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INVESTIGATION OF FISSION GAMMA RAYS

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ABSTRACT

The emission of gamma rays accompanying the fission process of 25 was investigated by the coincidence method. It was found that the total energy of the gamma rays is 5.1 Mev per fission and that the individual quanta have an average energy of about 1 Mev. No delayed gamma rays have been observed.

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INVESTIGATION OF FISSION GAMMA RAYS

INTRODUCTION

A knowledge of the number and the quantum energies of the gamma rays accompanying the fission process seems desirable not only from the point of view of obtaining a better understanding of the process itself, but also because those gamma rays might be utilised in experiments designed to test the gadget or other types of chain reactions.

The "prompt" fission gamma rays which were investigated in this experiment, i.e., those emitted within less than 2 microseconds of the fission process, may be expected to originate from the highly excited fragments long before their radioactive decay. It is not excluded that some gamma rays may be emitted by the compound nucleus 26 before fission. (We have limited our study to fission induced by thermal neutrons.) In order to investigate the fission gamma rays it is necessary to adopt a technique which will distinguish them from those due to radiative neutron capture by the various uranium isotopes and by other materials present, from those due to the radioactive decay of the fission and capture products, and from those emitted by the neutron source. The method adopted in our experiments was to observe coincidences between fission fragments in an ionisation chamber and gamma-ray pulses in a Geiger-Mueller counter.

Let us denote by ε_f the efficiency of the ionisation chamber used for detecting fissions (ε_f is very nearly unity); by $\Sigma\,\varepsilon_\gamma$ the sum of the efficiencies (including the effect of solid angle) of the gamma-ray counter for detecting the various gamma rays accompanying the fission process (if in the various modes of the fission process different types of gamma rays are

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emitted, $\Sigma\varepsilon_\gamma$ represents the average of the sums for the various modes; the individual ε_γ are of the order of 10^{-3}). Further let N_o be the number of fissions taking place per second, N_f the number of these observed in the fission chamber, N_γ the observed number of gamma-ray counts per second, N_B the number of these due to causes other than prompt fission gamma rays, N_c the rate of observed coincidences and N_r the number of these due to chance, i.e., due to gamma rays from other sources arriving within the resolving time of the instrument. Then

$$N_f = N_o\varepsilon_f; \quad N_\gamma = N_o\Sigma\varepsilon_\gamma + N_B; \quad N_c = N_o\varepsilon_f\Sigma\varepsilon_\gamma + N_r$$

from which we obtain $\Sigma\varepsilon_\gamma = (N_c - N_r)/N_f$ (1)

The rate of chance coincidences is given by the expression

$$N_r = N_f N_\gamma(\tau_1 + \tau_2)$$

where τ_1 and τ_2 are the durations of the two pulses at the coincidence circuit. We shall return to this last equation in the next section.

Since $\Sigma\varepsilon_\gamma$ can be determined, as shown in the above formula, the total energy carried by the gamma rays could be found if the spectral distribution of the fission gamma rays and the energy dependence of ε_γ were known. Fortunately, however, ε_γ for a counter constructed entirely of materials of low atomic number is very nearly proportional to the quantum energy so that $\Sigma\varepsilon_\gamma$ depends mostly on the total energy and a fairly rough determination of the average quantum energy is sufficient. If we denote this average energy by $\bar{E}$ and the efficiency of the counter for its detection by $\bar{\varepsilon}_\gamma$--this quantity can be determined by making use of known radioactive schemes as described

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later -- then the total energy of the "prompt" fission gamma rays is

$$E = \Sigma \varepsilon_\gamma \quad \overline{E}/\varepsilon_\gamma \qquad (2)$$

The determination of the mean quantum energy $\overline{E}$ was carried out by two methods. The first of these is based on the fact that the energy dependence of ε_γ is markedly different for a counter built of material with high atomic number than for one of low atomic number. Thus the ratio of the values of $\Sigma\varepsilon_\gamma$ determined with two such counters may be used to determine the average γ-ray energy, if the ratio of the efficiencies of these two counters is known for various gamma quanta. This can be achieved by calibration with gamma rays from radioactive substances with known disintegration schemes.

In the second method the single gamma-ray counter is replaced by a pair of counters equipped with thin mica windows. These counters are coupled in a coincidence circuit so that counts are registered only when a secondary electron produced in one counter enters the other one after traversing the windows. The output of this circuit is in turn used in coincidence with the fission-chamber pulses. By observing the dependence of the triple-coincidence rate on the thickness of absorbers placed between the two Geiger-Mueller counters the energy of the secondary electrons and thus of the gamma rays producing them can be determined. The calculation of the various background effects in this method is discussed in the appendix.

APPARATUS AND PROCEDURE

Inspection of the equations used to determine $\Sigma\varepsilon_\gamma$ given in the preceding section shows that for high accuracy it is desirable that $N_r \ll N_c$.

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i.e., that only a small fraction of the observed coincidences should be due to chance. This can always be obtained by making the source strength sufficiently small since the rate of chance coincidences is nearly proportional to the square of the source strength. However this leads to small counting rates and consequent statistical uncertainty in the results. In order to be able to use satisfactory counting rates while keeping N_r small the following conditions should be fulfilled: 1) ε_γ as large as possible, i.e., large solid angle subtended by the Geiger-Mueller counter at the fission chamber; 2) N_B as small as possible, i.e., proper selection of neutron source and of materials used in construction of parts exposed to neutron flux. 3) τ_1 and τ_2 as small as possible, i.e., electron collection in the ionisation chamber and fast response of electronic circuits. The width of the pulses cannot, however, be made much smaller than the rise time of the pulses in the Geiger-Mueller counter, which is of the order of several microseconds, otherwise some coincidences would be missed.

Condition 2) places very rigid limitations on the choice of materials which must have small neutron capture cross sections. We have used bismuth and lead as conductors and polyTFE as insulators wherever possible, despite their poor structural properties. Even so the fission gamma rays constituted only about 1 to 2 per cent of the total observed. If conventional materials such as aluminium or copper and lucite had been used this fraction would have been further decreased at least by a factor of five, rendering the experiment impracticable. The same holds true if hydrogen had been used as a slowing medium instead of graphite.

Fig. 1 shows a diagram of the apparatus. The cylindrical fission

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chamber was constructed of 1-mm-thick polyTFE and subdivided into four separate concentric compartments, with about 4-mm electrode spacing, by three foils of one-mil dural. Both the inner and the outer cylindrical polyTFE walls were coated with acquadag on the side facing the dural foils, thus forming another two conducting surfaces. Two of the foils were coated on both sides with a deposit of 0.4 mg/cm^2 of U_3O_8 containing about 20 per cent of 25; the total amount of 25 was thus about 30 mg. These films were prepared for us by Cpl. Miller of Group CM4. Alternate conductors were connected together, the group of three (two of acquadag and one of dural) carrying high tension of about +1200 volts, while the group of two on which the U_3O_8 was deposited was connected to the grid of the first preamplifier tube. The chamber was operated with room air, the high applied field permitting electron collection.

The Geiger-Mueller counter was placed on the axis concentric with the chamber. The design of the counter is also shown if Fig. 1. Two "single" counters of identical dimensions were constructed, one with a polyTFE wall (1 mm thick) coated on the inside with acquadag, and one with a lead wall.

The construction and assembly of the counters used in the coincidence-absorption experiment is shown in Fig. 2; both these counters have lead walls. In the experiment they were placed in the position of the "single" counter in Fig.1; the lead to the second counter passed through a hole in the other bismuth wall. All counters were filled with a Trost mixture of alcohol (1 cm Hg) and argon (10 cm Hg). The entire assembly was surrounded by a bismuth shield 3.5-cm thick and placed in a

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thermal-neutron flux in the graphite block near the cyclotron in building X.

Very careful electrical shielding was found necessary between the electrodes and leads of the ionisation chamber on one hand and those of the Geiger-Mueller counter on the other. Since the pulse in the counter is about 1,000 times as great as that from the fission particles a very small coupling will have serious effects in the "fission channel" of the circuit. In fact, before the shielding had been completed satisfactorily, the rather baffling result was obtained that the ionisation pulses due to fissions accompanied by gamma rays seemed to be smaller than the pulses not accompanied by gamma rays; this was found to be due to the fact that the positive ionisation pulse was reduced by a small cross-fed negative pulse from the Geiger-Mueller counter whenever a gamma ray was counted.

Fig. 3 shows a block diagram of the counting equipment used in the experiment with a single counter. The preamplifier and linear amplifier were of the standard medium-fast type and the rise time of the pulse was about 2.5 microseconds. The time constants of the blocking oscillators could be changed by means of a selector switch permitting a variation of the gate widths τ_1 and τ_2 from about 2 to 5 microseconds. The sum $\tau_1 + \tau_2$ was determined directly by feeding regular pulses into one channel and random pulses into the other, or alternatively by introducing a long delay (about 300 microseconds) into one of the channels. It is probable that a slight gain in the resolving time could be obtained by using faster amplifier circuits but the Geiger-Mueller counter does not permit truly "fast" operation with resolving times lower than a microsecond.

The use of three scalers made it possible to record simultaneously the rates of fissions, gamma rays, and coincidences between them, thus

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reducing the experimental error due to variation of the beam strength.

The variable-delay circuit, permitting the introduction of a delay from 2 μsec to 300 microseconds, could be incorporated into either channel, and was used to compensate the phase differences in the two channels and also to search for delayed gamma rays.

Most data were obtained with $\tau_1 = \tau_2 = 2.8$ microseconds, but the same results were obtained with gate widths of 4.1 microseconds; the results also remained unchanged when the phase of one channel was shifted over a certain range with respect to the other. In Fig. 4 the ratio of true coincidences to the fission count is plotted as a function of the delay in either channel. The points in Fig. 4A were obtained with a gate width of 2.8 μsec and those in 4B, with one of 4.1 μsec. It is seen that the points lie well on the curves showing the results expected for the given gate widths and rise time of the pulses. The fact that the coincidence rate is constant over the width of the pulse proves that all coincidences occurred within the resolving time.

Fig. 5 shows characteristic curves of the fission chamber. Curve I was obtained with an integral discriminator, i.e., it represents the rate of fissions as a function of the minimum firing pulse. Curve II shows the results with a differential discriminator, i.e., the actual distribution of the heights of fission pulses. As one would expect in a chamber of this type, the "plateau" is neither very long nor perfectly flat but appears good enough to ensure counting of at least 90 per cent of all fissions without interfering noise.

In the experiment with the two Geiger-Mueller counters, used in

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determining gamma-ray energies, an additional coincidence circuit with a resolving time of four μ sec was used and the output of that circuit fed into discriminator B on Fig. 3. The scaler C registered then triple coincidences between the two Geiger-Mueller counters and the fission chamber.

CALIBRATION OF THE GAMMA-RAY COUNTERS

The method of calibrating the efficiency of gamma-ray counters by coincidence counting is well known, and many known radioactive-disintegration schemes can furnish calibration points for different quantum energies. Unfortunately the most convenient substances such as Co^{60} and Mn^{54}, which emit nearly monochromatic radiations, were not readily available. It is, however, possible to calculate the energy dependence of the efficiency with reasonable accuracy from the known abosrption of gamma rays and electrons in the counter wall; these calculations are known from experience to describe the behavior of counters such as ours. This allows the use of substances with complex, but known, gamma radiations as calibration points.

The procedure of calibration was the following: a radioactive source was placed on one of the two dural foils carrying the uranium deposit and the gamma-ray rate recorded on the Geiger-Mueller counter (lead or polyTFE). A known fraction of the source was then placed, in a standard position, near another Geiger-Mueller counter which had been calibrated by the standard coincidence method. From the comparison of the observed activities the average efficiency of the investigated source spread uniformly over both dural foils was calculated by considering the areas of the foils.

The active substances used were Au^{198}, Mn^{56} and Na^{24} whose gamma rays cover the energy range from 0.42 Mev to 2.76 Mev. The calibration of

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the "standard" counter used for Au^{198} and Mn^{56} has been described in LA-100. The procedure for Na^{24}, which emits gamma rays of 1.38 and 2.76 Mev, was analogous. The results obtained in this way were then used to normalise the efficiency curve calculated at M.I.T. (Peacock, Ph.D. thesis, 1944). The curve for the lead counter was corrected for absorption of gamma rays in the wall. Fig. 6 shows the calculated efficiency-versus-energy curves normalised in this fashion and Table I shows the fit of the curves with the observed efficiencies. The curves represent the actual efficiencies of the counters used with a probable error of about 5 per cent in the absolute values.

TABLE I

	PolyTFE Counter		Lead Counter	
	Calculated	Observed	Calculated	Observed
Au^{198}	0.30×10^{-3}	0.30×10^{-3}	1.05×10^{-3}	1.03×10^{-3}
Mn^{56}	1.88 "	1.94 "	2.24 "	2.23 "
Na^{24}	3.88 "	3.84 "	4.03 "	4.06 "

In order to be able to determine the average energy of the gamma rays from the recoil-electron absorption curve the method was calibrated by measuring similar absorption curves for gamma rays of Na^{24}, Y^{86}, La^{140}, Ta^{181} and Au^{198}. In some cases the radiations were filtered through 3.5 cms of bismuth. Fig. 7A shows the average slope of the logarithmic absorption curves of the secondary electrons in lead as a function of the average gamma-ray energy. In calculating the average energy the variation of counter efficiency with energy and the effect of the bismuth shield where used were taken into consideration. In Fig. 7B the initial transmission, i.e., the

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ratio of coincidence rate to single counting rate with no absorber other than the counter windows, is plotted as a function of gamma-ray energy. This initial transmission defines the average probability for an electron produced in one counter to produce a pulse in the second counter.

RESULTS

Typical sets of data obtained in the measurements of $\Sigma\varepsilon_\gamma$ are shown in Table II; the first five measurements were taken with a resolving time of 2.8 µsec, the last three with 4.1 µsec. All these data were obtained with the lead counter; the setting of the fission-chamber discriminator was constant during these measurements. Curve III of Fig. 5 shows the value of $\Sigma\varepsilon_\gamma$ obtained for various settings of the discriminator bias in the fission chamber. As is seen, the coincidence rate is constant within the experimental error over the investigated range of fission pulses. It ought, however, to be pointed out that owing to the geometry of the chamber this method is not sensitive enough to detect a dependence of the energy of the gamma rays on the energy of the fission fragments.

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TABLE II

Time of measurement in minutes	Fissions recorded	Gamma rays recorded	Coincidences recorded	Chance coincidences calculated	$\Sigma\varepsilon_\gamma$
12	38976	40128	335 ± 18	12	$8.3 \pm 0.5 \times 10^{-3}$
16	43008	50240	335 ± 18	13	7.5 ± 0.4 "
6	31252	29248	267 ± 16	14	8.1 ± 0.5 "
6	29184	27840	229 ± 15	13	7.4 ± 0.5 "
10	42560	44416	356 ± 19	18	7.9 ± 0.4 "
10	90800	75776	832 ± 29	94	8.1 ± 0.3 "
10	78592	68096	716 ± 27	73	8.2 ± 0.3 "
12	91456	75072	817 ± 28	78	8.1 ± 0.3 "

The final results for $\Sigma\varepsilon_\gamma$ were: polyTFE counter $\Sigma\varepsilon_\gamma = 5.40 \pm 0.15 \times 10^{-3}$ lead counter $\Sigma\varepsilon_\gamma = 8.00 \pm 0.10 \times 10^{-3}$.

The absorption of the recoil electrons due to fission gamma rays is shown on Curve I on Fig. 8 where the transmission is plotted against the thickness of the lead absorber. Curve II gives the absorption of secondary electrons from the "background", i.e., those due to capture of neutrons in other elements, mainly in carbon. It is seen that those gamma rays are harder than the fission gamma rays. The slope of a logarithmic plot of the same data after subtracting background effects was 3.35 ± 0.45 for the fission gamma rays (in same units as those drawn on Fig. 7A) and the initial transmission without lead absorber was 0.032 ± 0.002.

From the ratio of the values of $\Sigma\varepsilon_\gamma$ and the curves in Fig. 6 one deduces an average quantum energy for the fission gamma rays of 0.8 ± 0.1 Mev;

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from the slope of the absorption curve of the secondary electrons (Fig. 7A), 1.5 ± 0.2 Mev; and from the initial transmission (Fig. 7B), 1.2 ± 0.1 Mev. The apparent discrepancy indicates a fairly wide spread in quantum energies; since the several methods weigh the various quantum energies differently in arriving at an average, the ratio of the $\Sigma \varepsilon_{\gamma}$ values favors the softer gamma rays while in the absorption method the harder quanta are more heavily weighed. It appears that about 1 Mev is a reasonable value for the average energy.

The total energy of the "prompt" fission gamma rays can be calculated according to formula (2) by assuming a value for the average quantum energy of the gamma rays and the corresponding efficiency of the counter from the curves on Fig. 6. In Table III we show the values of the total energy calculated for three values of energies assumed to be the average quanta of the fission gamma rays. Those values were calculated for the polyTFE counter and as would be expected they are nearly independent of the assumed value of the individual energy within the range of interest. The lead counter is not suitable for the determination of the total energy since its efficiency-versus-energy curve deviates too much from proportionality.

TABLE III

$\overline{E}$ Mev	E Mev
0.7	5.2 ± 0.3
1.0	5.0 ± 0.3
1.5	5.2 ± 0.3

From these figures we calculate that the total energy of the fission gamma rays emitted within two microseconds is 5.1 ± 0.3 Mev.

DELAYED GAMMA RAYS

The possibility that some of the fission fragments may be formed

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in a metastable state was investigated by searching for the existence of a delay between the fissions and the gamma rays. From the curve on Fig. 4 it is seen that the rate of true coincidences drops rapidly with increasing delay and reaches zero at a value to be expected for "instantaneous" gamma rays, after taking into account the gate width of the coincidence circuit. With longer delays all observed coincidences were found to agree with the value calculated for the chance ones within the experimental error. Thus we can state that no delayed gamma rays were observed. From the statistical error of the results with a longer delay we can calculate the upper limit of the fraction of gamma rays emitted with a given delay period. Results of these calculations are represented by the curve on Fig. 9 where the ordinates give the maximum percentage of the fission gamma rays which could be emitted with a mean life time drawn as abscissa. As is seen the results of these experiments do not exclude the possibility of a fairly high fraction of gamma rays being emitted with a period longer than 100 μsec, but from other considerations this seems rather unlikely.

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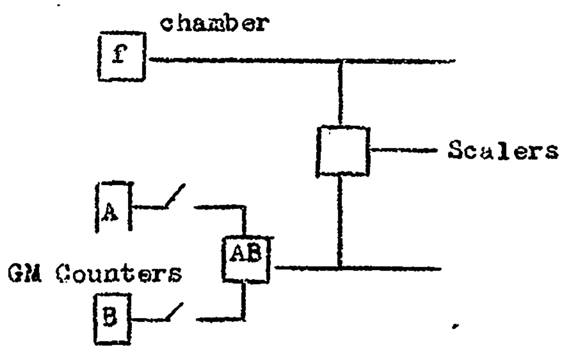
APPENDIX

In analyzing the "triple-coincidence" experiment one has to deal with a somewhat complex array of background effects. The accompanying sketch shows the connection of the several channels. The first coincidence circuit (A,B) is arranged in such a way that either coincidence pulses or pulses from one or the other of the G-M counters A and B can be mixed with fission pulses. In Table IV we denote the gate width of the several channels by $\tau_A = \tau_B = \tau_1$ for those entering the circuit (A,B) and τ_2 for the second coincidence circuit respectively. The counting rates in the three input channels are denoted by the letters A, B and f. Quantities in square brackets denote coincidence counting rates, e.g., $[Af]$ means the rate of coincidences between fissions and counts in counter A, etc. The subscript t outside a square bracket denotes true (i.e., causally connected) coincidences, and the subscript r denotes chance (random) coincidences. Thus $\left[[AB]_t f\right]_r$ is the rate at which fissions coincide by chance with truly coincident counts in A and B. The letter T refers to the probability that a secondary electron will enter one counter after having caused a count in the other one. T_f is this "transmission" in the case of fission gamma rays, T_o in the case of other gamma rays. Both of these quantities depend of course on the thickness of absorber between the two counters. The subscript γ refers to counting rates obtained with 0.7 gm/cm^2 of absorber, deemed sufficient to stop all recoil electrons without seriously affecting the background effects.



The quantity used in determining the gamma-ray energy is T_f obtained with several absorber thicknesses. It can be seen from the quantities listed in

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the table that

$$T_f = \left[[ABf] - [ABf]_r - [ABf]_{t_\gamma}\right] / \left[[Af] + [Bf] - 2\tau_2 f\ (A + B)\right]$$

The ordinates in Fig. 8 are plotted without subtracting $[ABf]_\gamma$. In determining the gamma-ray energy from Fig. 7b, T_f without absorber other than the counter windows, is used. For the determination from Fig. 7a a plot of log T_f vs absorber thickness is made and the slope of this entered in the curve in Fig. 7a. Table V shows a typical set of values obtained for the various quantities used in determining T_f. It should be borne in mind that the method of taking data was such that several background effects were observed together so that errors in calculating the several effects will tend to compensate each other. In calculating T_f from the several quantities, the results of several runs must, of course, be reduced to the same source strength. Thus, the five runs shown in Table IV, when reduced to a rate of 15,000 fissions per minute, yield

$$[ABF]_t = [ABf] - [ABf]_r = 4.8 \pm 0.4;\ [ABf]_{t_\gamma} = 0.4;\ [Af] + [Bf] - 2\tau_2 f(A + B) = 142$$

so that, from these runs, we find, for zero absorber

$$T_f = (4.8 - 0.4)/142 = (3.1 \pm 0.3) \times 10^{-2}$$

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TABLE IV

Connection	Observed Quantities	Coincidences
A and f	A, f, $[Af]$	True: $[Af]_t = f\Sigma \varepsilon_{\gamma A}$ Chance: $[Af]_r = 2\tau_2 Af$
B and f	B, f, $[Bf]$	True: $[Bf]_t = f\Sigma \varepsilon_{\gamma B}$ Chance: $[Bf]_r = 2\tau_2 Bf$
A and B	A, B, $[AB]$, $[AB]_\gamma$	True $[AB]_t$ $\{$ Recoil electrons accompanying fission: $\frac{f}{\varepsilon_f}(\Sigma \varepsilon_{\gamma A} + \Sigma \varepsilon_{\gamma B}) T_f$ Recoil electrons not accompanying fission: $(A + B)\ T_o$ Cosmic ray plus gamma-gamma: $[AB]_\gamma - [AB]_{r\gamma}$ Chance: $[Ab]_r = 2\tau_1\ AB$
AB and f	f, $[AB]$, $[ABf]$, $[Abf]_\gamma$	True $[ABf]_t$ $\{$ fission and recoil electrons: $\left([Af]_t + [Bf]_t\right) T_f$ fission and gamma-gamma: $[ABf]_\gamma - [ABf]_{r\gamma}$ Chance $[ABf]_r$ $\left\{ \left[[AB]_t f\right]_r + \left[[AB]_r f\right]_r + \left[[Af]_t B\right]_r + \left[[Bf]_t A\right]_r = 2\tau_2\ [AB]\ f + 2\tau_1\left([Af]_t B + [Bf]_t A\right) \right.$

TABLE IV

Typical results of the "triple-coincidence" experiment. Counts per minute.

Absorber gm/cm²	τ_2 (μsec)	τ_1 (μsec)	f	A	B	[Af]	[Bf]	[AB]	[ABf]	$[ABf]_r$ (calc.)	$2\tau_2 f(A+B)$ (calc.)
0	4.1	3.1	14976	7104	9792	70 ± 4	95 ± 4	640 ± 9	6.2 ± 0.6	1.45 ± 0.02	35
0	4.1		16896	8192	9472	89 ± 5	109 ± 6	723 ± 9	6.6 ± 0.9	1.82 ± 0.02	41
0	2.9		15360	8064	8192	97 ± 7	100 ± 7	698 ± 8	6.6 ± 0.8	1.21 ± 0.02	24
0.7	2.9		9856	4960	4672	41 ± 3	62 ± 6	32 ± 2	0.25 ± 0.17	0.07	10
0.7	2.9		5952	2880	2701	27 ± 2	43 ± 3	21 ± 1	0.18 ± 0.08	0.03	3

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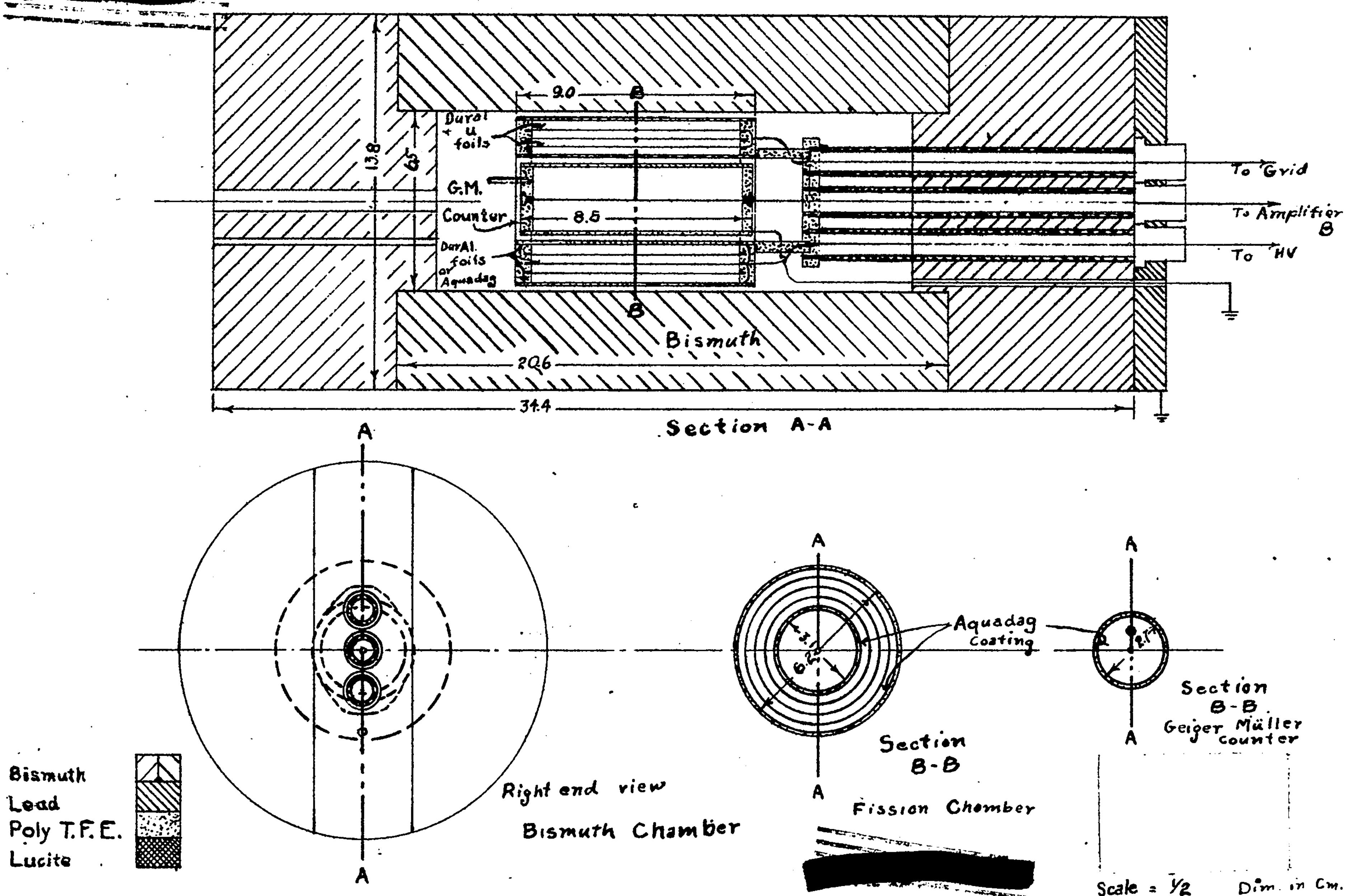
Figure 1



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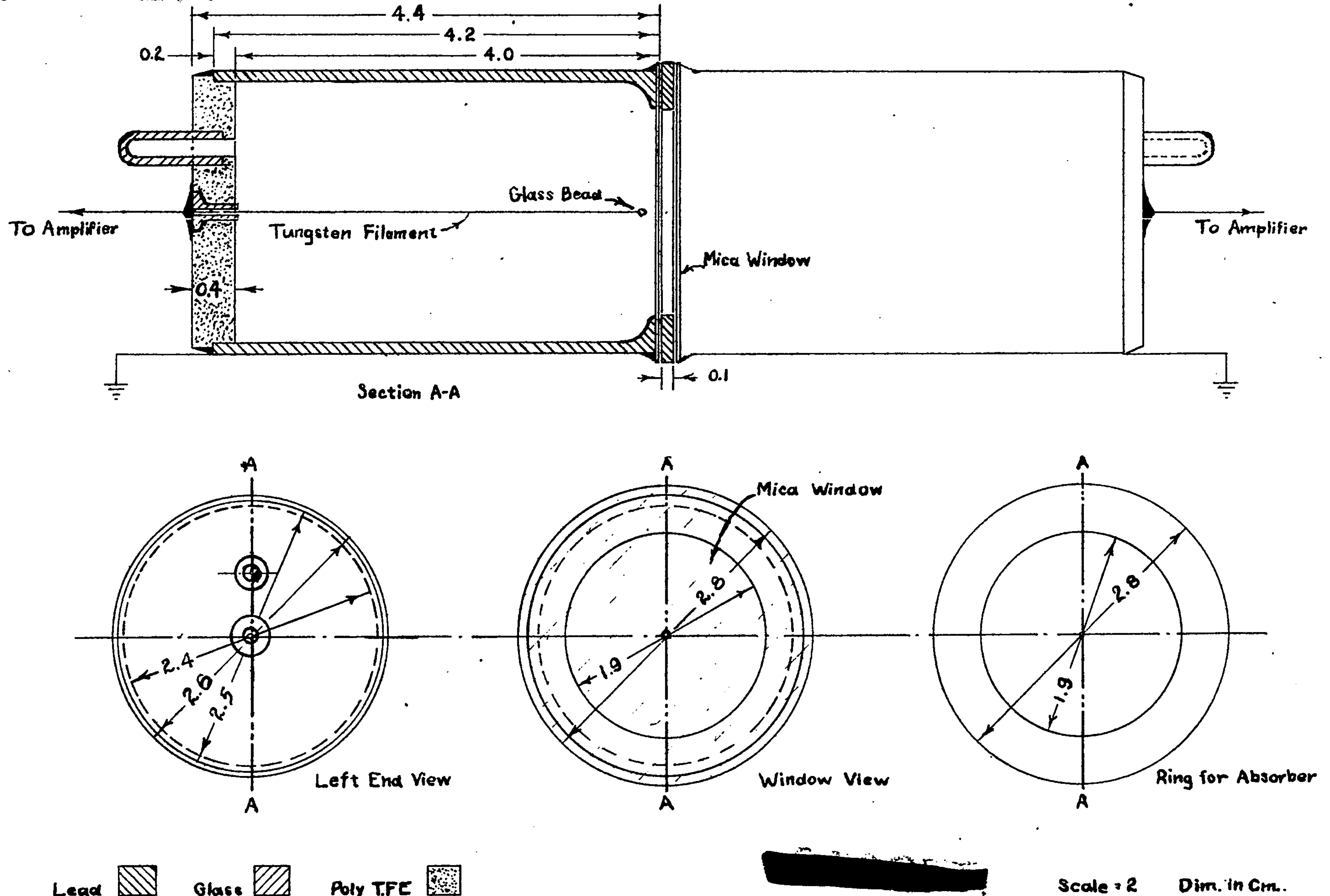

Figure 2

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Figure 3

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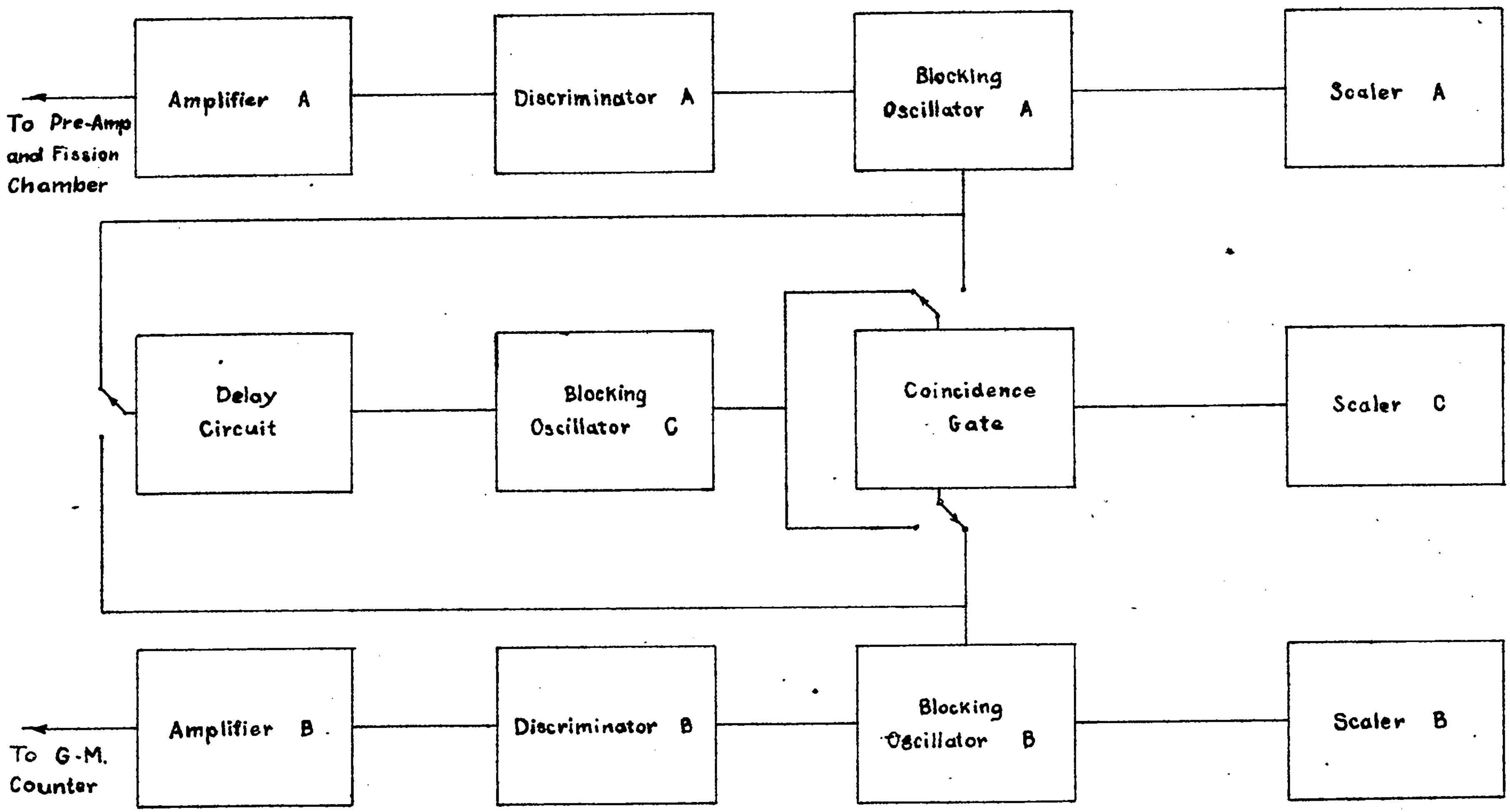

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Figure 4

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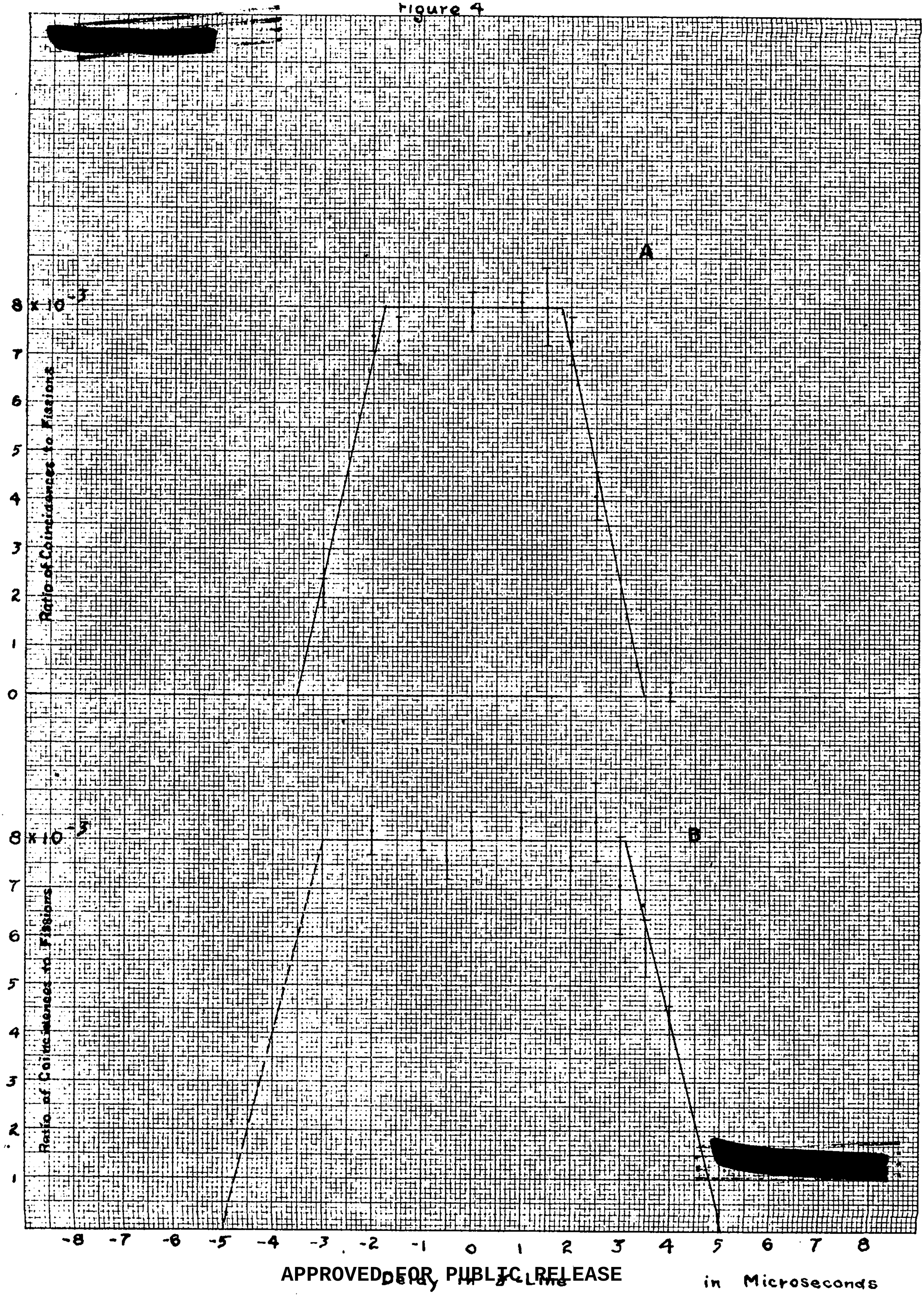

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Figure 5

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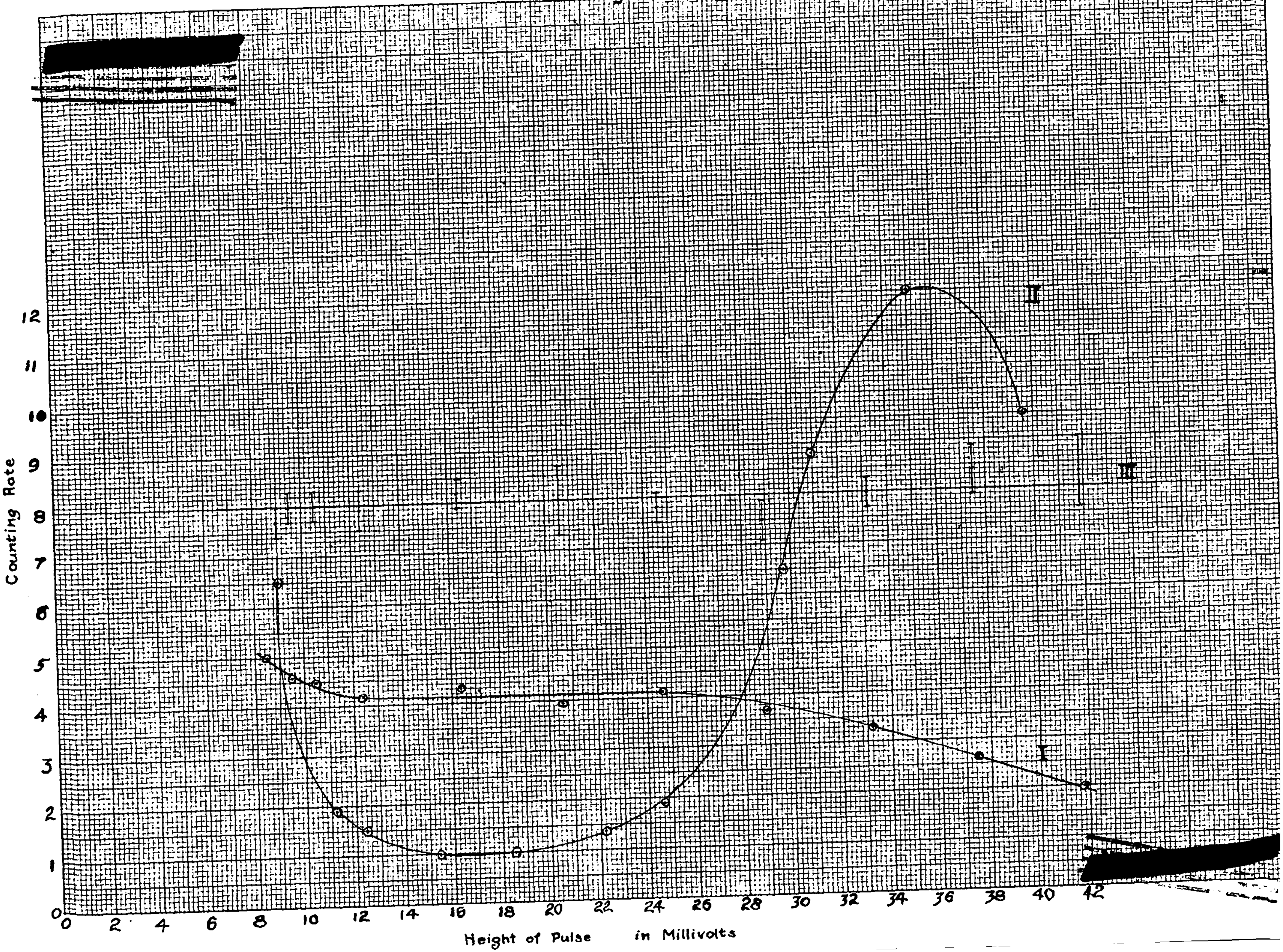

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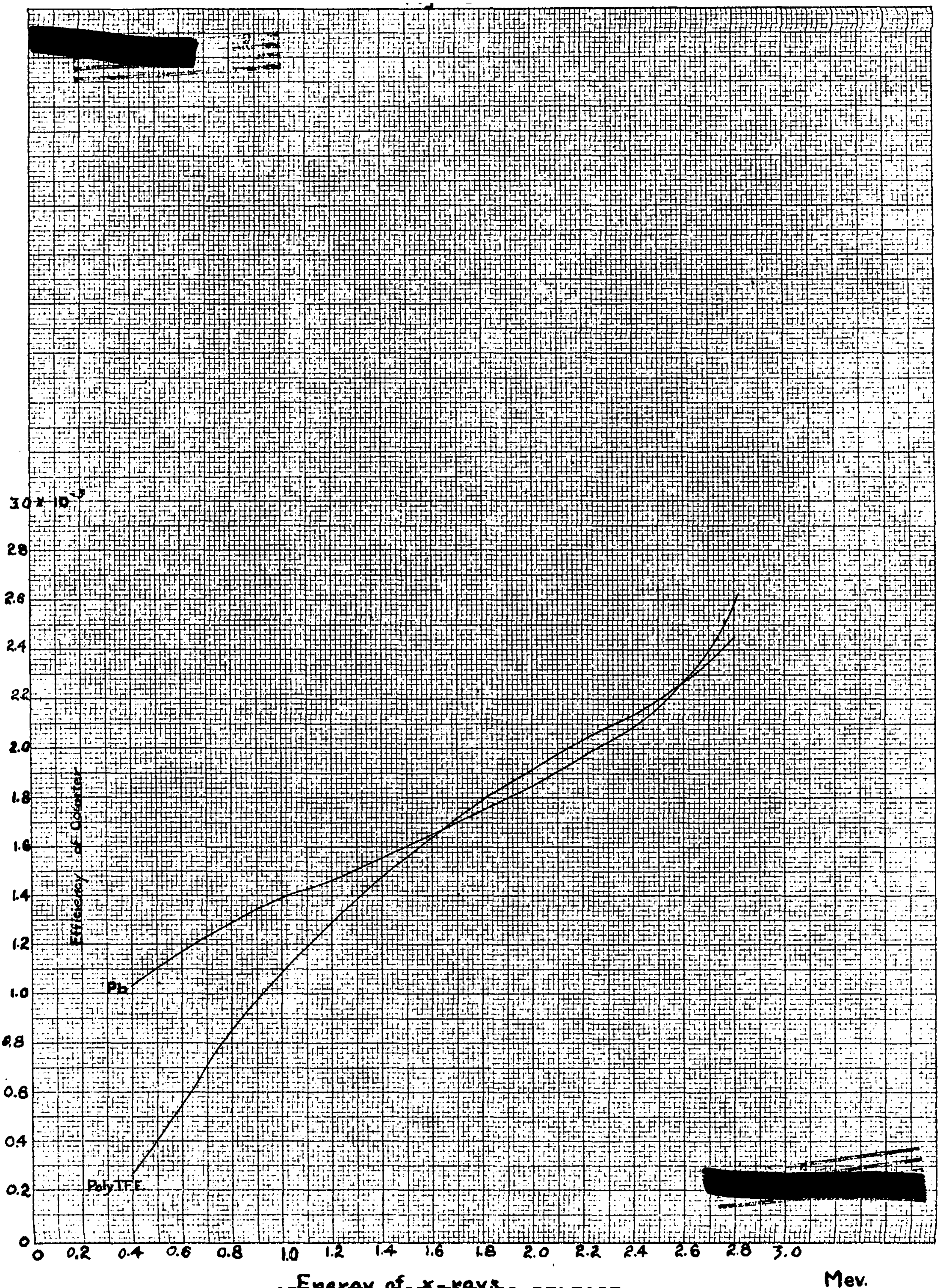

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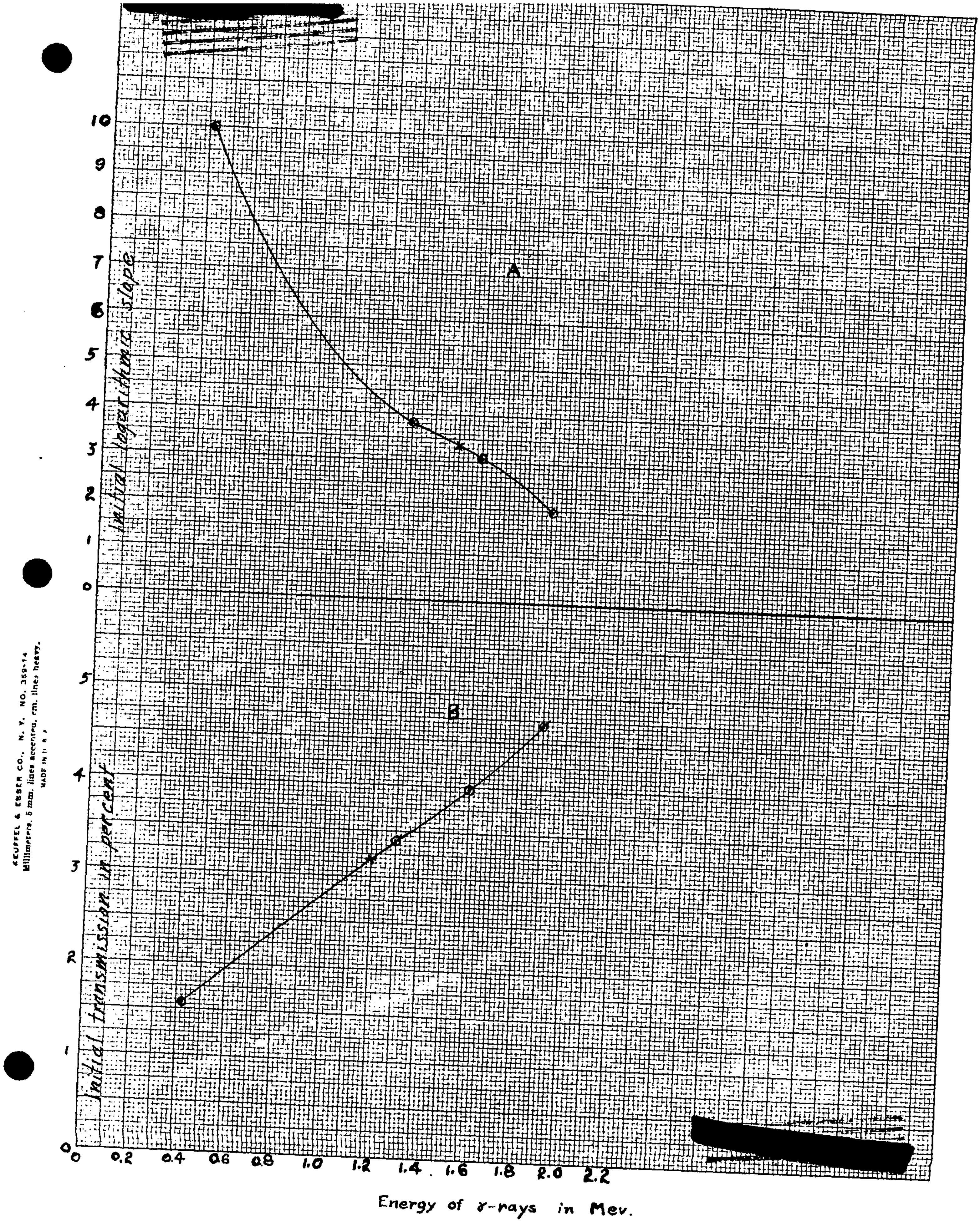
Initial logarithmic slope
10
9
8
7
6
5
4
3
2
1
0
A
B
5
4
3
2
1
0
Initial transmission in percent
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
2.2
Energy of γ-rays in Mev.
KEUFFEL & ESSER CO., N. Y. NO. 359-14
Millimeters, 5 mm. lines accented, cm. lines heavy.
MADE IN U.S.A.

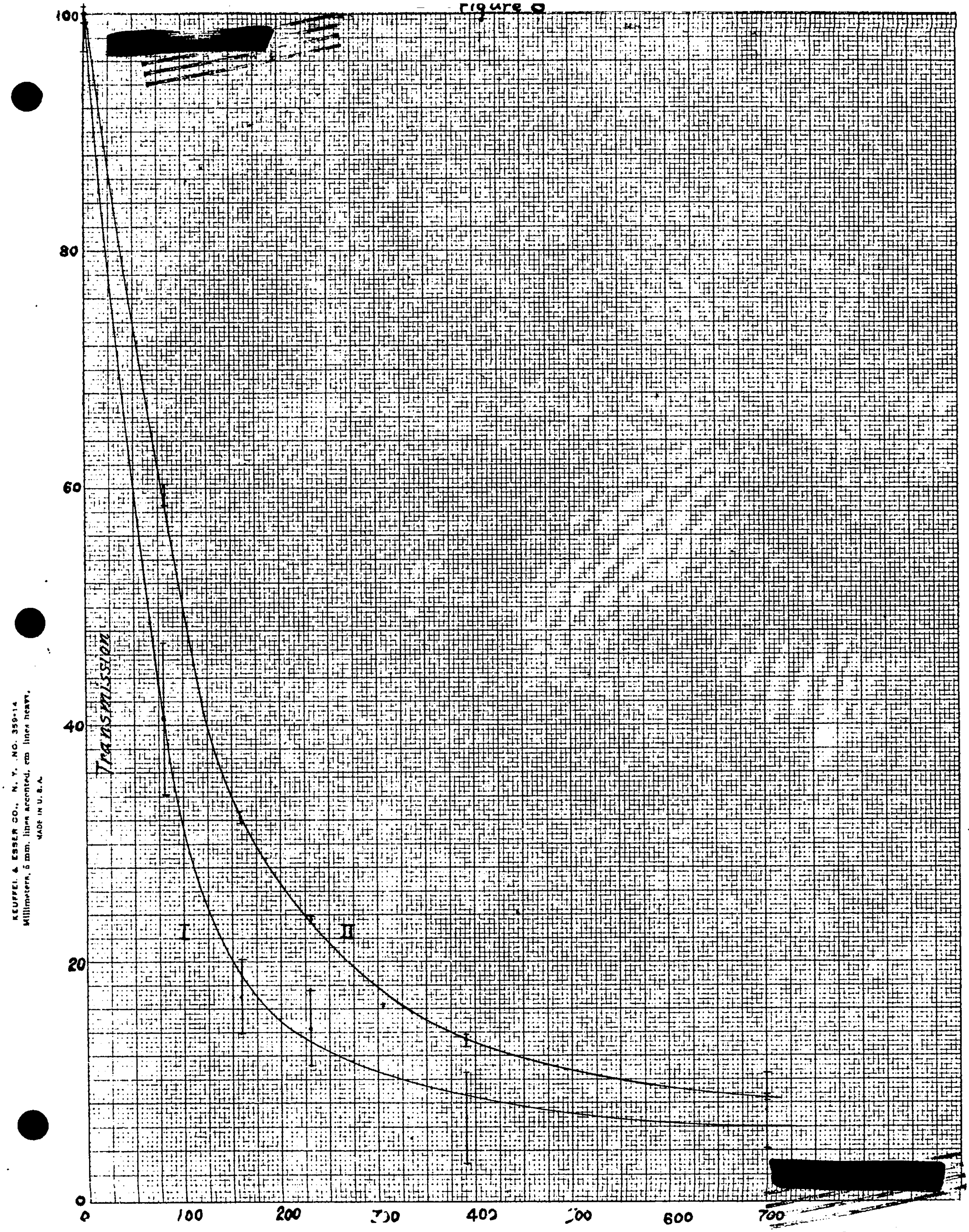
Figure 8
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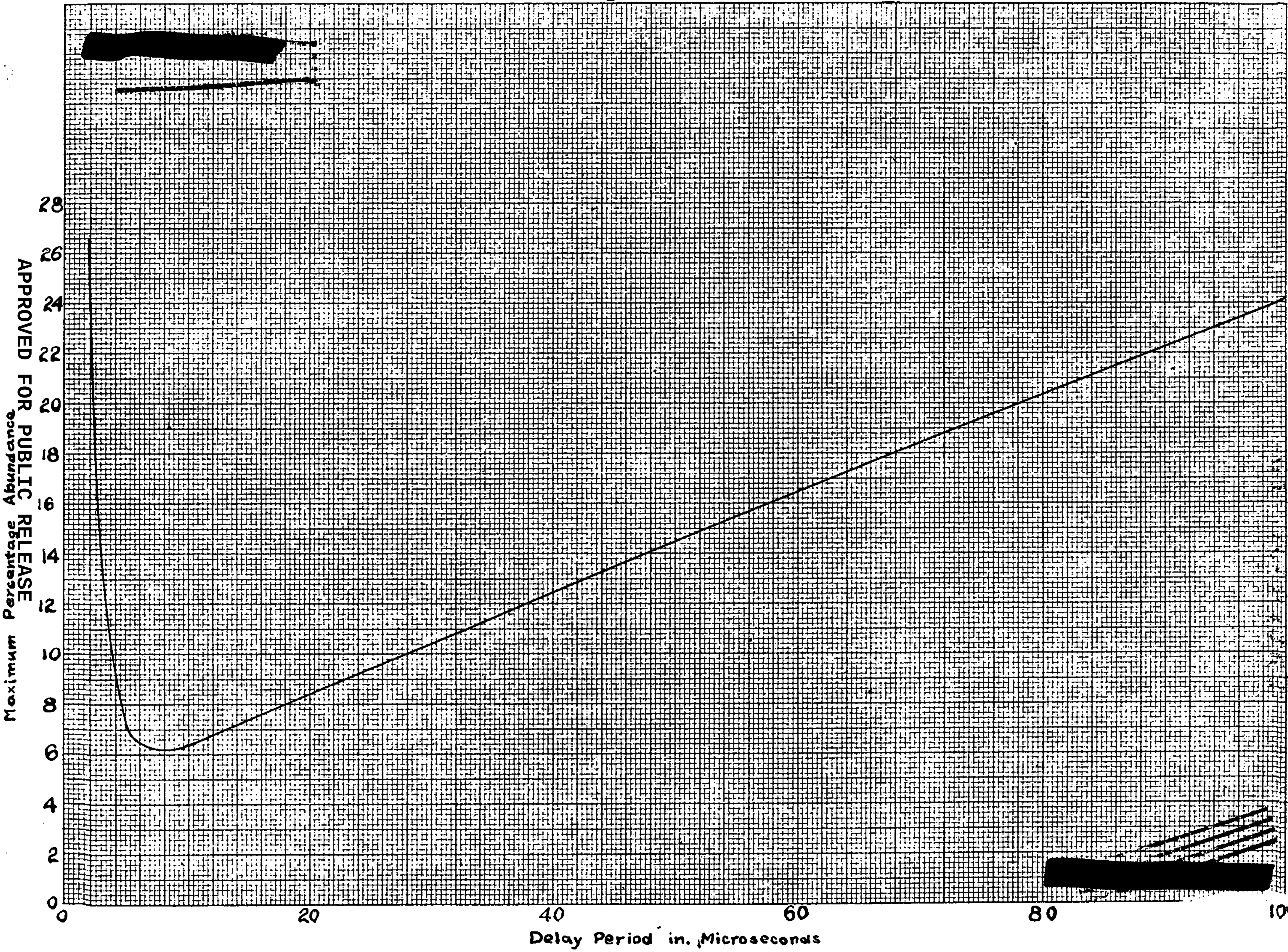
Transmission
100
80
60
40
20
0
0
100
200
400
600
700
I
II
[redacted]
mg/cm² Pb
KEUFFEL & ESSER CO., N. Y. NO. 359-14
Millimeters, 5 mm. lines accented, cm lines heavy.
MADE IN U. S. A.

Figure 9



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