







# THE DEVELOPMENT OF A SCINTILLATION. COUNTER

by

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## INTRODUCTION

Detection of ionizing radiations by the fluorescence they cause in certain substances was first performed by Lord Rutherford. It was found that a single  $\alpha$  particle on hitting a zinc sulphide screen produced enough light for the flash to be registered visually. This technique could not be applied to other radiations then known ( $\beta$  and  $\gamma$  rays) since these have considerably lower ionizing power and the energy released in the fluorescent screen was below the sensitivity of the eye. The principle however remained and awaited the appearance of a light detecting instrument more sensitive and reliable than the human eye.

A simple photo-cell could not be successfully applied to the problem, since the light energies involved are so low that the response to a scintillation is masked by the thermal or Johnson noise of the resistor coupling the photo-tube to the next stage.

An instrument suitable for the detection of individual scintillations is the electron multiplier photo-tube. This gives an internal amplification of the order of  $10^6$  and thereby raises the response

to a single scintillation far above the Johnson noise of the coupling resistor as well as all other statistical noises of the subsequent stages.

The photo-multiplier tubes now commercially available have considerable dark current. Thus the problem of constructing an efficient radiation detector is centred on the following points:

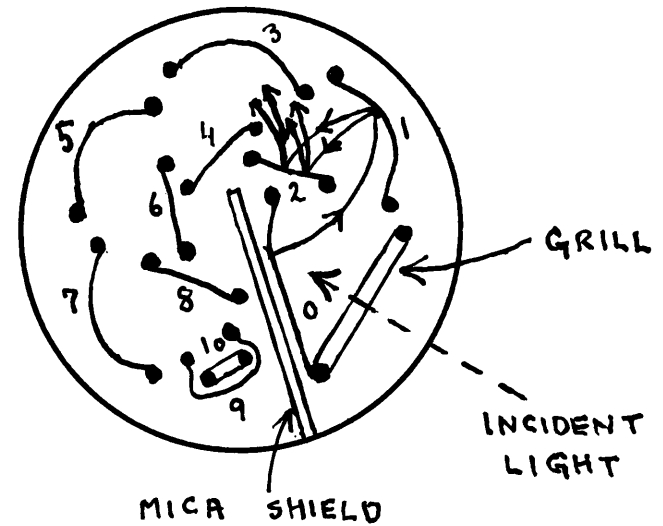
- (1) reducing the dark current;
- (2) finding effective means of differentiating between dark current pulses and useful pulses; and
- (3) selecting an efficient phosphor.

The object of this experiment was to construct a counter employing a fluorescent screen and a commercial photo-multiplier tube, suitable for work with  $\gamma$  rays. Particular attention was paid to the development of practical ways of reducing the high background rate of the counter, while keeping the intrinsic efficiency at a moderately high level.

## Photo-multiplier Considerations.

The electrostatically focused photomultiplier tube employs secondary emission multiplication to obtain an overall gain of approximately  $10^6$  at 100 volts per stage.

The gain varies considerably with the dynode voltages and is only 25,000 at 60 volts per stage.



0 = photocathode  
10 = anode  
1-9 = dynodes

The sensitivity of the multiplier tube photo-cathode is only about  $1/4$  of that of the two element photo-cell, the advantage of the photo-multiplier is that the large amplification obtained through secondary emission at the subsequent dynodes is accomplished with next to no Johnson effect, which appears only in the output stage. It is by virtue of this large internal amplification that external noise sources can be made negligible. The internal noise source - the dark current of the tube - has a magnitude of  $10^{-14}$  amperes at room temperature. According to R. Engstrom<sup>(1)</sup> it has the following origin:

- (1) thermal emission at the photo-cathode and succeeding dynodes
- (2) ohmic leakage
- (3) regenerative emission.

His results indicate that the second of these is dominant below 60 volts per stage and the third begins somewhere above 110 volts per stage depending upon the tube.

Since the amplified dark current of thermionic origin increases at the same rate as the useful signal, it is best to use the tube in the region where noise of other origin is relatively unimportant. This condition is satisfied by dynode voltages between 60 and 100 volts.

Engstrom's experimental measurements further show an increase in the signal to noise ratio with decrease of dynode voltages. In his experiments refrigeration of the tube to  $-192^{\circ}\text{C}$  gave a 100 fold decrease in dark current pulse count.

The output pulses of the tube will show a statistical variation in shape owing to the statistical nature of multiplication and also the differences in integrated transit times through the ten successive stages of the multiplier tube. Average width of a pulse is approximately 1  $\mu\text{sec}$ . and there are approx-

imately  $10^4$  dark current pulses per second at room temperature.

The most sensitive tubes are the R.C.A. types 1P21 and 931A. These have maximum sensitivity at  $4,200 \text{ \AA}$ . The construction of the two types is identical. They are graded on the basis of sensitivity, after manufacture. The 1P21 is the more sensitive one.

Since in this type of work the main consideration is the signal to noise ratio rather than the sensitivity, we considered it expedient to buy a large stock of the 931A type and select the best ones.

The selection was made by actual trial of the tube as a radiation detector. It was found necessary to give the tubes a preliminary warming-up period of about one hour, since the dark current of a tube during its first half hour or so of operation was seen to be unusually high. Out of 18 tubes tested, the three best ones were selected and these gave an approximately equal performance.

## Review of related Papers.

The first application of the photo-multiplier tube to measurement of radioactivity was made by Blau and Dreyfus<sup>(2)</sup> in 1941. The activity of  $\alpha$  ray sources was obtained by measuring the integrated current output of a 931A tube placed a short distance away from a copper-activated zinc sulphide screen. The instrument was calibrated by a source of known strength.

In 1947, Fitz Hugh Marshall and J.W. Coltman<sup>(3)</sup> used a photo-multiplier and silver-activated zinc sulphide (Patterson D) screen to detect and count individual  $\alpha$ ,  $\beta$  and  $\gamma$  rays. The screen was deposited on the envelope of the tube opposite the most sensitive part of the photo-cathode.

Most of the dark current pulses were discriminated against on the basis of their carrying less charge than useful pulses. In the case of  $\beta$  and  $\gamma$  rays, it was found necessary to use a collective mirror which focussed the light on the most sensitive part of the photo-cathode.

The efficiency of the counter for  $\gamma$  rays was reported to be somewhat higher than that of a Geiger counter. The dark current count was considerable.

J. Kallman suggested that the low efficiency is due to the fact that the phosphor used is opaque to its own luminescence. He used thick naphthalene blocks instead of zinc sulphide with much better results. He

further suggested the possibility of placing the crystal between two photo-multiplier tubes connected in coincidence.

This work was repeated by M. Deutch<sup>(4)</sup>, who claims a  $\gamma$  ray intrinsic efficiency of 20%, with presumably tolerable background. The background was greatly reduced by cooling to about  $-40^{\circ}\text{C}$ . He further claims good results with the coincident arrangement. The counter was insensitive to  $\alpha$  rays. Very low energy  $\beta$  rays could not be detected, the detection of moderately low energy  $\beta$ 's required refrigeration of the tube, while high energy  $\beta$ 's were detected with an efficiency approaching 100 %.

The decay time of the fluorescence was shown to be  $10^{-7}$  sec.

Since Geiger counters of high efficiency are available for detecting  $\alpha$  and  $\beta$  rays it appears that in its present stage of development the photo-multiplier counter is of interest chiefly as a  $\gamma$  ray counter, where its intrinsic efficiency is higher than Geiger's by a factor greater than 10. In the case of  $\alpha$  rays this type of counter allows the use of a thinner window with an efficiency of the same order (close to 100 %).

The speed of response of a naphthalene counter is limited by the speed of the photo-multiplier tube and therefore at least 10 times greater than the speed of response of a fast Geiger counter.

The chief drawback is the high background rate. Cooling of the photo-multiplier tube to reduce the background count is inconvenient, and renders the instrument cumbersome.

### Single Photo-multiplier Tube counter.

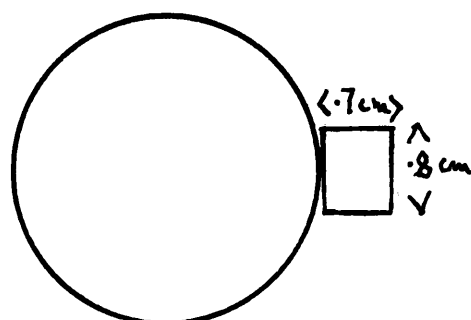
The design of the single photo-multiplier tube counter was patterned after Marshall and Coltman except that no collecting mirror was used.

Discrimination against most dark current pulses was on the basis of amplitude, after integration by the two RC circuits,  $R_1C_1$  and  $R_2C_2$  (Refer to Diagram 1.)

The voltages for the dynodes were supplied by a 1,000 volt unregulated power supply.  $R_0$  is a gain control for the photo-multiplier. The photo-multiplier output pulses were amplified by the pentode 220 fold.

Both Patterson D screens  
and naphthalene blocks were  
tried. The naphthalene block  
was approximately .7 cm.  
thick, the other two dimensions were .8 cm. and 1.35 cm.  
(Refer to Diagram). Naphthalene  
appeared to be more efficient

X (2)



than zinc sulphide for  $\gamma$  rays of Ra. by a factor of well over ten, but gave no response to  $\alpha$  rays.

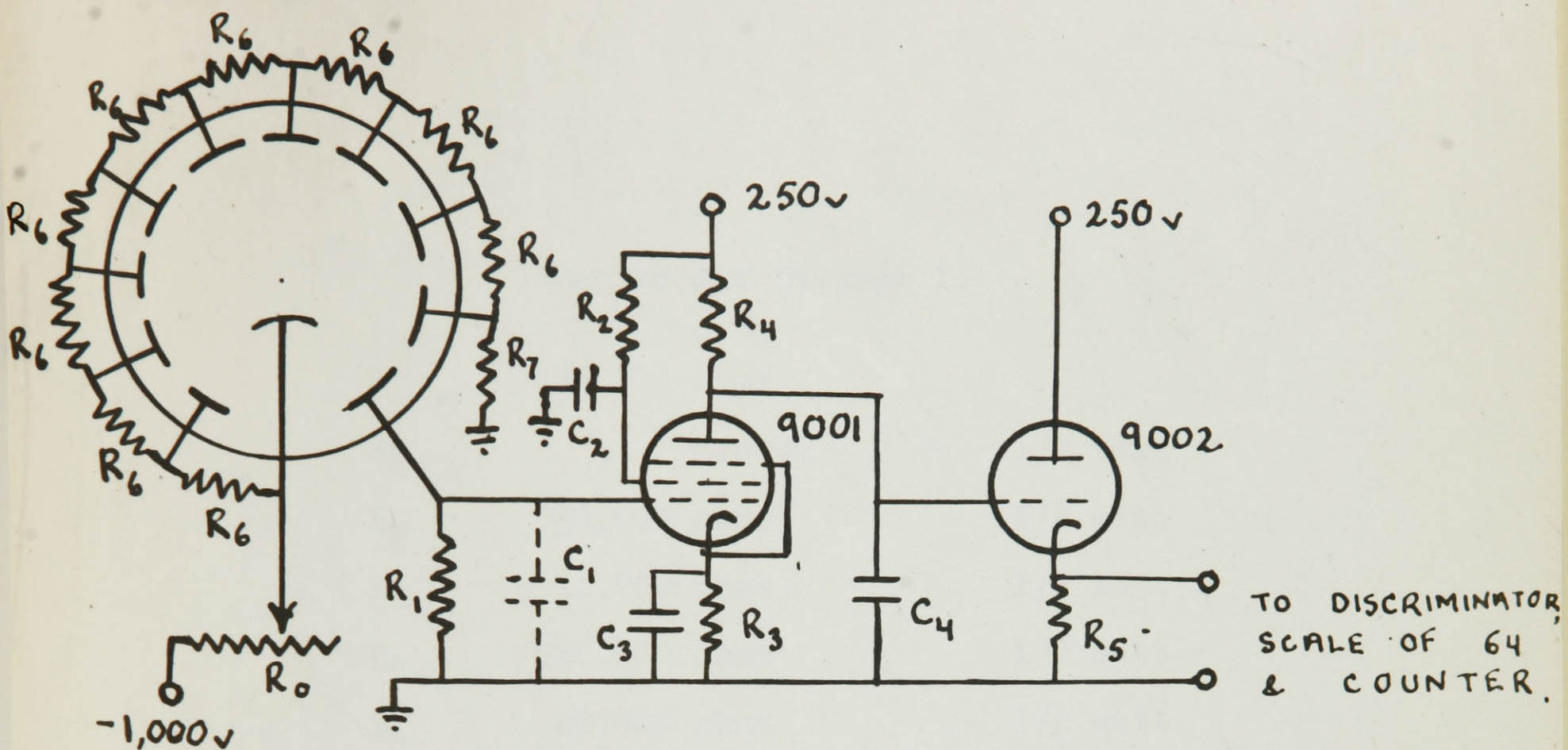
X (3)

Dark current pulses could not be completely eliminated by the method of discrimination used. For different settings of the discriminating bias, the efficiency increased together with the number of dark

current pulses transmitted. Decreasing the discrimination level below 12 volts, which corresponded to approximately 200 dark current pulses per second, there was a far greater increase in the dark current than in useful pulses. This setting corresponded to approximately 12% efficiency. No appreciable difference in the efficiency of the detector could be noticed on changing the position of the source as shown. (Refer to diagram, P.9)

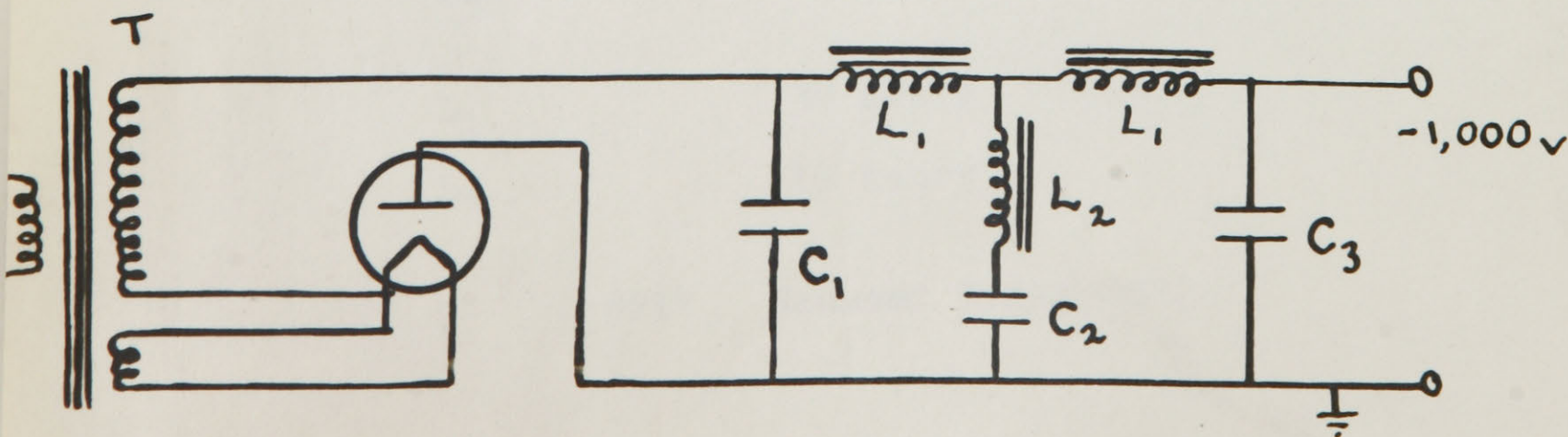
From the geometry of the arrangement it follows that the photo-cathode receives approximately between  $1/13$  and  $1/50$  of the light emitted at a point within the crystal, depending on the location of the point. The efficiency obtained, and the fact that even then many pulses detected by the photo-cathode were not counted, indicated that a scintillation due to a single  $\gamma$  photon usually yields a number of light photons much greater than one.

This and the failure to obtain any indication of a preference of direction on the part of light photons suggested grouping two photo-multiplier tubes around a single naphthalene block in coincidence, so as to increase the signal to noise ratio of the counter.



↑  
DIAGRAM 1.

DIAGRAM 2.



Legend for Diagram 1.

|                |              |          |
|----------------|--------------|----------|
| R <sub>0</sub> | 250,000 ohms |          |
| R <sub>1</sub> | 1 megohm     | 1/2 watt |
| R <sub>2</sub> | 330,000 ohms | 1/2 watt |
| R <sub>3</sub> | 2,200 ohms   | 1/2 watt |
| R <sub>4</sub> | 270,000 ohms | 1/2 watt |
| R <sub>5</sub> | 20,000 ohms  | 1/2 watt |
| R <sub>6</sub> | 50,000 ohms  | 1/4 watt |
| R <sub>7</sub> | 25,000 ohms  | 1/4 watt |
| C <sub>1</sub> | 11 $\mu$ pf  |          |
| C <sub>2</sub> | 0.05 $\mu$ f |          |
| C <sub>3</sub> | 10 $\mu$ f   |          |
| C <sub>4</sub> | 50 $\mu$ pf  |          |

Legend for Diagram 2.

|                |                          |
|----------------|--------------------------|
| C <sub>1</sub> | .2 $\mu$ f               |
| C <sub>2</sub> | .2 $\mu$ f               |
| C <sub>3</sub> | 2 $\mu$ f                |
| L <sub>1</sub> | 30 henry                 |
| L <sub>2</sub> | 10 henry                 |
| T              | #214 Hammond Transformer |

### The Coincidence Arrangement.

The arguments which have led to the idea of trying a coincidence arrangement can be extended to give an indication of how the efficiency of this arrangement will compare with that of the single tube counter.

The intrinsic efficiency of a single tube counter can be written as a product of two terms:

$n$  - the probability that a  $\gamma$  photon passing through the crystal will have a part of its energy converted into light energy  
and  $a$  - the probability that the photo-multiplier will detect a scintillation produced.

If it is further assumed that the major contribution to  $a$  comes from large scintillations, the efficiency of a coincidence counter will be given by

$$a^2 n$$

since for large scintillations the probability that one tube will detect the signal is almost independent of whether the other one has detected it or not.

Since  $n$  will be considerably less than one it can be expected that the efficiency of the coincidence counter will be appreciably higher than the square of the efficiency of the single tube counter, when both are operated with the same discriminator biases.

The weakness of the above argument is that in effect it assumes that a  $\gamma$  ray passing through a crystal produces a "large" scintillation or none at all.

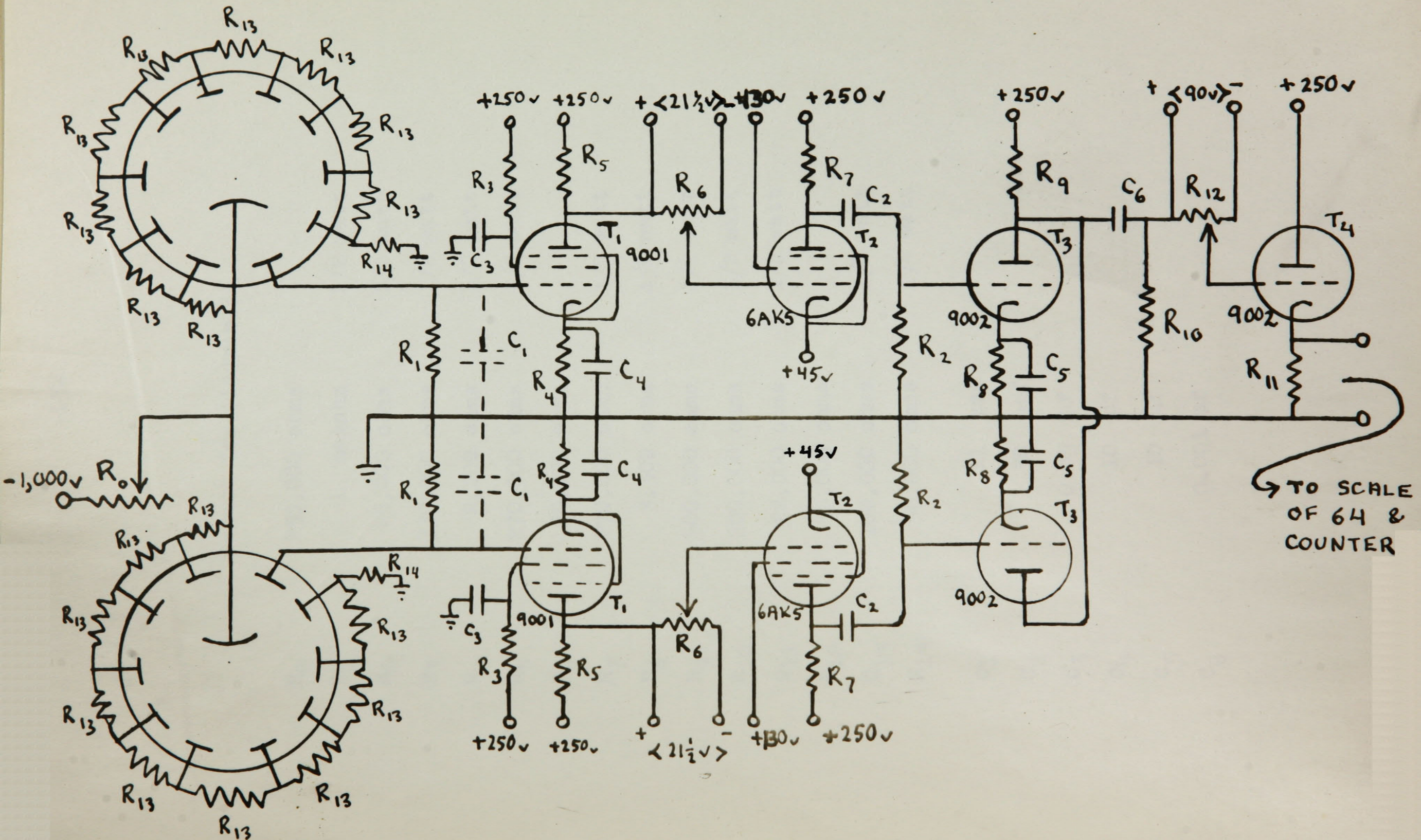
Diagram 3 gives the circuit used. The voltages for the dynodes of the two photo-multipliers were supplied by a common 1,000 volt power supply. Separate power packs were used for each amplifier with its discriminator and also for the final stage to avoid inter-coupling. All the power supplies were well filtered but unstabilized.

The output of each photo-tube is amplified by approximately 220 and then led to a discriminator which serves to cut out most of the dark current pulses and thus reduce the number of random coincidences. Discrimination is helped by the  $R_1C_1$  output of the photo-multiplier. The  $R_2C_2$  circuit serves to sharpen the pulses from the discriminator. Oscilloscope study of these sharpened pulses indicated half amplitude width of approximately 10 nsec. The pulses are then applied to the grids of  $T_3$ 's which are in Rossi arrangement for counting coincidences. The plates are joined together and when both tubes are conducting the common potential of the plates is 12 volts. If only one tube receives a

negative signal and is cut off, the potential of the plates rises to 23 volts, while if both receive a negative signal at the same time the potential rises to 250 volts. Non-coincidences are discriminated against with the help of  $T_4$ .

While the conventional Rossi circuit is designed for pentodes, it was found that better resolution could be obtained here by using triodes. This is due to the fact that pulses fed into the circuit were roughly triangular in shape and of uneven amplitude. The remote cut off of the triodes helped discriminate against incomplete coincidences.

DIAGRAM 3.



Legend for Diagram 3.

|                 |              |          |
|-----------------|--------------|----------|
| R <sub>0</sub>  | 250,000 ohms |          |
| R <sub>1</sub>  | 1 megohm     | 1/2 watt |
| R <sub>2</sub>  | 25,000 ohms  | 1/2 watt |
| R <sub>3</sub>  | 330,000 ohms | 1/2 watt |
| R <sub>4</sub>  | 2,200 ohms   | 1/2 watt |
| R <sub>5</sub>  | 270,000 ohms | 1/2 watt |
| R <sub>6</sub>  | 50,000 ohms  |          |
| R <sub>7</sub>  | 270,000 ohms | 1/2 watt |
| R <sub>8</sub>  | 3,300 ohms   | 1/2 watt |
| R <sub>9</sub>  | 500,000 ohms | 1/2 watt |
| R <sub>10</sub> | 200,000 ohms | 1/2 watt |
| R <sub>11</sub> | 22,000 ohms  | 1/2 watt |
| R <sub>12</sub> | 50,000 ohms  |          |
| R <sub>13</sub> | 100,000 ohms | 1/4 watt |
| R <sub>14</sub> | 50,000 ohms  | 1/4 watt |
|                 |              |          |
| C <sub>1</sub>  | 11 μpf       |          |
| C <sub>2</sub>  | 50 μpf       |          |
| C <sub>3</sub>  | 0.05 μf      |          |
| C <sub>4</sub>  | 10 μf        |          |
| C <sub>5</sub>  | 10 μf        |          |
| C <sub>6</sub>  | 0.001 μf     |          |

## Results.

The performance of the counter was tested with  $\gamma$  rays from .27 millicurie of Radium. As with the single photomultiplier tube counter it was found that the efficiency of the detector increased with the number of dark current pulses transmitted. An estimate of the intrinsic efficiency was made for discriminators settings such that two dark current pulses per second were counted. It was assumed that the disintegration of one atom of Radium yields on the average 2.29  $\gamma$  ray photons and any variation in efficiency according to the energy of the different groups of  $\gamma$  rays resulting from the disintegration of Radium was neglected. The result was approximately 6%.

This corresponds to a 100 fold decrease of the background rate for a two fold decrease in efficiency. Modern coincidence counters have a resolving time as small as  $1/3$   $\mu$ sec. Such equipment would reduce the dark current pulses counted by another factor of thirty.

An inverse square law plot was made to verify that the coincidences recorded are neither accidental nor due to integrated pulses.

A comparison of the efficiency of the coincidence counter with that of each of its two sections was made to test the conclusions reached on page 12.

This comparison could be made only at low efficiencies, on account of the high background count when the tubes were used singly. A sample result is given below.

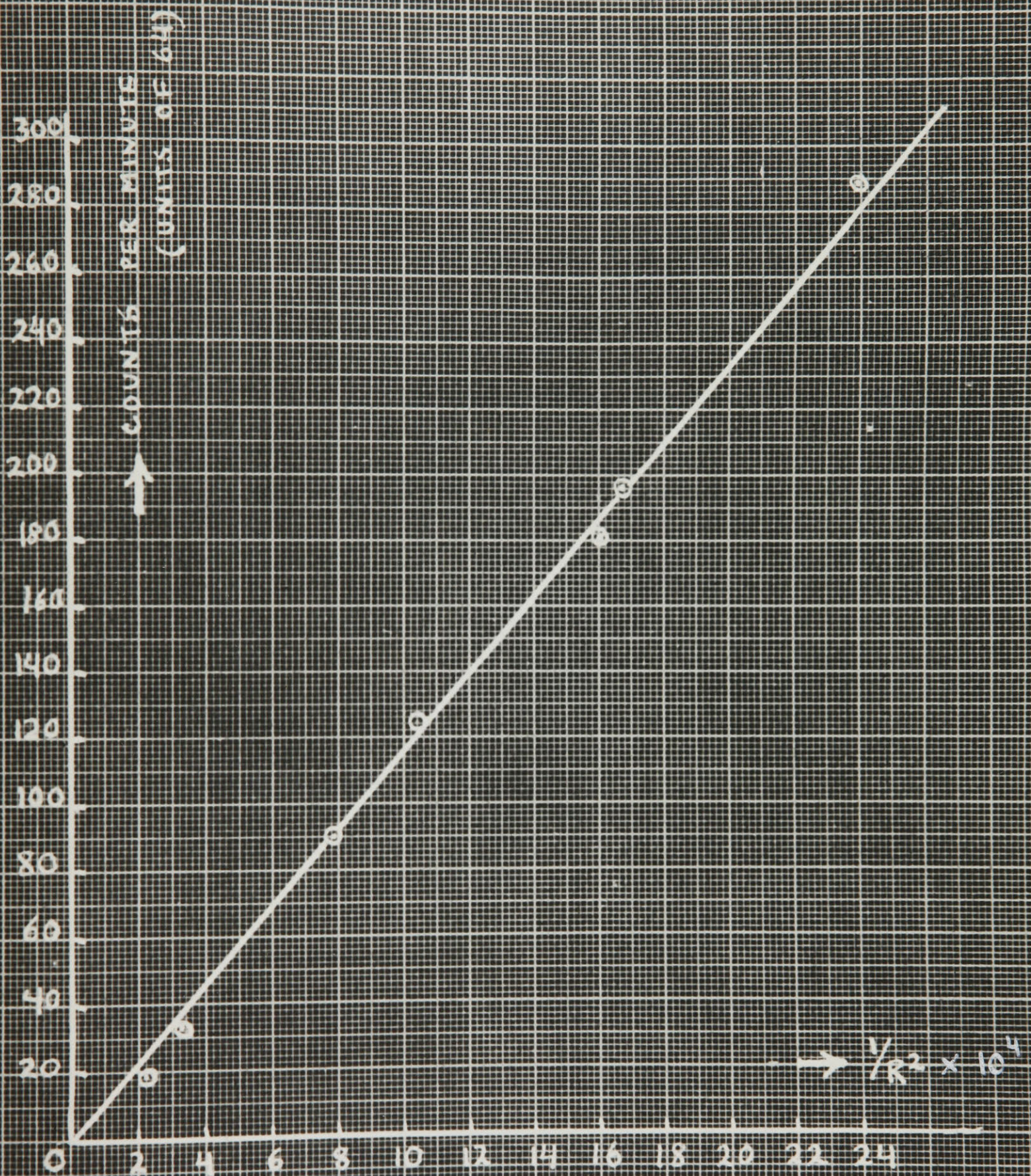
|                            |    |
|----------------------------|----|
| Efficiency of each section | 6% |
|----------------------------|----|

|                 |       |
|-----------------|-------|
| Square of above | 0.36% |
|-----------------|-------|

Efficiency of coincidence counter 1.3%.

A plot of relative efficiency against background count per minute was made. The result was a smooth curve without a plateau to indicate the best region of operation for the counter. At higher efficiencies the increase in the background became more and more rapid.

# INVERSE SQUARE LAW PLOT



Data for Inverse Square Law Plot.

| Distance of source<br>from counter in cm.<br>R | True counts<br>per minute<br>in units of 64 | $\frac{1}{R^2} \times 10^4$ |
|------------------------------------------------|---------------------------------------------|-----------------------------|
| 23.5                                           | 196                                         | 16.7                        |
| 36                                             | 90                                          | 7.85                        |
| 68                                             | 19                                          | 2.2                         |
| 55.5                                           | 33                                          | 3.3                         |
| 31                                             | 124                                         | 10.4                        |
| 20.5                                           | 289                                         | 23.9                        |
| 25                                             | 181                                         | 16                          |

Intrinsic Efficiency of counter.

No. of  $\gamma$  rays emitted by source per second

$$\begin{aligned}
 &= .27 \times 3.7 \times 10^7 \times 2.29 \\
 &= 2.29 \times 10^7
 \end{aligned}$$

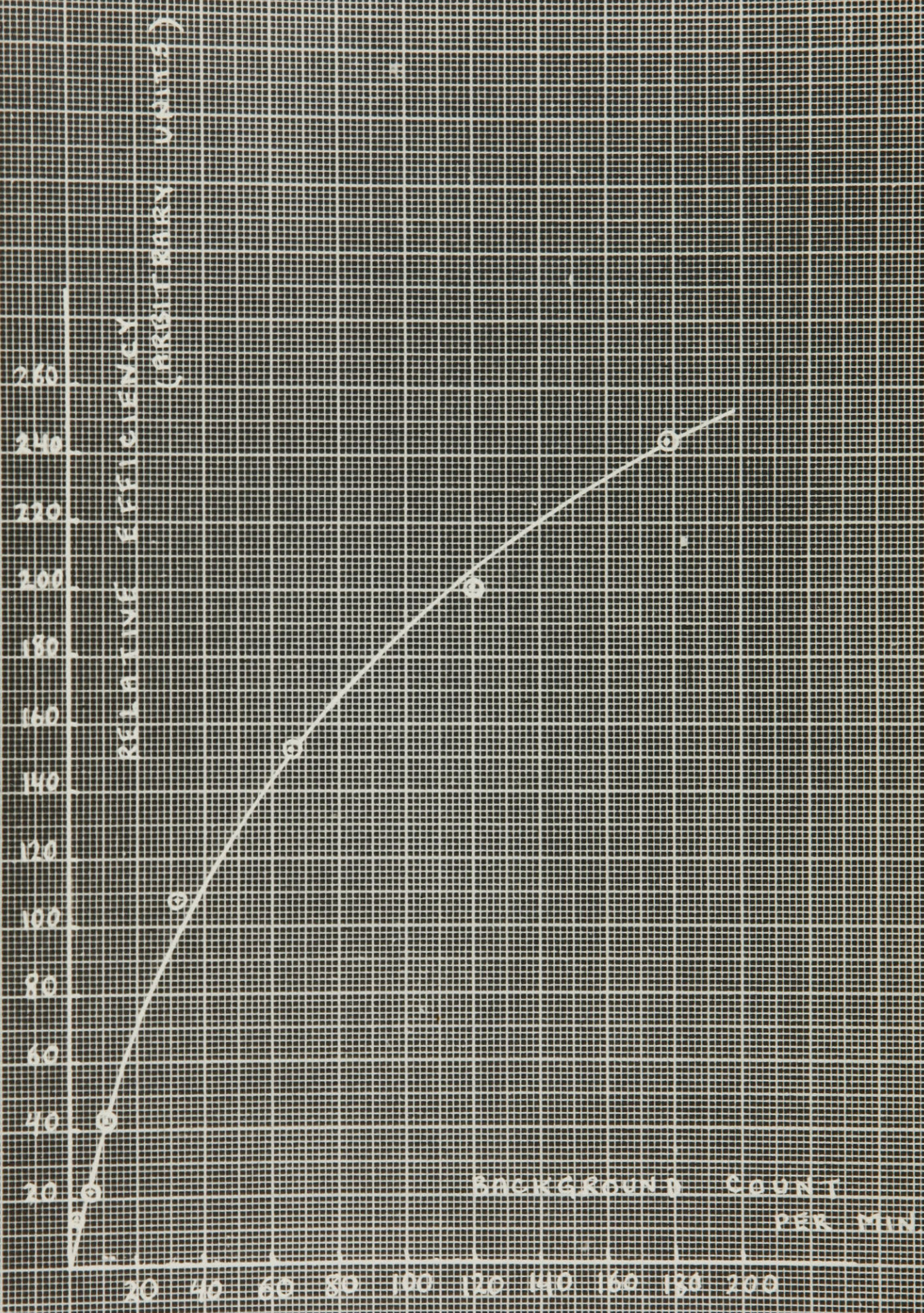
No. incident on crystal of surface area 1.08 sq. cm. and  
25 cm. away

$$= \frac{2.29 \times 10^7 \times 1.08}{25^2 \times 4\pi} = 3.14 \times 10^3$$

Hence Intrinsic Efficiency

$$\begin{aligned}
 &= \frac{187 \times 64}{60 \times 3.14 \times 10^3} \quad (\text{from graph}) \\
 &= 6.3\%
 \end{aligned}$$

RELATIVE EFFICIENCY AGAINST BACKGROUND RATE CURVE



Data for relative efficiency against background rate curve.

| No. of true counts per min.<br>in units of 64 | Background count per minute |
|-----------------------------------------------|-----------------------------|
|-----------------------------------------------|-----------------------------|

|     |     |
|-----|-----|
| 244 | 198 |
| 201 | 120 |
| 152 | 65  |
| 108 | 31  |
| 43  | 10  |
| 74  | 22  |
| 28  | 7   |
| 15  | 1.5 |
| 22  | 4.5 |

Note: Since the true counting rate depends not only on the background counting rate but also on the individual settings of the discriminators, in the two sections of the counter, only very general conclusions can be drawn from the curve.

### CONCLUSIONS

In view of the lack of detailed published reports on the results of work carried out along these lines elsewhere, it is extremely difficult to evaluate the performance of the instrument developed. It seems, however, that the practicability of the use of the coincidence arrangement as a method of reducing the background counting rate, with a correspondingly small loss of intrinsic efficiency, was demonstrated.

Unfortunately the geometry of this arrangement tends to be such that high total efficiency cannot be obtained. Thus in applications where total efficiency is the prime consideration or in coincidence experiments, where the comparatively high background rate would be unimportant a single tube counter would be preferred. Here larger size crystals could be used and the sample could be brought right up to the crystal.

A comparison with a Geiger counter is complicated by the fact that the relative importance of higher efficiency and lower background will depend on the strength of the source to be measured, and the time allotted for the measurement. It is clear, however, that in cases where the source is sufficiently strong

to make the probable error in the background count considerably smaller than the probable error in the true count, the photo-multiplier counter will give an equally reliable information in much shorter time.

The following figures will illustrate this point. Suppose that 2 minute activity measurements of a  $\gamma$  ray source are made with

- (1) a single tube photo-multiplier counter having a background of 200 counts per second and an overall efficiency of 1%
- (2) a Geiger counter, whose overall efficiency is 1/10 that of the photo-multiplier counter.

Let the results of the two measurements be

|                                   |        |
|-----------------------------------|--------|
| Photo-multiplier background count | 24,000 |
| Photo-multiplier total count      | 36,000 |

the standard deviations will be 157 and 190

The probable error will then be less than  $\frac{347}{12,000}$

i.e. less than 2.9%.

The Geiger counter will then give a count of 1,200 and a probable error of 34.6, i.e. about 2.9%.

These results would be obtained with a source emitting  $10^4$   $\gamma$  photons per second.

If the source used is 10 times stronger, the results would be

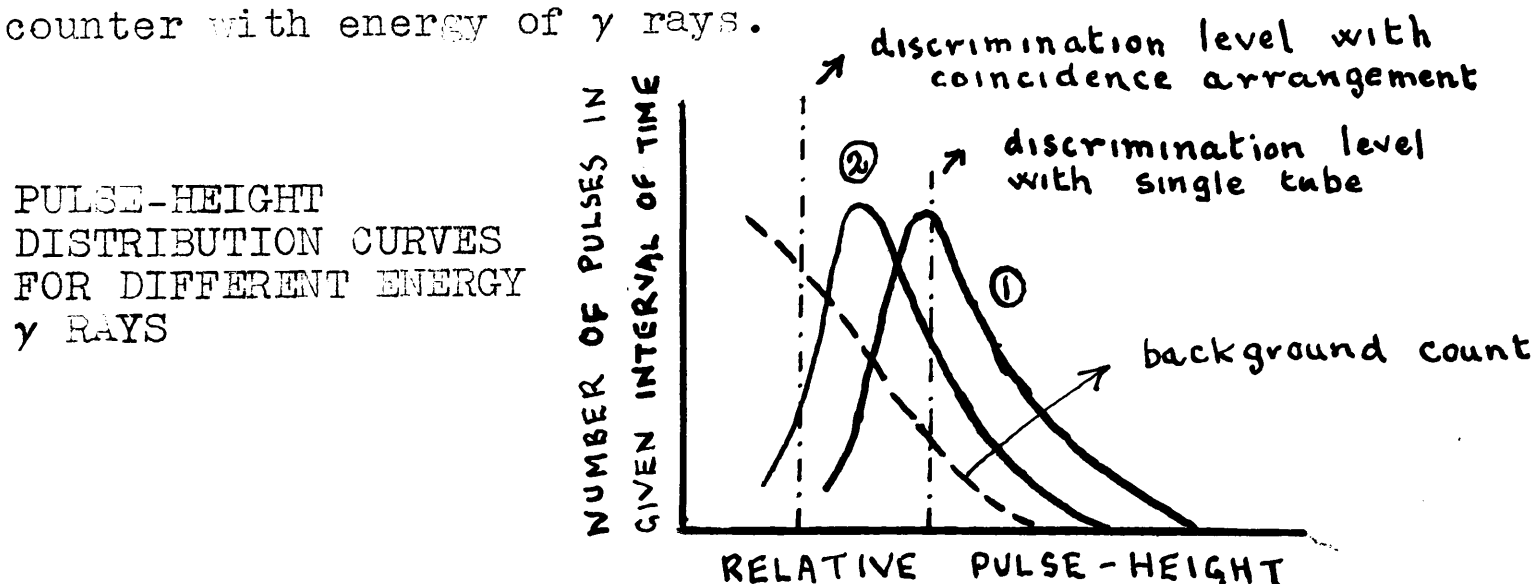
|                             |         |
|-----------------------------|---------|
| Multiplier background count | 24,000  |
| Multiplier total count      | 144,000 |

The standard deviations will be 157 and 380 which will imply a probable error less than  $\frac{537}{144,000}$  i.e. less than 0.37%.

The Geiger counter would give a count of 12,000 with standard deviation of 109 and a probable error of 0.9%.

The smaller dead time of the counter is an advantage to the extent of the inaccuracy attendant to dead time corrections.

It is interesting to notice the effect of the coincident arrangement on the variation of efficiency of the counter with energy of  $\gamma$  rays.



Such variation must result from the method of discrimination against dark current pulses used.

Let  $m_1$  be the efficiency of a single tube counter for a certain energy  $\gamma$  and corresponding to the level used. Let  $m_2$  be the efficiency for a different energy  $\gamma$ . The percentage variation in efficiency is then

$$\frac{m_1 - m_2}{m_1} \text{ and let } m_1 > m_2$$

A coincidence counter would use a lower discrimination level, and its photo-multipliers if used singly would have higher efficiencies, say  $n_1$  and  $n_2$  for the two energies we are considering.

The efficiency of the combination will then be proportional to  $n_1^2$  and  $n_2^2$  respectively.

Then the percentage variation in efficiency is

$$\frac{n_1^2 - n_2^2}{n_1^2} = \frac{(n_1 - n_2)}{n_1} \times \frac{n_1 + n_2}{n_1}$$

Since  $\frac{n_1 + n_2}{n_1}$  is  $< 2$ ,

it follows that if the discrimination level for the coincidence arrangement can be dropped sufficiently to make

$$\frac{n_1 - n_2}{n_1} < \frac{1}{2} \frac{m_1 - m_2}{m_1}$$

the coincidence arrangement will have a smaller variation in efficiency than a single tube counter.

## APPENDIX

Preparation and mounting of screens.

Zinc Sulphide Screen.

The following method was used to deposit a zinc sulphide screen on the glass envelope of the photo-multiplier. A pinch of the powder was placed on the glass envelope opposite the photo-cathode. By tapping the tube gently, the crystals were made to form a thin even layer. This was sprayed with methyl alcohol containing a few drops of shellac per 50 cc. of alcohol.

Unwanted portions of the screen were then scraped off, the clear part of the envelope given a coat of Kodak Opaque, several coats of black enamel, and the cylindrical part of the envelope wrapped in aluminium foil.

Napthalene crystals.

Melting pure napthalene in bulk and letting it cool gradually over a period of two days produced large clear portions in the solid. These were cut out, scraped to size on rough emory cloth and then polished to good transparency with fine emory cloth.

Since napthalene sublimates, the efficiency of a crystal drops during the course of a few days. It was therefore considered advisable to coat the crystals

with water glass. Thin layers are difficult to apply on account of the surface tension of water glass and the oily surface of naphthalene.

In the coincidence arrangement the crystal was housed in a box, three walls of which were formed by plane mirrors, two sides faced the photo-cathode and the remaining wall formed the window of the counter.

The whole was blacked out in a glyptol painted cardboard box.

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