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SPONTANEOUS FISSION IN SEPARATED URANIUM ISOTOPES

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ABSTRACT

The spontaneous fission rate of 28 is 24.0 ± 1.1 f/gm hr. The spontaneous fission rates of 25 and 24 are - 2.2 ± 4.6 and <60 f/gm hr respectively.

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SPONTANEOUS FISSION IN SEPARATED URANIUM ISOTOPES

Introduction

Spontaneous fission was discovered by Flerov and Petrzhak[1)] in common uranium. Subsequently several investigations on the subject have been made, always on unseparated isotopes. Kennedy and Segre[2)] have investigated for the first time separated 25 and concluded that its spontaneous fission rate was less than 10 times that of 28. Blanchard, Gofman and Seaborg[3)] investigated 24 and concluded that its spontaneous fission rate was less than 400 f/gm hr. We have continued and extended these investigations in the last year.

Experimental

The experiments consisted of putting certain amounts of normal uranium or of uranium whose isotopic composition had been changed in a known fashion in ionization chambers connected to linear amplifiers, and counting the fission pulses.

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1) G. N. Flerov and K. A. Petrzhak, Journal of Physics USSR, 3, 275 (1940)

2) CC-50

3) CF-220; CF-907

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The samples were thin layers (0.25 mg/cm^2) of U_3O_8 deposited electrolytically on platinum discs 0.05 mm thick and 4.3 cm in diameter. The discs were spot welded to copper discs of 12.7 cm diameter. A series of samples was prepared out of ordinary uranium (specific alpha activity 1500 α/mg min); another series of samples was of uranium of isotopic composition U^{238}: U^{235}: U^{234} = 2.76: 1.00: 0.0044 in mass (specific activity 16.44 x 10^3 α/mg min). The material of the enriched samples was analyzed by the Berkeley method[4]. All samples were weighed and alpha counted to determine their mass and check on their thinness. They were visually inspected for uniformity of thickness. The correction for self absorption of the fission fragments[5] is about 4%. In each ionization chamber we introduced approximately 49 mg of U_3O_8. Each ionization chamber has 3 electrodes, consisting of copper discs, 14 cm in diameter, spaced by 1 cm. The middle one is connected to the grid of the first stage of the amplifier. The other two, which carry the samples are at approximately - 500 volts. The chamber is filled with tank nitrogen, in which it is easy to collect electrons[6]. The amplifiers (constructed by Wiegand) are entirely battery operated and have a rise time of approximately 3 microseconds. Their output was connected to an ordinary mechanical counter and also to a pulse lengthener and amplifier which operated a recording milliammeter (Esterline, Angus). With this apparatus we could check that the pulse distribution in time followed Poisson's law.

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4) Kennedy, Segre, UCRL-9

5) Segre, Wiegand, LA-64

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6) The analysis of this water pumped nitrogen given by the manufacturer is: 99.3% N_2; 0.3% O_2; 0.4% water; trace hydrocarbon.

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In this experiment it is very important to be sure that the alpha activity of the samples does not give rise by its fluctuations to spurious counts, which would be mistaken for fission pulses. In order to make certain that no spurious counts due to these fluctuations were recorded, a Po sample of alpha activity greater than that of the strongest U samples was run periodically in each chamber. No counts were recorded for the Po sample in 1360 hours of observation.

Figs. 1 and 2 show bias curves for one of the chambers and amplifiers used. The curve of Fig. 1 was constructed from a relatively small number of counts recorded (during an actual 25 run) by an Esterline-Angus recording galvanometer. The curve of Fig. 2, very similar in appearance, was made with the aid of a Ra-Be neutron source, using the 25 sample for which the spontaneous rate was measured. It is to be noted that the fission plateau is satisfactorily flat, and that the operating bias is well above the level of alpha disturbances.

Figs. 3 and 4 show the results of an analysis of pulses recorded during actual runs (of equal duration for Po and 25) by an Esterline-Angus recording galvanometer. Fig. 3 shows alpha pile-ups of height less than the minimum required to count, and fission pulses of height greater than the minimum. Fig. 4 compares alpha pile-ups from Po and 25 (observed consecutively in the same chamber at the same amplifier gain) and demonstrates the greater probability of spurious counts from Po than from 25.

The sample of 24 (kindly loaned by Dr. Latimer) was prepared by chemical extraction of UX_1, from uranium and subsequent decay. It contained

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about 10 effective micrograms of 24. It was observed for 1735 hours and it gave no fissions; from this we deduce that λ_z is less than 60 f/gm hr.

Results

If we call λ_x, λ_y, λ_z the spontaneous fission decay constants of 28, 25 and 24 in f/gm hr, we have the observed counting rates for the three samples given by

$$\begin{aligned} X_1\lambda_x + Y_1\lambda_y + Z_1\lambda_z &= C_1 \\ X_2\lambda_x + Y_2\lambda_y + Z_2\lambda_z &= C_2 \\ \lambda_z Z_3 &= C_3 \end{aligned} \qquad (1)$$

It is clear that the term in λ_z can always be neglected in the first two equations (1) because its value is of the order 10^{-3} against terms of the order of unity for the 28.

Using equations (1) we obtained from experiments performed in Berkeley (sea level) $\lambda_x = 20.5 \pm 2.0$, $\lambda_y = 37 \pm 13$. When the experiments were repeated in Los Alamos (Pajarito Canyon Station, altitude 6200 ft) with larger samples, we obtained $\lambda_x = 24.6 \pm 1$, $\lambda_y = 56 \pm 8$.

The agreement in λ_x is satisfactory, but the discrepancy in λ_y is conspicuously large. An estimate of the possible cosmic-ray neutron effect was performed by Bethe on the basis of the paper by Bethe, Korff and Placzek[7], and it was found that, with the known slow neutron cross section of 25 (600×10^{-24} cm^2 for thermal neutrons) and the estimated cosmic-ray

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7) Bethe, Korff and Placzek, Phys. Rev. 57, 573 (1940)

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neutron distribution, one might obtain effects of the order of magnitude of the observed ones.

Shielding by 2 gm/cm^2 of B_2O_3 should, however, be sufficient to absorb practically all the effective cosmic-ray neutrons. The experiments were then repeated with such a shield and we obtained

$$\lambda_x = 24.0 \pm 1.1$$

$$\lambda_y = -2.2 \pm 4.6$$

As to be expected, λ_x was unaffected by the shielding because 28 does not undergo fission by slow neutrons. This confirms the correctness of the interpretation of the effect of the shield. The fast neutron effect is an exceedingly small one.

We shall discuss now the estimated errors in the measurements as given above. In the final runs we have (see equations (1))

$$C_1 = 602/628 = 0.958 \text{ c/hr} \qquad C_2 = 578/859 = 0.673 \text{ c/hr}$$

with standard deviations

$$\Delta C_1 = 0.039 \qquad \Delta C_2 = 0.028$$

The masses X_1 and Y_1 are calculated from the alpha activity of sample 1 (normal uranium) and checked by its weight. We estimate that they may be in error by 2%. We call $X_1 + Y_1 = m_1 = 42.02$ mg of metal. In sample 2 (enriched uranium), the Berkeley system of analysis gives $R = X_2/Y_2 = 2.76$ and the mass $m_2 = X_2 + Y_2 = 41.24$ mg. We estimate that R may be in error by as much as 5% and m_2 by 2%.

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Using the standard formulae of the theory of errors one finds, neglecting some small terms:

$$(\Delta\lambda_x)^2 = (1/m_1)^2 (\Delta C_1)^2 + (C_1/m_1^2)^2 (\Delta m_1)^2 \tag{2}$$

$$(\Delta\lambda_y)^2 = \left[(1+R)/m_2\right]^2 (\Delta C_2)^2 + (R/m_1)^2 (\Delta C_1)^2 + (C_1R/m_1^2)^2 (\Delta m_1)^2 + \left[C_2(1+R)/m_2^2\right]^2 (\Delta m_2)^2 + (C_1/m_1 - C_2/m_2)^2 (\Delta R)^2 \tag{3}$$

The values of the various terms in (2) and (3) for our experiments are

$$(1/m_1)\Delta C_1 = 1.0 \qquad (C_1/m_1^2)\Delta m_1 = 0.5$$

entering in (2), and

$$\left[(1+R)/m_2\right]\Delta C_2 = 3.0 \qquad (R/m_1)\Delta C_1 = 2.8 \qquad (C_1R/m_1^2)\Delta m_1 = 1.3$$

$$\left[C_2(1+R)/m_2^2\right]\Delta m_2 = 1.3 \qquad (C_1/m_1 - C_2/m_2)\Delta R = 1.0$$

entering in (3). As it is easy to see, all the contributions are of the same order of magnitude. A considerable improvement of the precision of the measurements could only be achieved by very great effort and does not seem warranted at present.

ν for spontaneous fission

From our measurements combined with other data it is also possible to calculate the number of neutrons emitted per spontaneous fission. Maurer and Pose[8)] have measured the number of neutrons emitted per second by 8.82 kg of uranium. Their data need recalculation because they are based upon an

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8) Maurer and Pose, Zeitschrift fur Physik, 121, 285, 293, (1943)

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incorrect standardization of the number of neutrons emitted per second by a Rn + Be source. If we assume for this constant 11,350 n/sec per mC of Rn we obtain from the data of Pose that 1 gm of uranium emits 50 neutrons per hour. Dividing this by λ_x we find

$$\nu = 2.1$$

Fermi (private communication) has found that 1 gm of ordinary uranium emits 54 n/hr. From this number, which is probably more accurate than the data of Pose, we find

$$\nu = 2.2$$

These figures may be in error by about 15%.

Another measurement of the neutrons emitted spontaneously by ordinary uranium has been made by Scharff-Goldhaber and Klaiber[9], who found for normal uranium a spontaneous emission of 63.7 neutrons per tram-hour. Only neutrons of energy greater than 100 kv were detected. Assuming that 90% of the neutrons emitted are of energy greater than 100 kv, and taking our value of 24 fissions per gram-hour, one finds from their value for neutron emission

$$\nu = 2.95$$

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9) G. Scharff-Goldhaber and Klaiber, CN-1463

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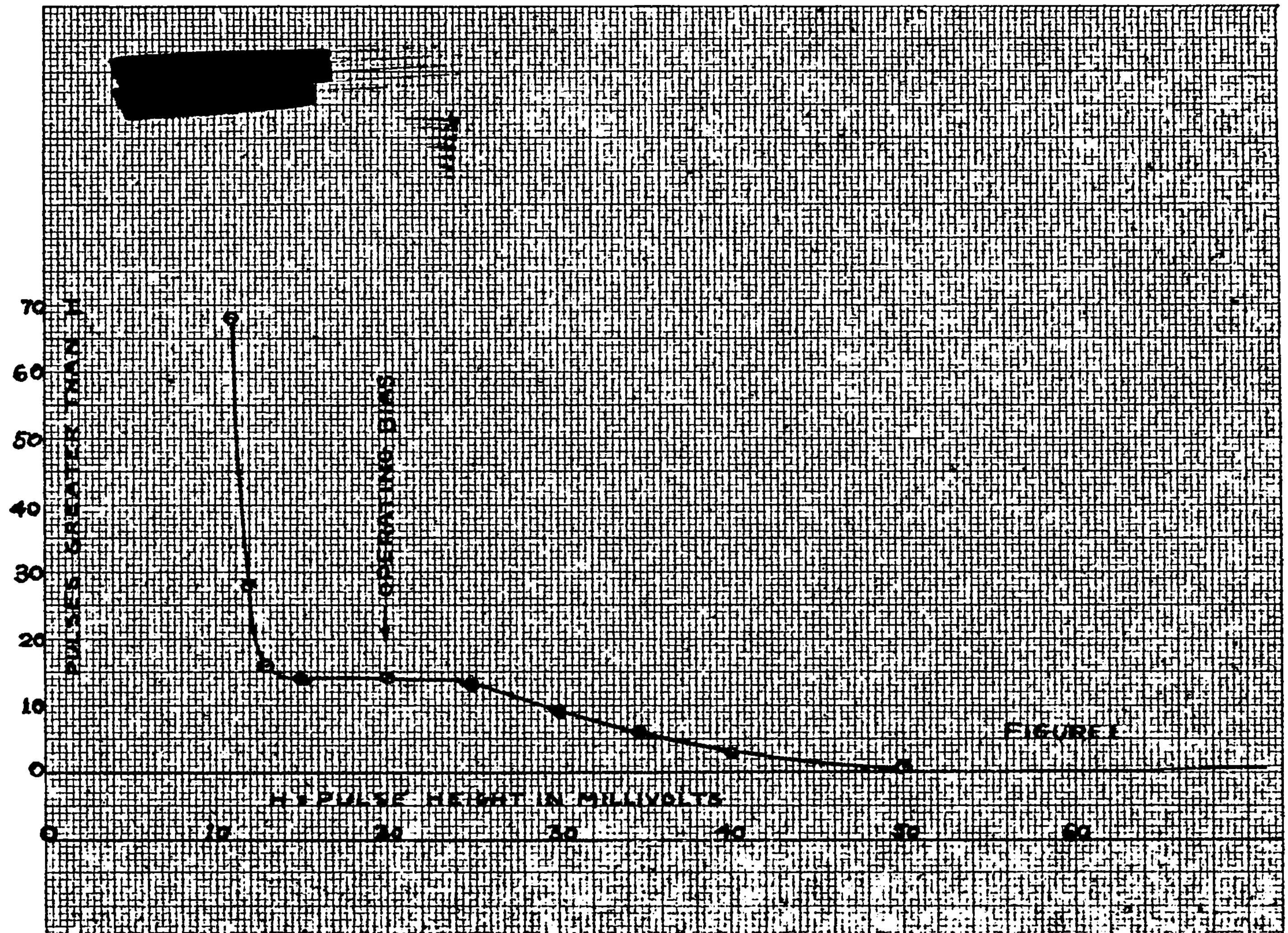

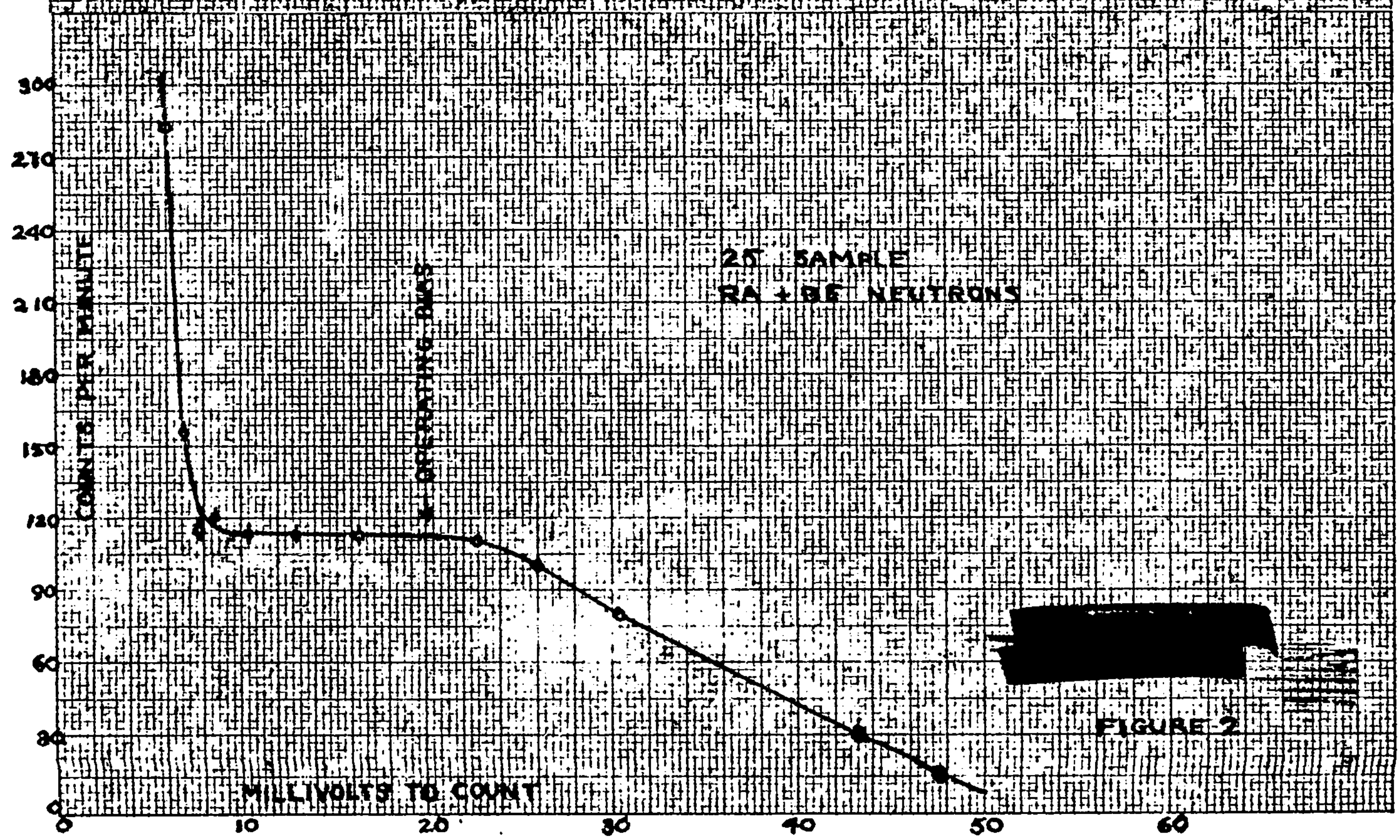

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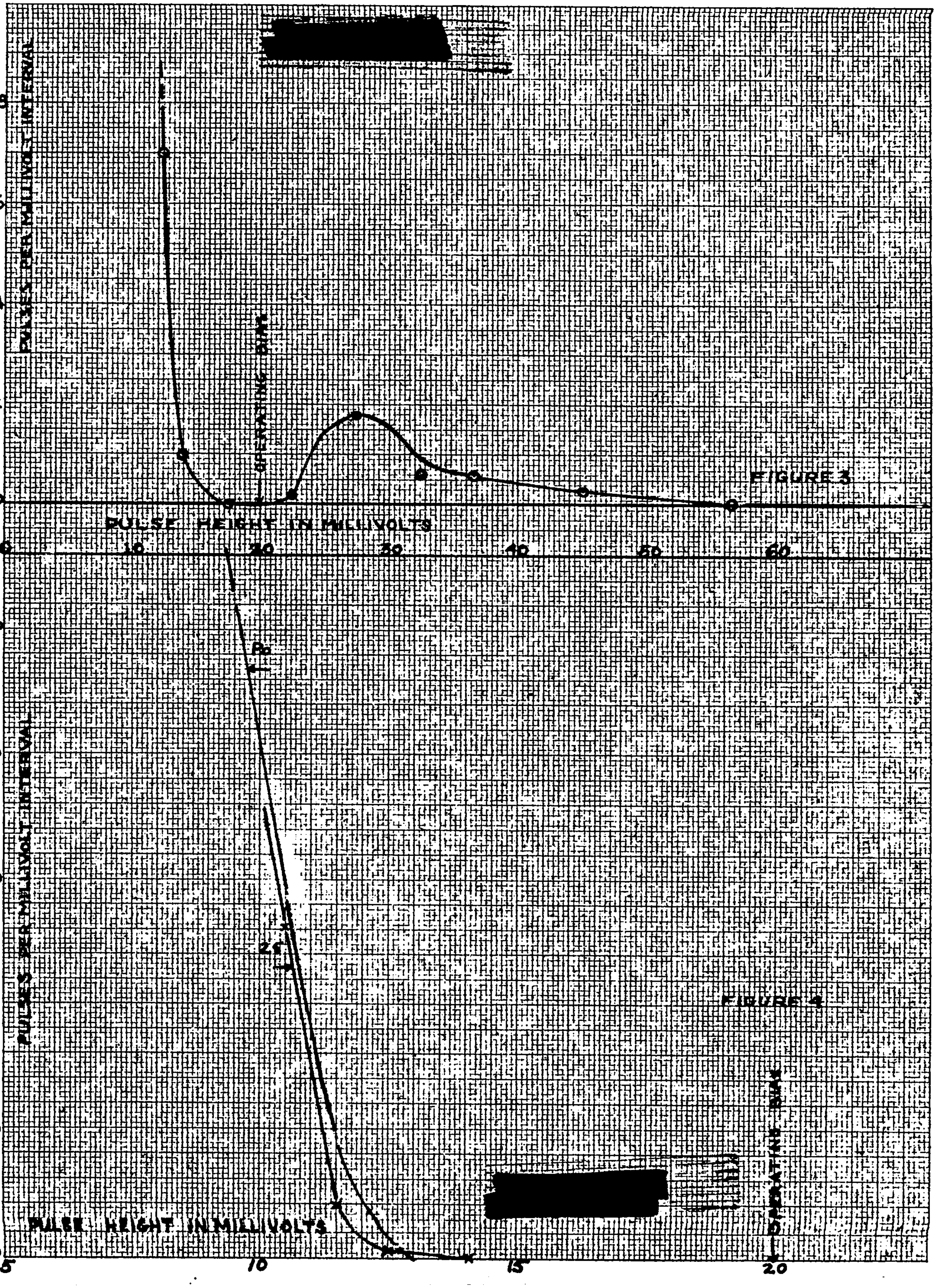
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PULSES PER MILLIVOLT INTERVAL
8
6
4
2
0
OPERATING BIAS
FIGURE 3
PULSE HEIGHT IN MILLIVOLTS
0
10
20
30
40
50
60
500
400
300
200
100
0
Po
PULSES PER MILLIVOLT INTERVAL
FIGURE 4
OPERATING BIAS
[redacted]
PULSE HEIGHT IN MILLIVOLTS
5
10
15
20

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