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MEASUREMENT OF THE CAPTURE-FISSION RATIO IN 25 FOR THERMAL NEUTRONS

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-2-

ABSTRACT

New measurements of the ratio  $\alpha = \sigma_c / \sigma_f$  in  $^{25}\text{Mg}$ , where  $\sigma_c$  is the cross section for  $^{25}\text{Mg}$  formation and  $\sigma_f$  is the fission cross section, for thermal neutrons, are described. Three methods were used: (1) a comparison of the  $^{10}\text{B}(n, \alpha)$  disintegration rate of weighed boron films relative to the fission rate of weighed  $^{25}\text{Mg}$  films in the same thermal-neutron flux, with the known relative absorption cross sections; (2) a determination similar to (1) except that the  $^6\text{Li}(n, \alpha)$  reaction in weighed  $\text{LiF}$  films was used; (3) comparison of the disintegration rate in a  $\text{BF}_3$ -filled counter of known active volume with the fission rate in weighed  $^{25}\text{Mg}$  films in the same thermal flux. The three methods yield respectively these results: (1)  $\alpha = 0.18 \pm .03$ ; (2)  $\alpha = 0.154 \pm .02$ ; (3)  $\alpha = 0.155 \pm .035$ . The average is  $\alpha = 0.16 \pm .03$ .

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-3-

MEASUREMENT OF THE CAPTURE-FISSION RATIO IN 25 FOR THERMAL NEUTRONS

The total absorption cross section  $\sigma_{25}$  of 25 for thermal neutrons has been reported by Fermi<sup>1)</sup>. This cross section includes both capture to form 26 and fission; i.e.,  $\sigma_{25} = \sigma_c + \sigma_f$ , hence it is of interest to measure the ratio  $\alpha = \sigma_c / \sigma_f$ .

Fermi<sup>2)</sup> has determined  $\alpha$  for neutrons of thermal energy. He reports  $\alpha = 0.21$  (but see note 2).

We have made new measurements of  $\alpha$ , by three methods.

I. BORON - 25 FILM EXPERIMENT

Our first method was the following: thin films of boron and 25-enriched alloy, of known mass, deposited on metal foils, were placed in the same thermal neutron flux, in adjacent ionization chambers, and the ratio  $r$  of fission and  $B^{10}(n, \alpha)$  disintegration rates obtained. The ratio of these rates which would occur if all thermal neutron absorption by 25 were due to fission (i.e.,  $\alpha = 0$ ) is

$$r_0 = \frac{N_B \sigma_B}{N_{25} \sigma_{25}} = \frac{235 M_B \sigma_B}{10.82 M_{25} \sigma_{25}} \quad (1)$$

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- 1) Fermi, E., CP-1389.
  - 2) Fermi, E., CP-1531. His value of  $\alpha$  was later measured to be 0.18. It is to be emphasized that Fermi's observations are treated without corrections for the thickness of the 25 and Li foils used. The value of observed counting rates,  $r'$ , is thus subject to a correction which we estimate should increase the value of  $\alpha$  obtained by him.

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where  $N_B$  and  $N_{25}$  are the numbers of atoms present and the  $M$ 's are masses. The actual disintegration ratio is given by

$$r = \frac{N_B \sigma_B}{N_{25} \sigma_f} \quad (2)$$

so that

$$\frac{r}{r_0} = \frac{\sigma_{25}}{\sigma_f} = \alpha + 1. \quad (3)$$

Because the films are not really infinitely thin, the true value of  $r$  is not observed, but rather a value  $r'$ , which needs to be corrected for the thickness of the films. In our work these corrections were made; they are described at the end of this paper.

The thin weighed boron samples were prepared by decomposition of  $B_2H_6$  gas impinging on hot tungsten foils<sup>3)</sup>. The technique is very difficult, but three satisfactory films were produced. They were made as heavy as was consistent with thinness, in order to increase the percentage accuracy in weighing. To test for the presence of impurities, one film was analyzed spectroscopically; the principal foreign material expected was silicon, because of the technique, and this was not found in any appreciable quantity. Moreover, the  $B_2H_6$  gas used was 99.6 percent pure.

The 25 sample in most of the experiments used was a thin film electroplated on platinum, code number E50; its 25 content has been variously

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3) Russell, H., report in preparation.

measured<sup>4)</sup>. Chamberlain's result of 763  $\mu\text{gm}$ , obtained by comparison with samples of the E10 series, was adopted, partly because other E10 material was used by Fermi in the work on  $\sigma_{25}$  and  $\alpha$  quoted above. For one run an E10 sample was used which had been weighed by  $\alpha$ -counting and also by fission counting, in the course of this work, by comparison in a thermal flux with a sample of normal alloy. These two weighings agreed within 1 percent.

The ionization chambers, made of aluminum throughout, are drawn to scale in Fig. 1. The samples were mounted on either side of the common high-voltage electrode, and so were separated by only  $3/64$ " of Al. The gas used was spectroscopic argon at about 2 atmospheres, with a chamber depth of 7 mm.

In the course of these observations two neutron sources were used. First, well-thermalized neutrons were obtained in the carbon block in Building X, and a Cd ratio of over 1000 in the particular spot chosen. Second, it was found that with a sufficiently large paraffin block and primary neutron energy less than 100 kv from the  $\text{Li}(p,n)$  reaction, the observations of  $r'$  could be duplicated in Building W, with a Cd ratio of nearly 1000.

In obtaining  $r'$ , several precautions were observed. Runs with dummy samples established that the background was negligible. Care was taken to establish bias plateaus. This was easy in the 25 chamber; a representative bias curve is given in Fig. 2. It was achieved in the boron case by using an amplifier of low (120 kv) noise level and stable gain, preparing sufficiently thin samples and always measuring  $r'$  as a function of counter bias. A representative

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4) Hush, J. M., (unpublished); Chamberlain, O., (unpublished).

bias curve is given in Fig. 3.  $\text{Li}^7$  recoils as well as  $\alpha$ -particles are detected; in Fig. 3 the  $\alpha$ -particles are cut off at about bias 45, hence the  $\text{Li}$  recoils are counted out to about bias 23. Thus in this work, as well as in the later work, the probability of detecting a disintegration is almost unity. The value of  $r'$  is taken just above the noise level. In this way  $r'$  could be observed to 1 percent. Repeat runs were taken in some cases; the values of  $r'$  proved reproducible within 1 percent.

In calculating the value of  $\alpha$  from these observations we used  $\sigma_B = 703 \times 10^{-24} \text{ cm}^2$  and  $\sigma_{25} = 640 \times 10^{-24} \text{ cm}^2$ , values reported by Fermi (1,5).

The results are summarized in Table I.

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- 5) Fermi, E., CP-1098. In regard to  $\sigma_{25}$ , for the neutron energy distribution present in our experiments, see Section IV, page 15.

TABLE I

B film	M <sub>B</sub> μgm	25 film	M <sub>25</sub> μgm	r <sub>0</sub>	r <sub>c</sub>	Net correction factor <sup>6)</sup>	α ± 1
W2	221 ± 5	E5C	763 ± 14	6.92 ± .20	7.77 ± .07	1.018	1.143 ± .035
W2		E10 (I)	57.3 ± 1	92.0 ± 2.5	101 ± 1	1.024	1.124 ± .035
FW7	271 ± 5	E5C	763 ± 14	8.48 ± .22	9.83 ± .1	1.023	1.185 ± .035
FW8	169 ± 4	E5C	763 ± 14	5.29 ± .16	6.35 ± .07	1.010	1.213 ± .035

6) See pages 15 to 20.

The average (since the W2 trials are not independent) is  $\alpha = 0.18 \pm .03$ . The probable errors are those arising from uncertainties in the masses and observations on these foils and do not contain uncertainties in  $\sigma_B$  or  $\sigma_{25}$ .

Since the value of  $r'$  for any one foil could be closely reproduced, the most likely explanation for the spread in answers is error in the mass of boron; such errors are hard to avoid when dealing with such small weights.

## II. LITHIUM - 25 FILM EXPERIMENT

By the same method as in Section I, lithium was compared with 25, using the  $\text{Li}^6(n, \alpha)$  reaction. The same ionization chambers and source of slow neutrons were used; the same equations apply if one replaces  $M_B$  by  $M_{Li}$ ,  $\sigma_B$  by  $\sigma_{Li}$ , and the atomic weight factor 10.82 by 6.94.

The lithium fluoride films were prepared by evaporation from a steel furnace heated to about  $900^\circ \text{C}$ . Before the  $\text{LiF}$  was deposited the furnace was cleaned and blanks were run. A blank of three micrograms in five minutes was achieved before the  $\text{LiF}$  was put in the furnace. Since the deposition time was only about fifteen seconds, the blank correction was negligible. The  $\text{LiF}$  used was taken from an optical blank supplied by the Harshaw Chemical Company. A spectroscopic analysis on the starting material showed traces of less than .001 percent of Si, Mg, B, Fe, Ag, Na, and Ca, 0.001 - 0.01 percent of Cu and Pb and 0.01 - 0.1 percent of Al. No other metallic impurities were found. An optical examination of the infra-red transmission of a similar crystalline  $\text{LiF}$  has revealed only a very small amount of  $\text{LiHF}_2$  as an impurity<sup>7)</sup>. One of the finished

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7) Wright, N., Review of Scientific Instruments, 15, 22 (1944).



LiF films was also spectroscopically analysed. The major impurities were 0.02 percent of boron, 0.01 percent of silicon and 0.02 percent of silver; the only other metal found was a trace of Ca. The analyses are accurate to about 50 percent. Apparently the elements with more volatile fluorides were concentrated by the evaporation technique.

The impurities are given as the weight percent of metal based on the weight of LiF; if we convert to the fluoride, we obtain 0.13 percent of  $\text{BF}_3$ , .037 percent  $\text{SiF}_4$ , and 0.024 percent  $\text{AgF}$ . The LiF is then 99.8 percent pure. For nuclear reactions, the important ratio is that of boron atoms to lithium atoms. This ratio is 0.00048:1. Since the boron cross section for slow neutrons is about eleven times that of Li the ratio of boron counts to lithium counts is 0.0053:1. The correction therefore in the observed counting rate per unit weight of lithium is + 0.2 percent in the weight and -0.5 percent for boron impurity or a net correction of -0.3 percent. This correction will probably vary somewhat from film to film since the first films made from a fresh charge of LiF will probably have a higher percentage of boron than succeeding films. For this reason and because this error is considerably smaller than some of the other experimental errors in the measurements, a correction does not seem desirable. We can say with considerable confidence that the error due to impurities in the lithium fluoride does not exceed 0.5 percent.

Again ESC was the principal 25 sample. Three runs were taken in which lithium films were measured both against ESC and against an E10 film of 25. How these compare may be seen in Table II.

It was easier to measure  $r^1$  than for boron, because of the greater particle energy. To stop the  $\text{H}^3$  recoils, the ionization chamber was deepened to

-10-  


13 mm and the pressure of spectroscopic argon to about 4 atmospheres.  $r'$  was again measured as a function of bias. A sample bias curve is shown in Fig. 4;  $r'$  is taken just above the noise level. The 25 chamber had the same bias curve as before. Thickness corrections were calculated as in the boron case.

The value of  $\sigma_{Li}$  was taken at  $64.9 \times 10^{-24} \text{ cm}^2$ ; this is the latest result reported<sup>8)</sup> from Chicago. The lithium results are given in Table II.

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8) CP-1531.



TABLE II

Li Film	M <sub>Li</sub> μgm	25 films	M <sub>25</sub> μgm	r <sub>0</sub>	r'	Net correction factor <sup>6)</sup>	1 + α
Gpt1	146.1 ± 1%	E5C	763 ± 14	.658 ± .014	.770 ± .007	1.006	1.179 ± .027
Gpt2	106.2 ± 1%	E5C		.478 ± .010	.552 ± .006	1.002	1.159 ± .027
Gpt2		E10H15	67.5 ± 1.4	5.40 ± .12	6.04 ± .05	1.003	1.122 ± .027
Gpt2		E10H12	74.3 ± 1.4	4.90 ± .10	5.60 ± .06	1.003	1.145 ± .026
Gpt4	110.0 ± 1%	E5C		.495 ± .010	.557 ± .006	1.002	1.129 ± .027
Gpt4		E10H12		5.08 ± .11	5.63 ± .07	1.003	1.113 ± .028
Gpt5	16.3 ± 7%	E5C		.0735 ± .0053	.0936 ± .003	.993	1.27 ± .1
Gpt6	141.5 ± 1%	E5C		.637 ± .013	.738 ± .007	1.006	1.165 ± .028
Gpt7	52.7 ± 2%	E5C		.237 ± .006	.267 ± .005	.997	1.122 ± .035

6) See pages 15 to 20.

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Combining the GPt2 and GPt4 results with the different 25 films, and weighing the answers linearly according to their accuracies, one obtains  $\alpha = 0.154 \pm .02$ . The errors stated do not include any probable error in  $\sigma_{Li}$ .

We have enough lithium films in these experiments so that the errors in weighing should average out fairly well; the principal remaining source of uncertainty seems to be the mass of 25.

### III. BF<sub>3</sub> - 25 COMPARISON EXPERIMENT

In order to make a determination of the quantity  $\alpha$ , which was independent of the mass determination of the boron or lithium-fluoride films, the counting rate of an ionization chamber containing BF<sub>3</sub> gas was compared with that of an almost identical ionization chamber containing two foils covered with enriched 25 when the two chambers were in a uniform flux of thermal neutrons.

The cross section of the BF<sub>3</sub> chamber is shown in Fig. 5. The ionization chamber for the 25-foils is of the same construction except that the ends of the hollow high voltage electrode are not spherical so that the circular foils diameter 1-1/4" would lie flat on the inner wall. As noted on the diagram, the chambers were constructed of dural to reduce the absorption of the slow neutrons, and the gaskets which seal the chambers are of lead to eliminate any material which would absorb or contaminate the BF<sub>3</sub> gas. The high voltage insulators are of porcelain and the collector electrode insulators are of glass.

The neutrons were provided by the short tank in W. Their original energy was 100 kv or less, and they were thermalized by a block of paraffin 18"x18"x20" with a 6"x6"x14" hole in it for the chambers. This gave a layer of paraffin six inches thick on all sides of the chambers except in the direction away from the neutron source. The axes of the chambers were parallel and two

and a half inches apart. They and the preamplifier were suspended so their positions could be interchanged periodically during the run to compensate for any non-uniformity in the neutron flux in the hole in the paraffin block. When interchanged in this way the ratio of counting rates changed 6 percent or less.

The effective counting volume of the  $\text{BF}_3$  chamber was determined by filling the high voltage electrode with water. The volume of water was measured from a burette and this was checked by weighing the electrode before and after filling. As a check this was done separately by two persons. From this volume was subtracted the volume of the collector electrode, and a small correction was made for the region where the collector electrode enters the high-voltage electrode. The effective volume was considered to be  $15.96 \pm 0.04$  cc.

The  $\text{BF}_3$  gas was purified and put in the chamber by Russell. He measured the pressure to be  $10.12 \pm .05$  cm Hg at a temperature of  $25^\circ \text{C}$ . To shorten the range of the disintegration particles and thereby make the effective counting volume better defined, pure argon was added to make the total pressure two pounds above atmospheric.

The high voltage electrode was held at a positive potential of 2200 volts.

During the preliminary testing of the chamber it was filled and emptied several times. It was found that sufficient boron remained on the walls of the chamber to give a residual counting rate which was 4 percent of the rate when it contained  $\text{BF}_3$  at 10 cm pressure. After several cycles of filling and emptying this background did not increase further. It was compared with the normal rate by using a Ra-Be source in a standard paraffin geometry and was found not to change appreciably over a period of two weeks.

When a curve of counting rate vs the bias of the scaler discriminator is plotted (Fig. 6) it is found that the curve is not exactly flat in the region just above noise level, but has a slope such that the extrapolated counting rate of zero bias is 4 percent higher than the rate for a bias of 20 units. For a maximum signal of 100 units the curve is straight from a bias of 12 units to a bias of 35 units, so this 4 percent extrapolation can be easily determined. The bias curve probably slopes in this way because of a somewhat non-uniform collecting field in the counting region, particularly where the collector electrode enters the high voltage electrode. This bias curve was measured several times during the final runs by means of a second scaler connected in parallel to the one set constantly at a bias of 20 units.

The ionization chamber containing the 25 foils was filled with 55 pounds per square inch gauge pressure of pure argon in order to make the range of the fission fragments small as compared to the curvature of the electrode walls. If this had not been done a considerable fraction of the fission fragments would have been stopped by the walls before giving up a sufficient fraction of their energy. No correction for this was necessary as a bias curve for this chamber had an appreciable flat region. To secure satisfactory collection the high voltage electrode was held at 4000 volts positive.

The 25 was on two foils, E10H7 and E10H8, with a total mass of 25 of 219.6  $\pm$  5.5 micrograms.

The final data was taken in six runs, between each pair of which the positions of the chambers were reversed as explained above. The second pair of runs was taken with a counting rate which was about one-third the others to show that the ratio of counting rates was independent of the number of counts per

second. Each run had 1600 to 2000 fission pulses and the  $\text{BF}_3$  rate was over one hundred times this, so the probable error on each run was 2-1/2 percent or less. The three runs in one position gave ratios of 116.5, 116.9, 113.7 (average 115.7), while the three runs in the other position gave 119.2, 120.8, 122.0 (average 120.7). The overall average is then  $118.2 \pm 1.6$ .

The computed ratio of counting rates, obtained by use of  $\sigma_B = 703$  and  $\sigma_{25} = 640$  barns with the measured pressure and effective volume, is  $102.4 \pm 2.75$ . The measured ratio is 15.5 percent higher than this. On this basis the value of  $\alpha$  would be  $0.155 \pm .035$ .

#### IV. MOST PROBABLE VALUE OF $\alpha$

The three determinations averaged linearly yield  $\alpha = 0.16$ , and a reasonable limitation on the error is  $\pm .03$ .

As previously stated, the probable errors in  $\sigma_B$ ,  $\sigma_{25}$ , and  $\sigma_{Li}$  are not taken into account in calculating the error in  $\alpha$ .

The values of these cross sections used in these calculations are the values at 2200 meters/sec neutron velocity. Now the neutrons used in the present work were distributed in energy, so that, if the cross sections do not obey the same law as a function of energy, the effective cross sections will differ relative to one another from the values used in the calculations.  $\sigma_B$  and  $\sigma_{Li}$  may be taken as  $1/v$  in this region, but data of McDaniel et al.<sup>9)</sup>, involving fission and absorption methods, indicate that  $\sigma_{25}$  does not follow that law exactly.

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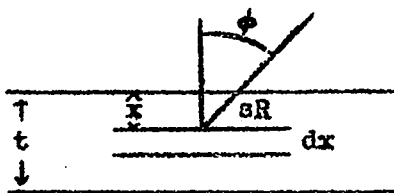
9) Anderson, Lavatelli, McDaniel and Sutton, LA-91.

Assuming a Maxwellian distribution of neutron velocities which is certainly not true for the neutrons degraded by paraffin, and using the McDaniel fission data for  $\sigma_{25}$  as a function of energy, one obtains an effective  $\sigma_{25}$  about 2.5 percent lower than that which we have used. This correction to  $\sigma_{25}$  would lower the value of  $(1 + c)$  by the same ratio.

However, further experiments of McDaniel on 25 in this region, using absorption detection, tend to show a nearer approach to the  $1/v$  law than is indicated by the experiments using fission. It is therefore felt that it is somewhat dubious whether this correction to  $c$  should be applied. It may suffice at this time to point out that if sufficiently accurate data on the behavior of  $\sigma_{25}$  is obtained, the revision in the value of  $c$  may be made.

#### V. THICKNESS CORRECTION

The thickness corrections are calculated by the method described by Hull<sup>10)</sup>. In a film of thickness  $t$ , imagine a slab of thickness  $dx$ , whose distance below the surface is  $x$ . Let  $s$  be the fraction of the range  $R$  of a



particle in the substance which it may spend in the slab and still be detected. Let  $\phi$  be the angle at which the particle may be emitted and still travel only the distance  $sR$  in the film.

Then

$$y = \text{no. detected from } dx = \frac{1}{4\pi} (\text{solid angle defined by } \phi) \times (\text{no. emitted from } dx)$$

$$= (1/2)(1 - x/sR) \times (\text{no. emitted from } dx).$$

10) Report A-1235.



Let  $n$  be the number emitted per  $\mu\text{gm}$ . Then the number emitted from  $dx$  is  $n\rho A dx$ , where  $A$  is the area of the film and  $\rho$  is its density. So

$$y = (1/2)(1 - x/sR) \cdot n\rho A dx,$$

and integrating

$$\begin{aligned} \text{total no. detected} = Y &= (1/2)n\rho A \int_0^t (1 - x/sR) dx \\ &= (1/2)n\rho At(1 - t/2sR). \end{aligned}$$

Then the fraction detected is

$$\frac{\text{No. detected}}{\text{No. emitted}} = \frac{Y}{n\rho At} = (1/2)(1 - t/2sR).$$

If we include both emitted particle and recoil nucleus from a reaction, the total fraction detected is

$$F = 1 - (t/4)(1/s_1R_1 + 1/s_2R_2).$$

The correction factor by which  $r'$  must be multiplied to give  $r$  is then  $1/F$ .

The factors entering the above equation are given in Table III.

The quantity  $s$  is somewhat indefinite, since some particles with energy less than 120 kv will be detected and some with a little more energy will be missed.

TABLE III

		s	Range cm	Area cm <sup>2</sup>	Density gm/cm <sup>3</sup>
Boron case	$\alpha$ 's	.93	$4.9 \times 10^{-4}$	5.1	2.3
	Li's	.90	$2.8 \times 10^{-4}$		
Lithium case	$\alpha$ 's	.93	$.63 \times 10^{-3}$	8.0	2.5
	H <sup>3</sup> 's	.93	$3.1 \times 10^{-3}$		
Fission case	E5C	.7	$1.4 \times 10^{-3}$	7.2	7.2
	E10	.7	$1.4 \times 10^{-3}$	8.0	7.2

In the boron case the ranges were estimated with the aid of Weisskopf. The ranges in air are known<sup>11)</sup>; extrapolating from nearby elements<sup>12)</sup>, one estimates that the atomic stopping power of boron relative to air is 0.77. For lithium, the ranges in air are also known<sup>13)</sup>, and the atomic stopping power can be estimated from theory<sup>14)</sup> as 0.86 times air. For the fission case, the ranges of the fragments may be roughly estimated from the data of Sogre and Wiegand<sup>15)</sup> on the stopping of fission fragments in  $U_3O_8$ .

The most reasonable assumption concerning the density is that the films are deposited at the nominal density. But since the thickness and range are affected in about the same way by changes in density, the exact value is not very important. One may point out that errors of say 10 percent in  $t$ ,  $s$ , and  $R$  do not have appreciable effects on  $F$ .

The calculated correction factors are given in Table IV.


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- 11) Rotblat, Nature, 158, 202 (1936). He has verbally corrected the Li range given there from 0.38 cm to .36 cm.
  - 12) Livingston and Bethe, Rev. Mod. Phys., 9, 245 (1937).
  - 13) Livingston and Hoffman, Phys. Rev., 50, 401 (1936).
  - 14) Rasetti, Elements of Nuclear Physics, pg. 62.
  - 15) LA-64.
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TABLE IV

Film	Correction factor
Boron-W2	1.030
Boron-FW7	1.035
Boron-FW8	1.023
LiF GPt1	1.015
2	1.011
4	1.011
5	1.001
6	1.015
7	1.005
25-E5C	1.011
E10(I)	1.006
E10H15	1.007
E10H12	1.007

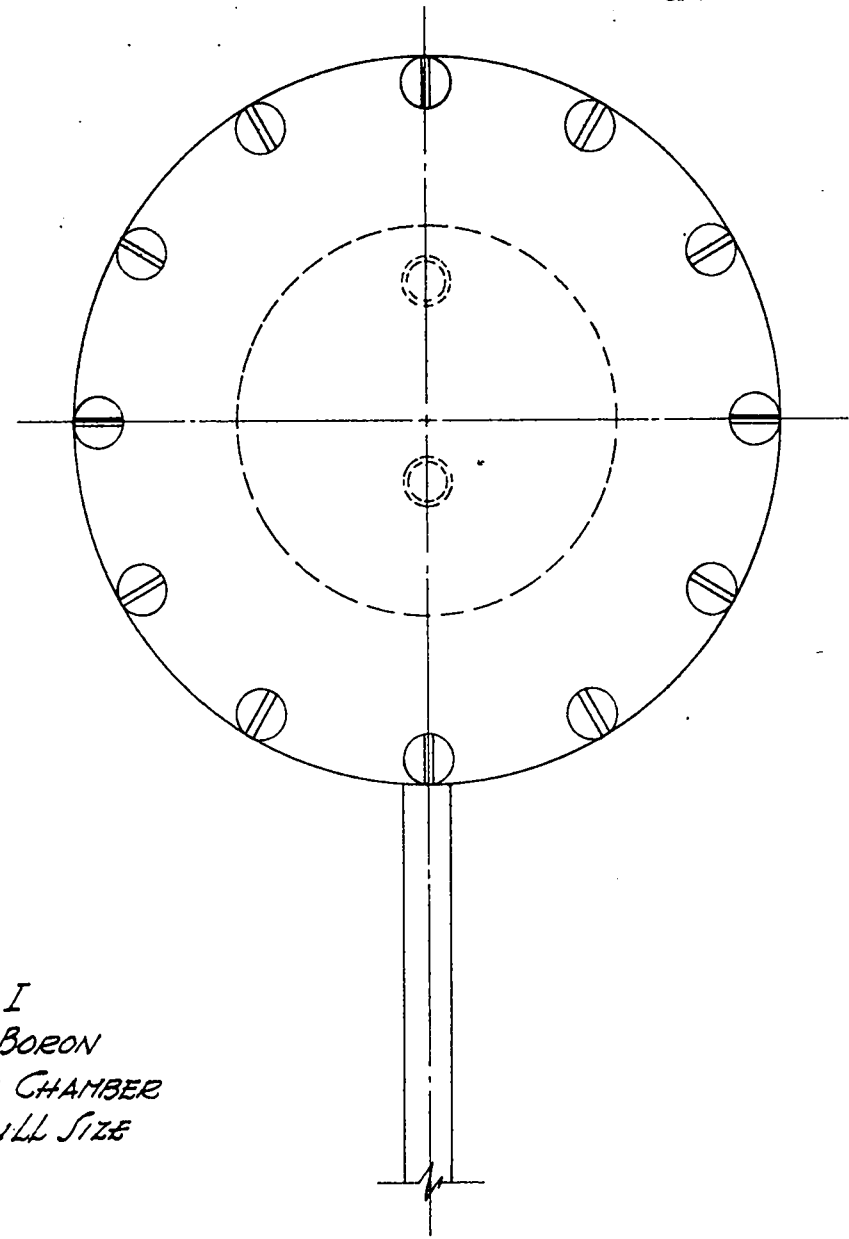
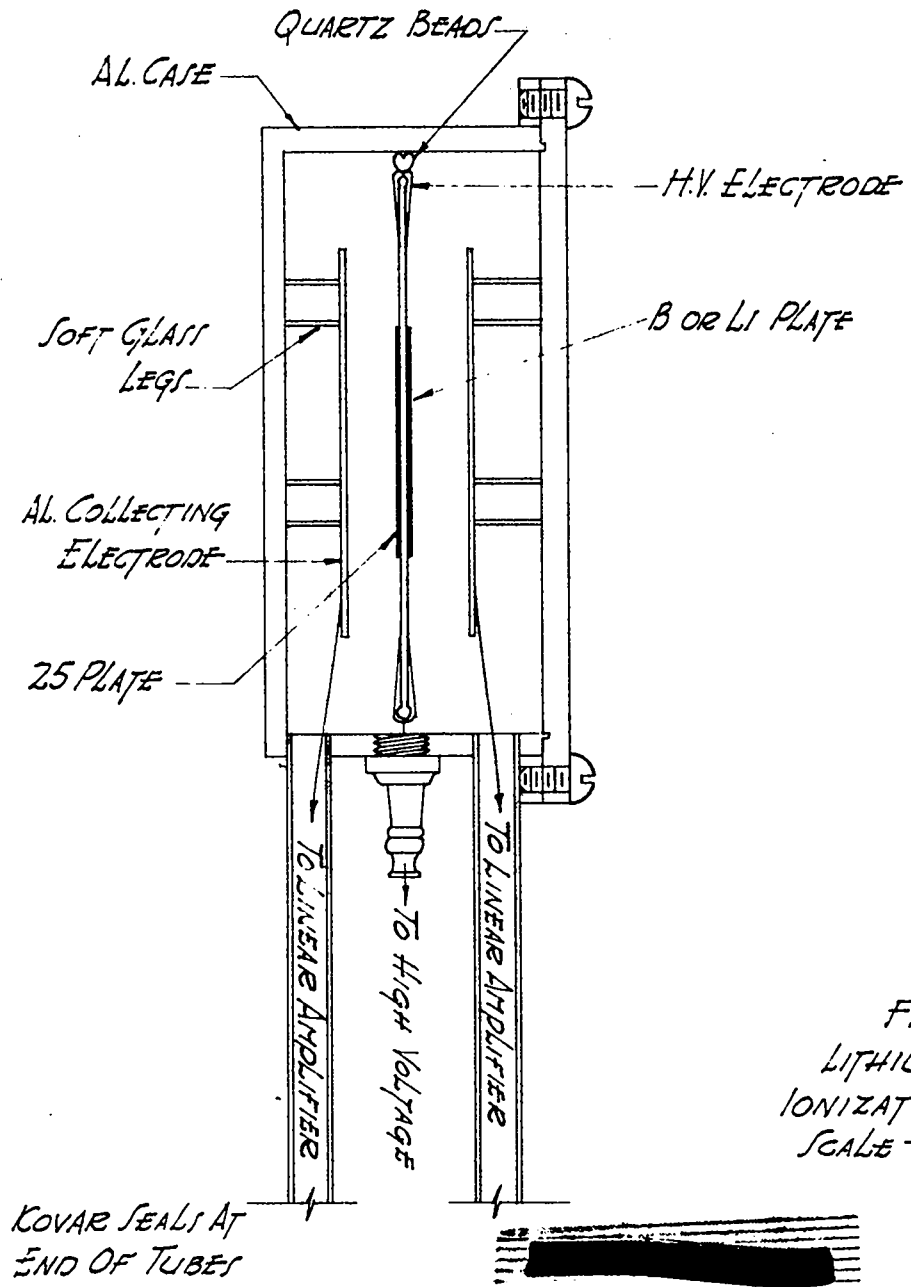
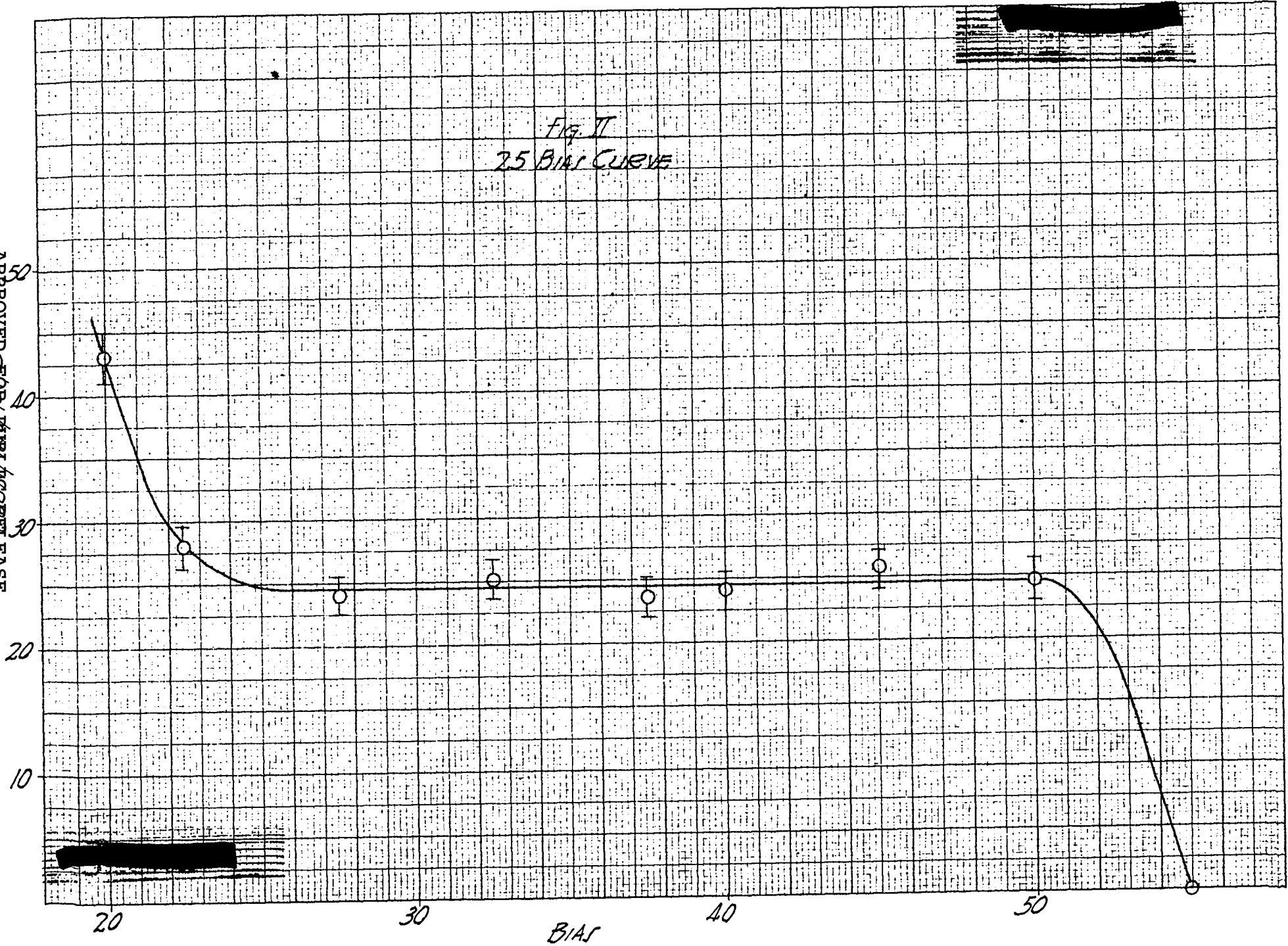


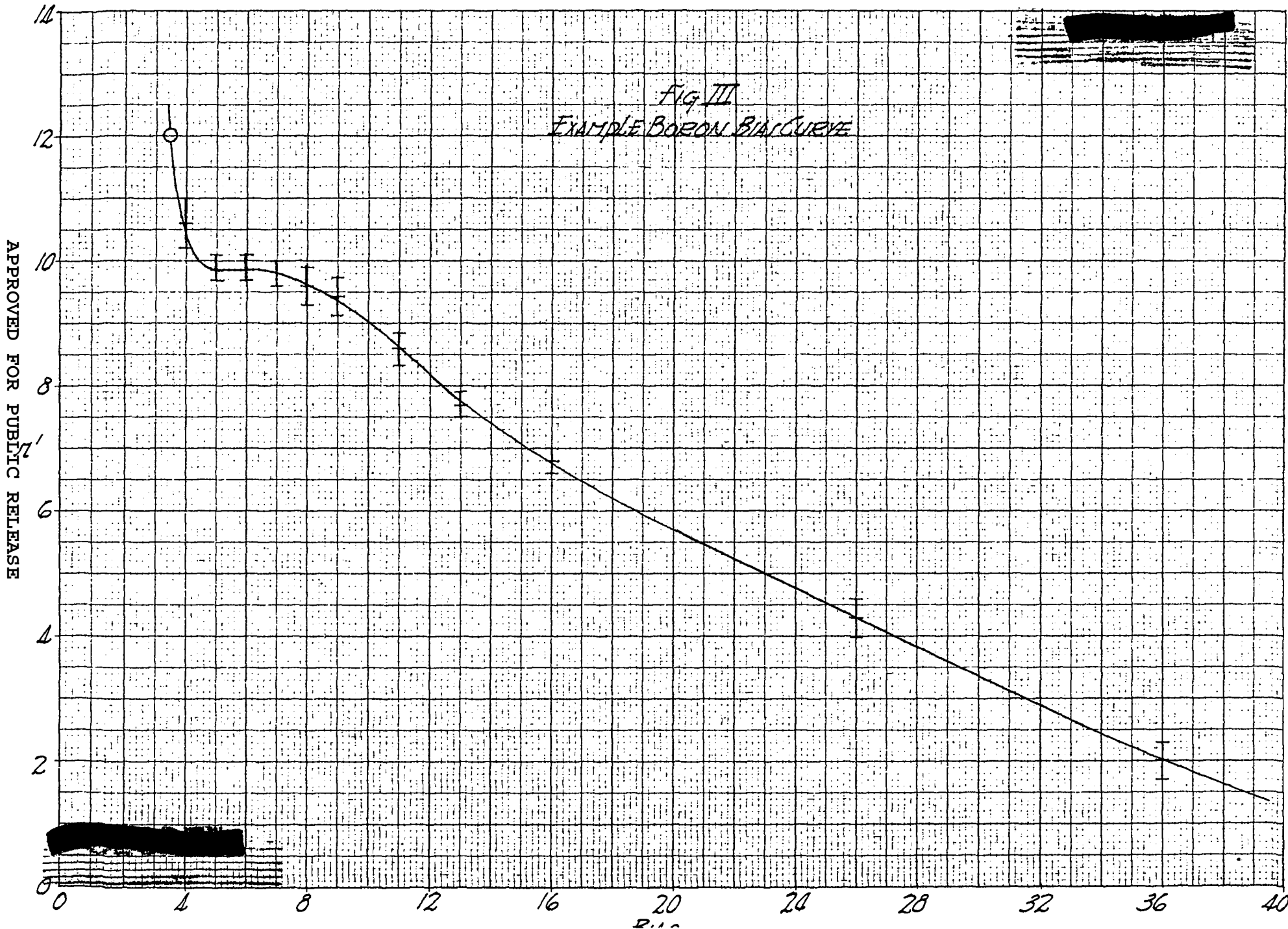
FIG. I  
LITHIUM-BORON  
IONIZATION CHAMBER  
SCALE - FULL SIZE

Fig. II  
25 Bias Curve

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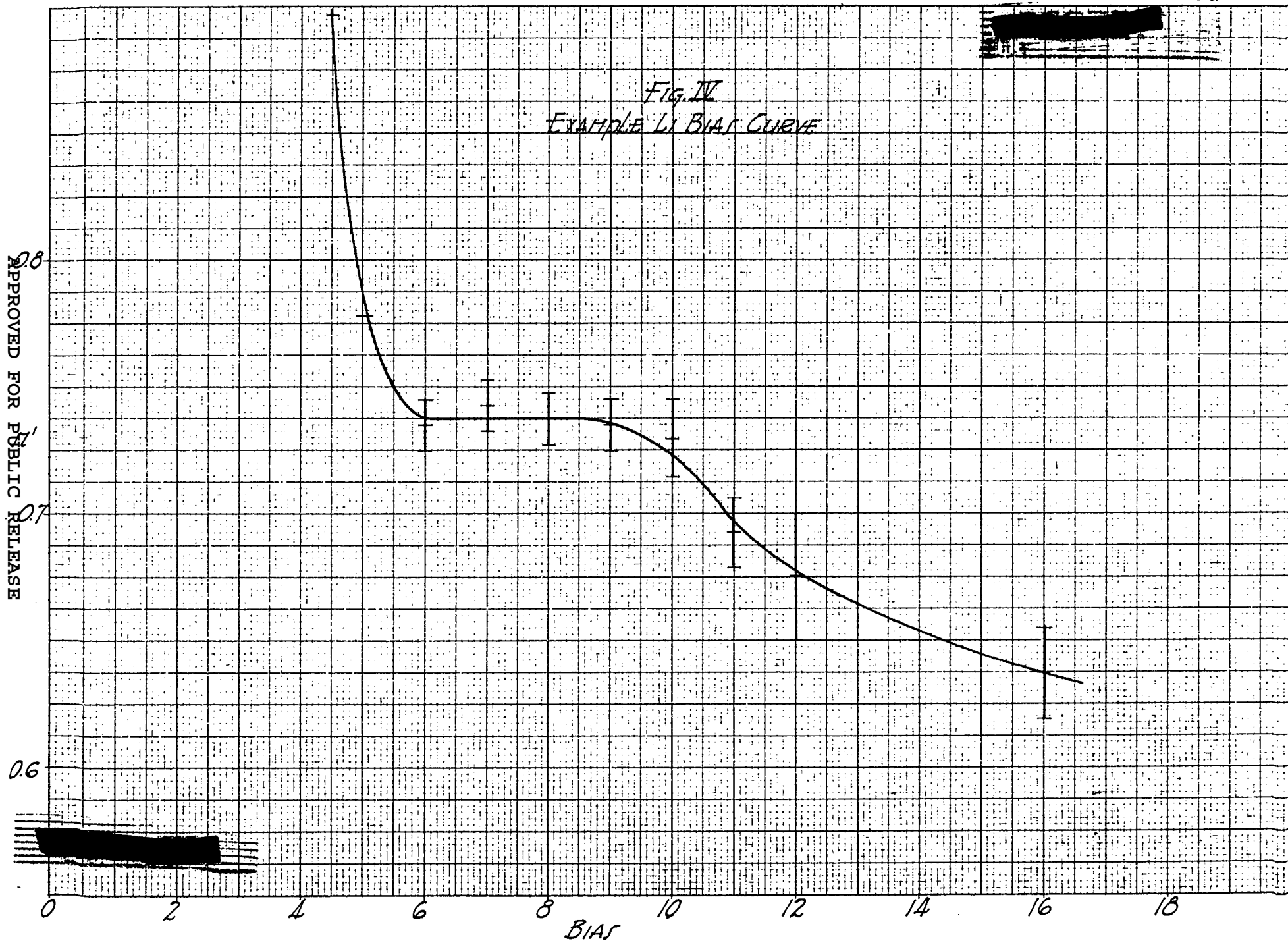
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FIG. IV  
EXAMPLE LI BIAS CURVE

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0.6

0.8

0.7

0

2

4

6

8

10

12

14

16

18

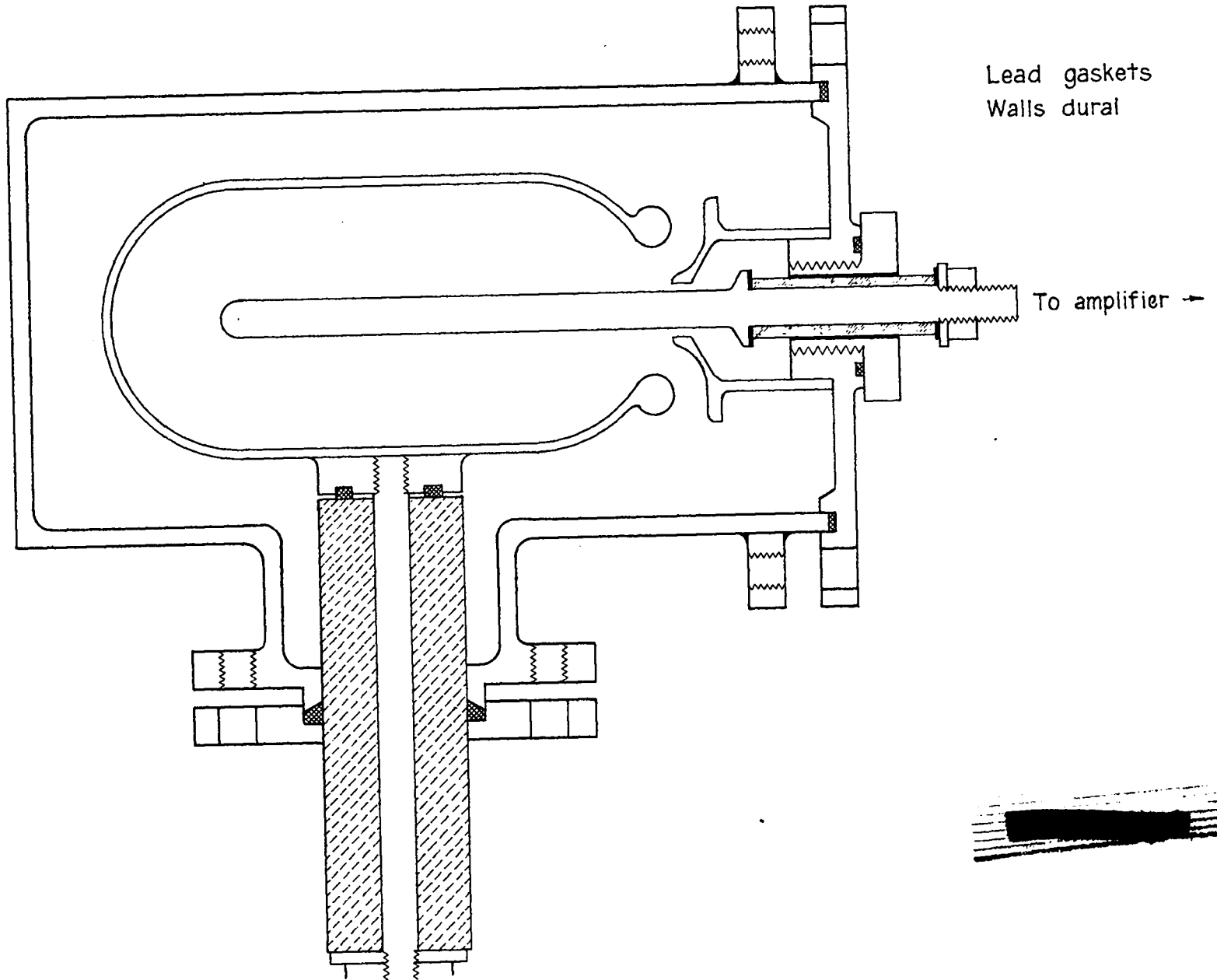
BIAS



BF<sub>3</sub> CHAMBER

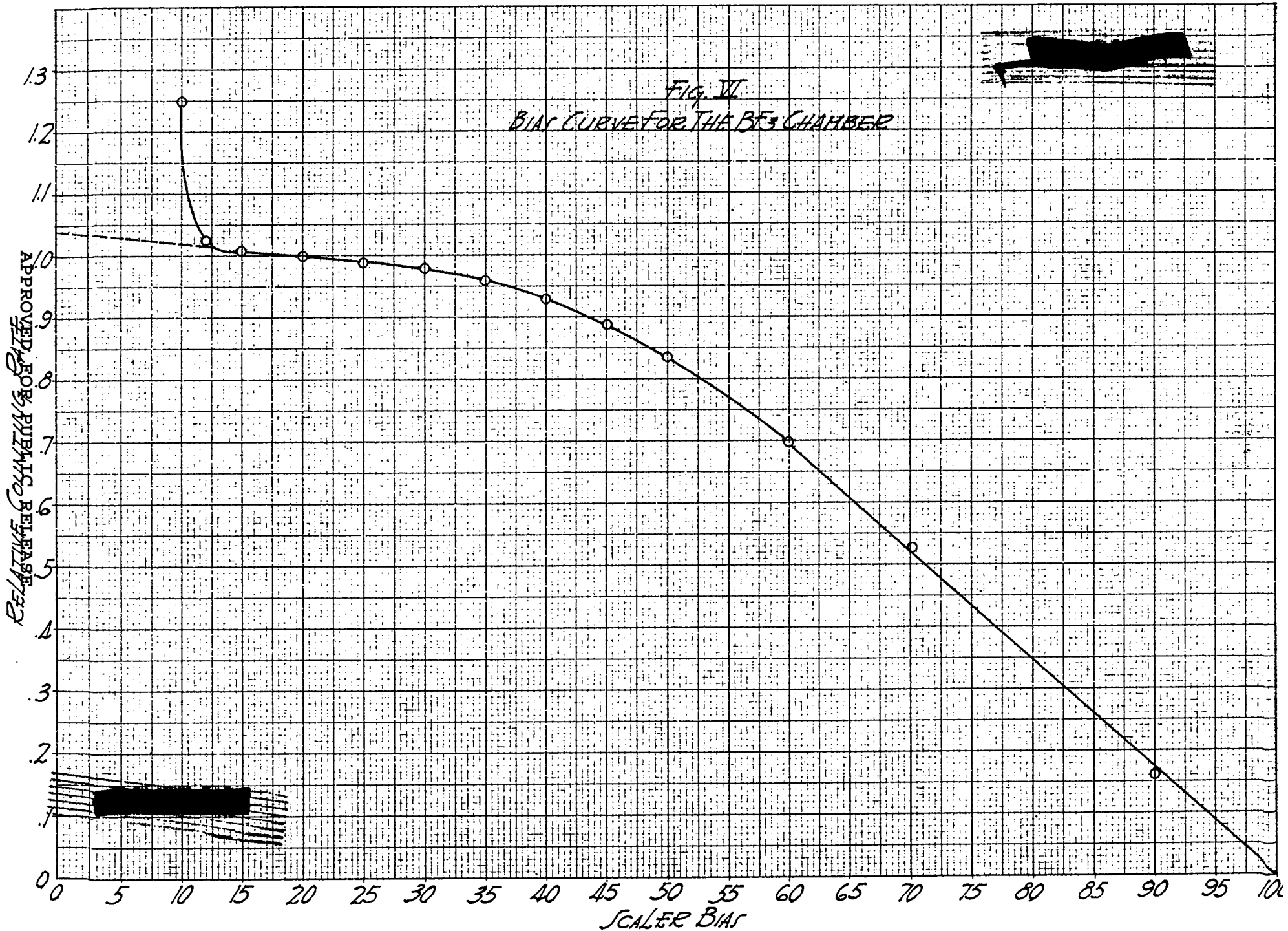
Twice full size

Fig. V



Lead gaskets  
Walls dural

To amplifier →



UNCLASSIFIED

EQUIPMENT ROOM

REC. FROM       

DATE JUN 14 1944

REC. NO. REC.       

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