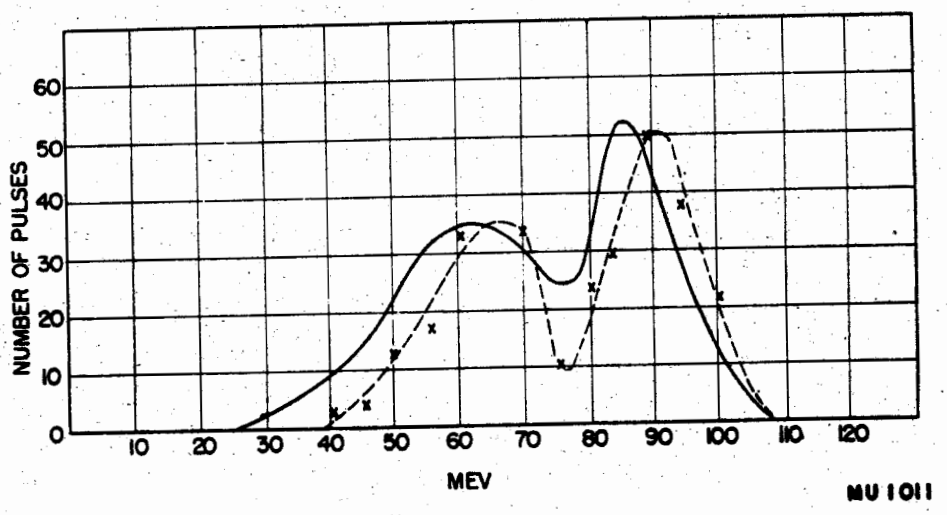

Figure 11

Energy of Fission Fragments