Declaration.

The author desires to state that he received no assistance in the writing of this thesis. The conditions in which the work described in Part I and Part II was carried out are given in the Foreword.

Foreword.

The course of research carried out by the present author consisted of the following work. During the first year of study the author took part in the design and erection of the main columns and pumping system of the Glasgow High Tension Set, under the supervision of, and in collaboration with Dr. J.B. Warren. From August, 1947 to January, 1948 an investigation was made of the properties of a specially designed high pressure ionization chamber incorporating a gridded electrode. This study, the results of which are reported in Part I of this thesis, was carried out solely by the author, except for the initial stages of development, in which he collaborated with Dr. Warren. Starting in January, 1948 the author undertook an investigation which had been initiated by Dr. S.C. Curran of the properties of proportional counters with special reference to the possibility of the measurement of energies and intensities of electrons and electromagnetic radiations of low energies. In the initial stages of this study he worked under the supervision of Dr. Curran, and in the subsequent stages he actively collaborated with Dr. Curran and Mr. A.L. Cockcroft. The studies were extended to an investigation of the beta-ray spectra of tritium and carbon 14 and the gamma ray spectrum of radium D. The results of this investigation are given in Part II of this thesis. In the initial experiments the author was concerned principally

in the investigation of the characteristics of the counters for the recording of X-rays and electromagnetic radiations, and carried out the investigation of the resolution of the instrument and the variation of out-put pulse size with the voltage applied to the cathode, the results of which are contained in Section 5. He was also concerned principally in the measurements carried out on radium D described in Section 9. In the more recent experiments on tritium and carbon he participated fully in the preparation of the sources and in the recording and analysis and interpretation of the results.

Acknowledgements.

The author would like to thank Professor P.I. Dee and Professor T. Alty who acted as supervisors during this research for the interest which they showed in these studies and the advice which they gave.

He would also like to thank Dr. J.B. Warren with whom he collaborated in the initial stages of the work described in Part I of this thesis, and Dr. S.C. Curran and Mr. A.L. Cockcroft with whom he collaborated in the work described in Part II. He has pleasure in acknowledging the many discussions which he had with them and the advice which they gave.

The author is much indebted to the Ministry of Education of Northern Ireland from whom he received a research grant which enabled him to carry out this work.

PART I.

THE INVESTIGATION OF SMALL AMOUNTS OF IONIZATION WITH A GRIDDED IONIZATION CHAMBER.

John Angus, M.Sc.

March, 1949.

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SECTION 1.

INTRODUCTION.

Studies of the ionization produced by the interaction of a fast particle with the atoms of the gas encountered along its track form an important field of research in nuclear physics. In the case of a particle which begins and ends its trajectory within the volume under observation the complete track of the particle is obtained, and the total number of ion pairs produced by the particle may be measured. If the average energy required to produce one ion pair is known, for example by means of a calibration experiment using particles of known energies, the energy of a particle may be calculated from the number of ion pairs which it produces.

On the other hand, for a particle with a considerably longer range, as, for example, an electron or meson of high energy, information of a different nature may be obtained. For such a particle only a small section of the track may be measured, and for this section the ionization per centimetre of track may be obtained. This quantity, the specific ionization, depends on the nature and pressure of the gas and is usually corrected to normal temperature and pressure. It also depends on the charge and velocity of the particle and, if the charge is known, an estimation of the velocity of the

particle may be made. The rate of change of the specific ionization with the velocity varies considerably for different velocities and, while the determination of the specific ionization may be reasonably high, the value of the velocity may be rendered inaccurate for particles with very high energies. If the momentum of the particle is measured simultaneously by magnetic curvature or range estimations, the mass and energy of the particle may be calculated. The accuracy is usually sufficiently high to identify the type of particle as, for example, in the case of the meson, although very accurate mass determinations are somewhat more difficult. A considerable amount of experimental work has been carried out to determine the specific ionization of the meson since its discovery, and also of the electron, since the variation of the ionization with the energy is of considerable theoretical importance for both these particles.

It may be stated here that, since a large amount of the ionization may be produced by secondary particles of relatively high energies arising from collisions in the gas, at least two specific ionization values may be defined. The primary specific ionization is the number of ion pairs produced by primary ionization processes per centimetre of track and is obtained by counting as a single event groups of ion pairs formed by secondary process. The total specific ionization includes all ionization resulting from secondary

processes and represents the total loss of energy of the particle in ionization processes. The former is obtained from cloud chamber measurements, and the latter from ionization chamber measurements, usually at high pressure.

The three principal methods of studying ionization of fast particles are:-

(1) Ionization chambers, using electric fields sufficiently high to ensure the collection on an electrode of all the ions formed; these are used in conjunction with an electrometer or linear amplifier for detection and measurement.

(2) Cloud chamber photographic technique, in which the expansion of the chamber is delayed to allow the ion pairs to diffuse in a weak electric field before becoming condensation nuclei for the drops. Individual drops may then be counted and the number of ions determined.

(3) Proportional counters, which employ electric field conditions in which the ions produced are multiplied by a gas collision process before their collection; these are used in conjunction with a linear amplifier in exactly the same manner as an ionization chamber.

The latter method has been applied only to the measurement of large amounts of ionization, as, for example, that produced by a proton or alpha-particle. In Part Two of this thesis a detailed account is given of investigations extending the scope of this technique to the measurement of

the ionization produced by low-energy electrons and electromagnetic radiations.

The cloud chamber method is very laborious, and, while a high degree of accuracy may be obtained in the measurement of a single track when the conditions of expansion are carefully controlled, investigations of the ionization of particles using the cloud chamber technique usually extend to relatively small numbers of observations. The method is not therefore suited to a statistical analysis of the specific ionization of a large number of particles, such as would be required, for example, for a study of the specific ionization of the meson or of high energy electrons. Such an analysis is considerably simpler if the amounts of ionization produced by individual particles are recorded electronically.

The ionization produced by individual particles may be obtained with an ionization chamber if the ions collected are made to cause a change in the voltage of the first grid of a linear amplifier. This voltage change may be converted into a pulse of suitable shape, amplified in the various stages of the amplifier, and be made to produce an output pulse sufficiently large to allow accurate measurements to be made. With an anplifier of suitable design this may be accomplished without distortion and the output pulse is directly proportional to the original voltage change. If the voltage change is also directly proportional to the number of ion

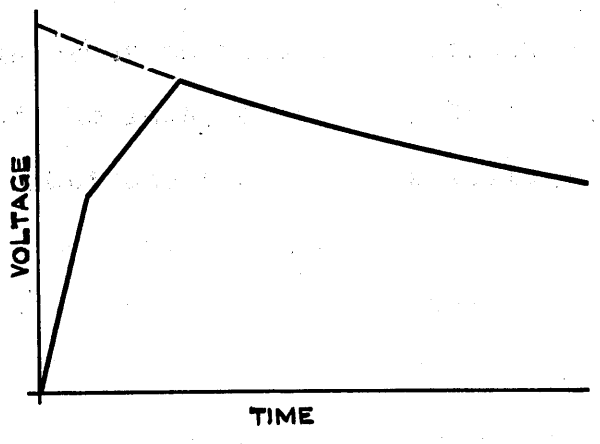
pairs formed by the particles, the system may readily be used for measurements of the ionization.

A large variety of designs have been used for ionization chambers, largely depending on the type of radiation being investigated and the method used for its study. For example, in the measurement of gamma radiation, high energy electrons and cosmic rays, a considerable number of investigations, particularly in the earlier work, were carried out in which the ionization was measured as a continuous current. The ionization chambers usually consisted of simple parallel plates, cylinders with a central collecting wire, or spherical chambers with a central probe. In each of these the current was directly proportional to the number of ions being collected, and the principal problem was to ensure that neither the positive nor the negative components were reduced by recombination with each other, or with the atoms of the gas on their passage to the electrodes. This may be effected by the use of a sufficiently high electric field strength, depending on the pressure of the gas, and the current flowing between the plates is then a direct measure of the number of ions being formed per unit time interval. This effect is also present when the chamber is being used to measure a single ionization event, but in this case other problems also arise. The conversion of the voltage change on the collecting electrode into a pulse which may be amplified on an A.C.-coupled

amplifier necessitates a consideration of the various stages of growth and decay of the voltage with time, in order that a suitable frequency band-width may be chosen for the amplifier. This time variation of the voltage depends upon three factors:-

(a) If the ionization results in the formation of electrons and positive ions, these will travel towards their respective electrodes with different velocities, and the voltage rise will consist of two portions, a steep increase when both groups are moving and a more gradual increase when only the slower component is moving. Thus, while the voltage change finally resulting from a single ionization event is independent of the position of formation in the active volume, the relative sizes of the two portions depend on the relative distances of the position of formation from the two electrodes.

(b) The rate of change of voltage at the collecting electrode is proportional to the strength of the field through which the ions are travelling and the voltage rise in each portion will therefore depend on the configuration of the electrodes; thus, for a parallel plate ionization chamber, the two portions would produce voltage changes of the type shown in the sketch below, whereas, for a cylindrical chamber, the increases would both be non-linear.

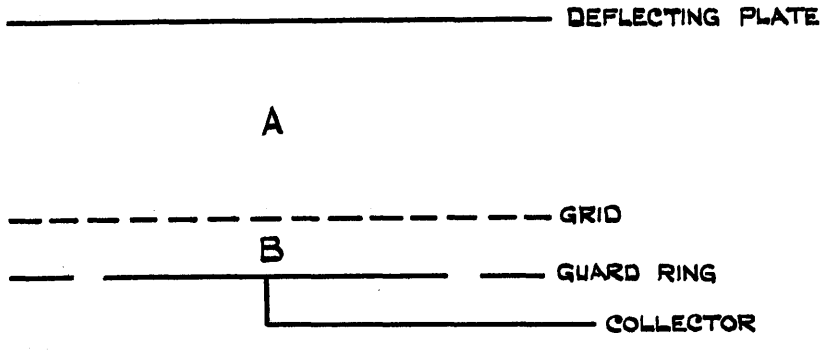


(c) The voltage will decrease when the charge has been collected, the rate at which the decrease takes place being determined by the leakage resistance and the capacity of the collecting electrode; this rate must be sufficiently low to allow the voltage to rise to very close to the full extent before the decay is appreciable.

It is normal to allow both groups of ions to reach the electrodes, and the frequency response of the amplifier is set to cover the whole time required for the collection. This is frequently much lower than is desired since the time constant of such an amplifier must be greater than about 10 milliseconds. This time constant may be reduced considerably if only the electrons are to be collected, since electron drift velocities of the order of 10^6 cm/sec may readily be obtained in some gases.(1) This would result in the measurement of only the initial steep rise shown in the sketch, and would not

therefore give a constant pulse size for every ionization event involving the same number of ions.

In order to reduce the effect of the position of formation of the ions, a grid may be interposed between the collecting electrode and the high voltage plate, as shown in the sketch below. This will at the same time minimise the difficulties associated with the measurement of tracks which are not parallel to the electrodes.



The ionization events are confined to the region A and the function of the grid is to screen the collecting electrode from any field changes arising in the area of formation. The voltage rise on the collecting electrode results almost entirely from the passage of the electrons across the region B. If the grid can successfully be made to provide a screen between the two regions, and if no electrons are lost in their passage through the grid, the voltage change will be directly proportional to the number of ions formed, and will possess a very rapid rise time. This pulse may then be amplified in an amplifier with a high frequency

response, overcoming the difficulties associated with microphony in the chamber. It was hoped that such an amplifier could be made to have a much lower noise level than a corresponding low-frequency type.

An ionization chamber of this type was built and an investigation of the ionization produced by alpha particles of polonium (radium F) was carried out at reduced pressures. The variation of output pulse size with the voltages of the electrodes and the pressure of the gas was measured, the number of ions produced being calculated from the Bragg curve and a knowledge of the section of the track being investigated. The widths of the groups were also measured and the resolution of the instrument obtained. The results of the investigations showed that the pulse size was directly proportional to the number of ion pairs produced, the measurements extending from 1.65×10^5 ions down to 1.02×10^4 ions, which represented the minimum number which could be detected with accuracy above the noise level. The average resolution, represented as the full width at half height of the group, depended on the scale setting of the amplifier; on the maximum gain setting it corresponded to 3,000 ion pairs, which was almost identical with the mean square value of the noise.

A summary of the conclusions is given at the end of this part of the thesis. It was considered that the facilities afforded by the development of the proportional

counter methods were much superior to those afforded by a pressurised ionization chamber for investigation of small amounts of ionization. Proportional counters also permit the measurement of very low energy radiations in studies which are of greater interest.

(1) The Kinetic Theory of Gases - Loeb.

SECTION 2

PREVIOUS WORK.

The Measurement of Single Ionization Events.

A number of studies have been made of the ionization produced by alpha particles and protons using pressurised chambers. In many of these the effect has been measured as a continuous current, and attention has been directed principally to the efficiency of collection as a function of voltage and pressure. In several more recent experiments the ionization produced by single particles has been measured, and these measurements have normally involved the use of an amplifier with a sufficiently long time-constant to allow the collection of both positive and negative ions. A summary of the results has been given in a recent paper by Corson and Wilson[2].

Kema and Barschall[3] investigated the saturation characteristics of a parallel plate ionization chamber for various gases. They used a balanced circuit for measuring the ionization produced by the alpha particles of polonium, investigating the mean current as a function of pressure and field strength. The results showed that the elimination of gases which readily form negative ions allowed very small fields to be used, and that the mobilities of the negative carriers were very much higher in these gases. The curves for pure argon

showed that 100 percent collection could be obtained with field strengths of the order of 10 volts per cm. per atmosphere. They point out that the Jaffe theory of columnar ionization would not be applicable to this case, since the diffusion will be very rapid and the effect of columnar formation almost negligible.

Barschall and Kanner[4] describe the results of experiments to determine the scattering distribution of neutrons in hydrogen and other gases. A parallel plate ionization chamber was used, and the size distribution of the individual pulses measured. Pressures up to 9 atmospheres were used and the collecting field gradient was 3750 volts per cm. Calibration with alpha particles from polonium showed that the spread introduced by the chamber was less than 5 percent in either direction. Since the pulse size is independent of the position of formation of the ions, the time constant of the amplifier being equal to 0.02 sec., this spread must be due almost entirely to amplifier noise.

Koontz and Hall[6,7] describe the results of a set of experiments using a cylindrical chamber which may also be operated as a proportional counter. Since the amplifier had a shorter time-constant than in the previous experiments the pulse size depended on the position of formation of the ions. An assymmetrical distribution of pulses is obtained when the chamber is used without gas amplification, but this disappears

almost entirely when gas amplification is used. In the latter case the greater proportion of the electrons measured are formed very close to the collecting electrode. The width obtained with the proportional counter was therefore less, and, for the 0.6 Mev protons from the n,p reaction in nitrogen, the spread corresponded to a value of 35 kev.

Measurements of the specific ionization of fast mesons and electrons have normally been carried out by the continuous current method and a large number of such investigations have been reported. These investigations, of which the results of Clay[7] and Broxon and Meredith[8] are typical, have been directed mainly to a study of the saturation characteristics at high pressures. Two studies in which individual particles were measured have, however, been reported.

Swann[9] measured the specific ionization of single cosmic ray particles with a cylindrical chamber 30 cm. in length. This could be pressurised to 10 atmospheres, and a voltage of 1100 applied to the case. It was claimed that the collection time was less than 0.05 sec. and an amplifier with a long time-constant was used. Curves are shown with values as low as 6,000 ion pairs, indicating some degree of accuracy in this region.

Dunlap[10] repeated this experiment with a parallel plate ionization chamber using a track length of 17 cm. The chamber was pressurised to 27 atmospheres and a field of 1900

volts per cm. applied. The collection time was of the same order as the time constant of the amplifier, 0.01 sec., and corrections were applied for this fact. The results indicated a specific ionization of 67 ion pairs per cm. in argon, but the spread of the values was considerable. This may be due to the energy spread of the particles, but uncertainties are introduced by the close proximity of the collection time and the time-constant of the amplifier.

The Use of Gridded Ionization Chambers.

Bunemann, Cranshaw and Harvey[11] have described the results of an investigation of the properties of an ionization chamber having a grid between the collecting plate and the deflecting plate. As already pointed out, the function of this grid is to screen the collecting plate from any field changes in the region between the grid and the deflecting plate, and at the same time to permit the passage of the electrons to the collector. If this is possible a linear amplifier of high frequency response may be used, the pulse obtained being formed solely by electrons.

These authors also give the results of a theoretical analysis of the effect of the grid. In this they assume that the grid consists of a set of parallel wires with radius r spaced a distance d apart, and that the spacing between the grid and the collector is p. They define the inefficiency of screening, s, as dE_B/dE_A where E_B is the field between the

grid and collector and E_A is the field in the collecting volume. They find that s is very closely equal to $d/2\pi p.\log d/2\pi r$; this quantity must be as small as possible if the pulse size is to be independent of the position of formation of the charge. The condition that all lines of force leaving the plate should by-pass the grid and reach the collector is:-

$$E_B/E_A > (1 + 2\pi r/d)/(1 - 2\pi r/d).$$

If this condition is observed no electrons should be lost by capture on the grid.

These conclusions were tested with a number of grids and the results found to be in agreement with the experiments. An investigation of the alpha particles of polonium (5.3 Mev) showed a full width at half height corresponding to 60 kev. The majority of this spread was attributed to the amplifier noise.

In the work described in the following sections a grid was used in which the wire diameter was 0.005 in. (No. 36 S.W.G.) and the spacing 0.05 in. The distance between the grid and collecting plate was 0.67 cm. The value of s calculated for this grid was 0.04 and the condition that all lines by-pass the grid was that E_B/E_A should be greater than 2.0. The latter value may be compared with the experimental ratio of 1.8 : 1.

(2) Corson and Wilson, Rev. Sci, Insts., 19, 207, (1948)

(3) Kema and Barschall, Phys. Rev., 63, 18, (1943)

(4) Barschall and Kanner, Phys. Rev., 58, 590, (1940)

(5) Koontz and Hall, Rev. Sci, Insts., 18, 643, (1947)

(6) Koontz and Hall, Phys. Rev., 72, 196, (1947)

(7) Clay, Phys. Rev., 52, 143, (1937)

(8) Broxon and Meredith, Phys. Rev., 55, 883, (1939)

(9) Swann, Phys. Rev., 44, 961 (1933)

(10) Dunlap, Phys. Rev., 67, 67, (1945)

(11) Bunemann et al., unpublished communication.

SECTION 3.

DESCRIPTION OF APPARATUS.

The method consisted of the use of an ionization chamber with intermediate grid electrode in conjunction with a linear amplifier of fairly high frequency response. The ionization chamber was made sufficiently robust to stand pressures up to 30 atmospheres. A variable-bias discriminating circuit was used to analyse the pulses.

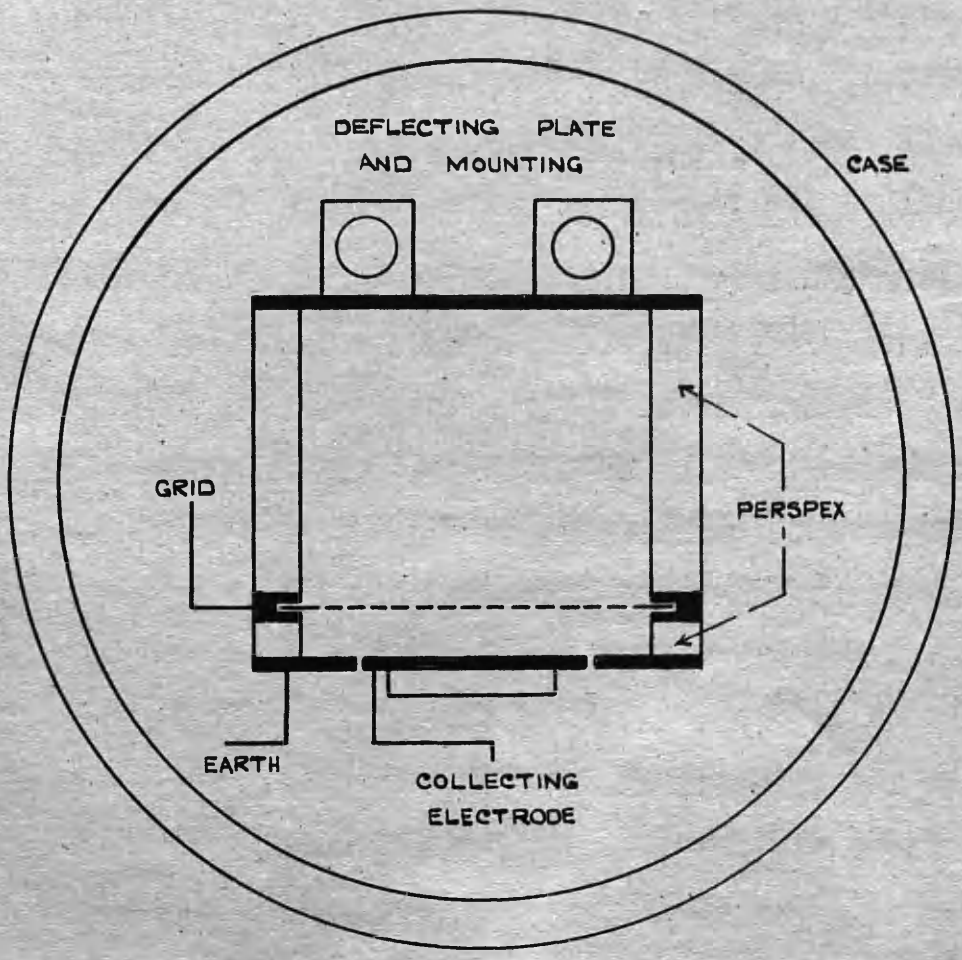
The characteristics of the chamber were measured at reduced pressures using an alpha particle source. In each individual study the integral distribution of the pulses was obtained by counting with a scaling unit the numbers of pulses with amplitudes greater than the bias settings. The pulse size distribution was then obtained by differentiation, and the average pulse size and the full width at half height calculated. Various voltages were applied to the deflecting plate and the grid for the various pressures, and the variation of pulse size with these quantities observed.

The Ionization Chamber.

A sectional diagram of the ionization chamber is given in Figure 1. The electrode system consisted of a collecting plate 3 cm. broad and 21 cm. in length surrounded by an earthed guard-ring 1.5 cm. in width. These two electrodes were separated by a gap of 1 mm. and held together

FIG. 1

SECTIONAL DIAGRAM OF IONIZATION CHAMBER



by a strip of perspex running under the assembly. This strip was recessed near the gap in order to increase the insulation between the two electrodes. The grid and the deflecting plate were mounted on the guard-ring by means of perspex spacers extending over the whole length of the assembly. The grid consisted of a mesh of nichrome wires 0.005 in. in diameter spaced 0.05 in. apart; it was mounted on a light frame located in a metal slot at a distance of 6.7 mm. from the collecting electrode. The deflecting electrode was also mounted on perspex, and two copper rods attached to this electrode provided a means of supporting the whole assembly from the end-plate of the outer case. These rods were led out through the case by bonded metal-to-rubber seals, thus providing a vacuum-tight insulated mounting. The whole electrode system consisted of a box 6 cm. x 6 cm. in cross section and 25 cm. in length. Copper-to-perspex seals were effected by cement, which was baked to hardness after assembly.

The leads to the grid and the collecting electrode were introduced through the end-plate by means of co-var seals. Adequate insulation was obtained with these. The guard-ring was earthed internally to the case of the chamber.

The case consisted of a cylinder 5.5 in. outer diameter and 13 in. in length, with a wall thickness of 0.25 in. The end-plates were 0.75 in. thick and were attached to flanges of the same thickness on the main case. The vacuum seal was

provided by two gaskets of rubber 0.25 in. x 0.25 in. in cross-section.

Source.

The polonium source used in these experiments was separated from an equilibrium mixture of radium D, E and F in solution in the following manner. The solution was evaporated and the residue re-dissolved in 0.1 N hydrochloric acid. A silver disc 0.6 in. diameter was rotated in the solution and polonium deposited on it. The activity of this source appeared from absorption measurements to be entirely free from any beta-activity.

The source was mounted from the end-plate of the chamber opposite the electrode assembly. It was covered by a collimator consisting of a large number of holes 0.03 in. in diameter drilled in a plate 0.25 in. thick. This limited the ionization produced by all but a small number of the particles to the active volume of the chamber.

With this source and collimator a total activity of 1,700 counts per minute was obtained.

The Linear Amplifier.

With the exception of the scaler the electronic equipment used in this experiment was of Montreal design. The head amplifier, the main amplifier and the discriminating unit were considerably modified to suit the requirements of

this experiment. In the final form the head amplifier consisted of one stage of amplification and a cathode-follower. The first valve, a 6 AK 5 pentode, was triode-connected and drew a few milliamps of current. The grid of this valve was connected to the collecting electrode, no grid resistance being used, and the input capacity of these was 35 picrofarads. The cathode follower was provided with a number of cathode resistances in series, allowing several gain settings of the amplifier, each approximately one-half of the previous setting.

Six stages of amplification followed in the main amplifier, each stage being resistance-capacity coupled with a small amount of negative feed-back. The time-constants of these stages were 0.7 milliseconds, except for the first stage, which had a time-constant of 3 microseconds. This stage also had a high-frequency by-pass in the anode circuit with a time constant slightly longer than that of the grid circuit. The output stage of the amplifier was a cathode follower capable of developing voltages between 100 and -100. The valves in the head amplifier and the first three valves in the main amplifier were heated by accumulators, the remainder being heated by a 6.3-volt A.C. supply.

The overall gain of the amplifier at maximum setting was 2.7×10^6; the frequency response curve is given in Figure 2. The maximum noise level was 25 microvolts, corresponding to

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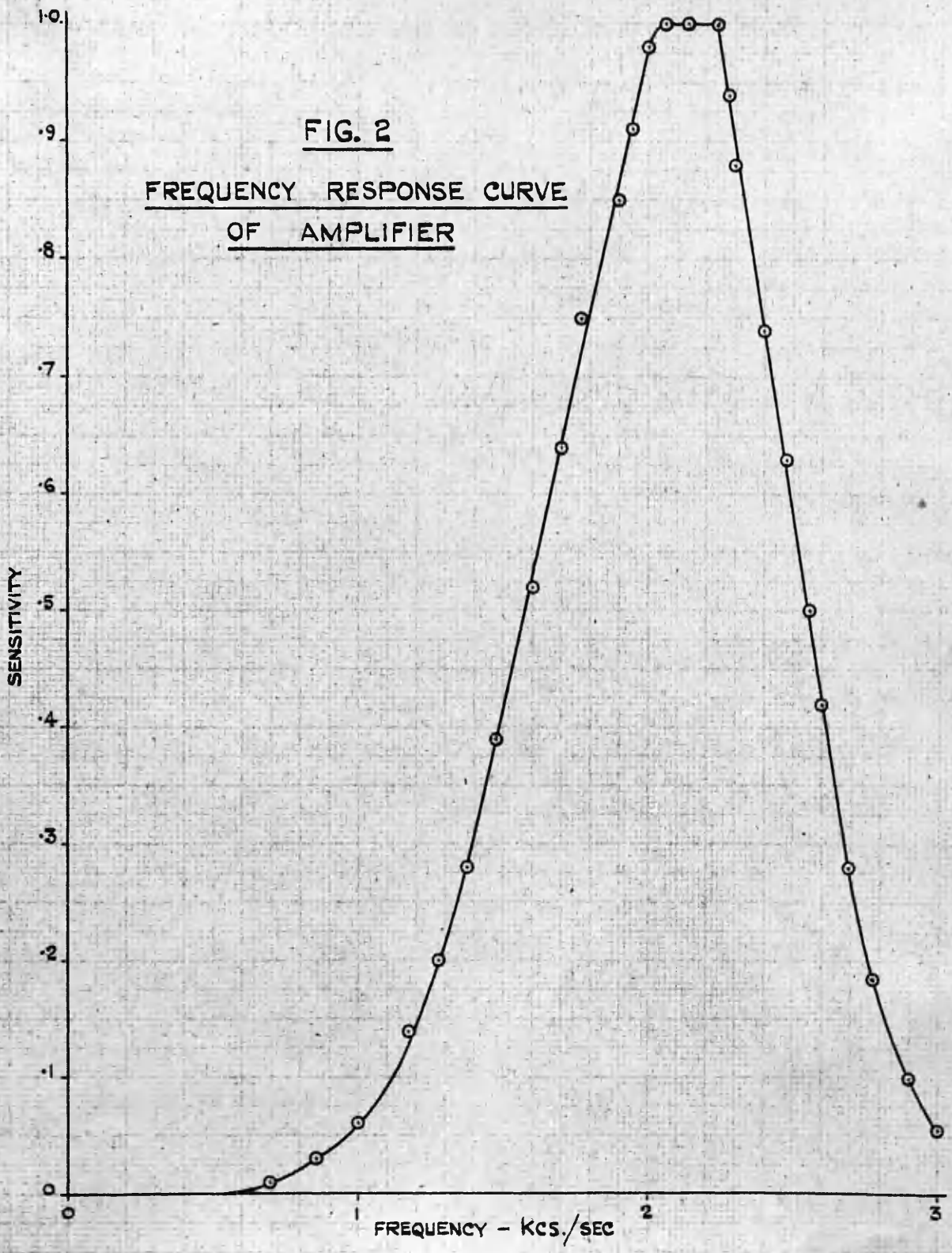
FIG. 2

FREQUENCY RESPONSE CURVE OF AMPLIFIER



about 6,000 ion pairs, and the root mean square noise level 8 microvolts, corresponding to about 2,000 ion pairs.

The Discriminator.

A circuit diagram of the discriminator is given in Figure 3. It consisted of two 6 AB 7 television pentodes with a common cathode load of 10,000 ohms, these valves being chosen for their short grid-base. Stabilised supplies of 150 and -150 volts were used, and the common cathode potential was 11 volts above earth. With no pulses incident on the circuit the left hand valve was cut off and the other was drawing the full current required to maintain the cathode at the required potential. A positive pulse arriving at the grid of the first valve caused it to conduct, and the drop in voltage on its anode was used to cut off the second valve and divert all the current through it. After the removal of the positive pulse the circuit returned to its normal position. The arrival of a pulse of sufficient magnitude to open the first valve therefore resulted in a large voltage change on the second valve, and this voltage could readily be used to operate a scaling unit or oscilloscope. The resistance-capacity network of the first valve had a time constant of 0.05 seconds, which was sufficiently long to prevent spurious counting of double pulses.

A negative bias voltage between 0 and 100 could be applied by means of the potentiometer chain across the whole

TO FOLLOW PAGE 21

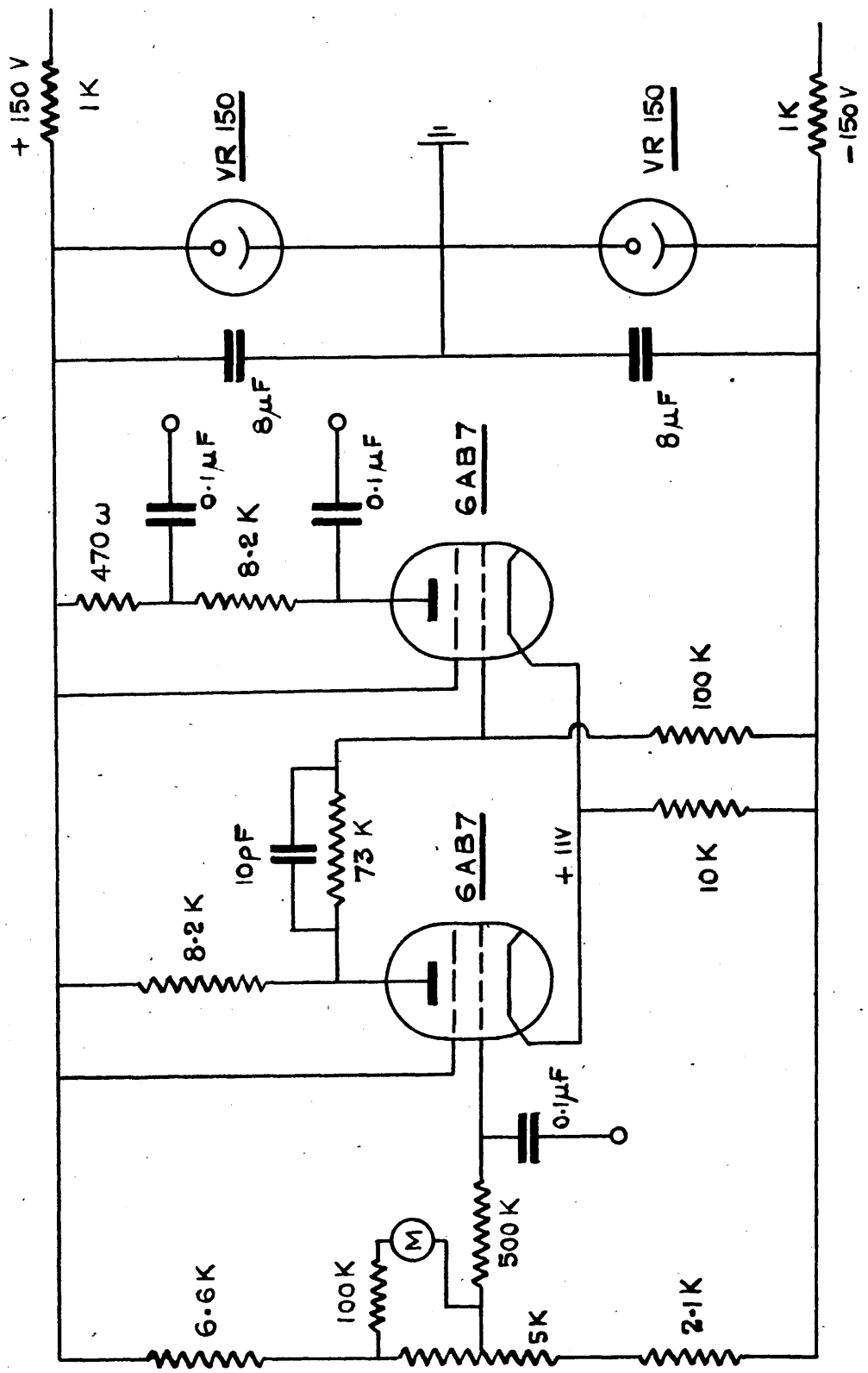


FIG. 3

CIRCUIT DIAGRAM OF DISCRIMINATOR

stabilised high voltage supply. This bias was registered on a meter which could be read to 0.5 volts. The bias setting could be reproduced to a satisfactory degree of accuracy for the experiments described below.

Power Supplies.

The high-voltage supplies for the amplifier and discriminator were of conventional design and were stabilised by neon tubes. No measurable gain change was observed when the mains voltage was varied by 10 volts.

The high-voltage supplies for the grid and deflecting plate of the chamber were obtained from a specially designed 2-kilovolt rectifier unit; this unit incorporated stabilisers providing independence of the current and voltage being used, and the various voltages used in these experiments were obtained from a potentiometer unit attached to the output.

The high-voltage leads and the connections between the amplifier and the ionization chamber were carefully screened to prevent interference from external radiations of high frequency.

SECTION 4.

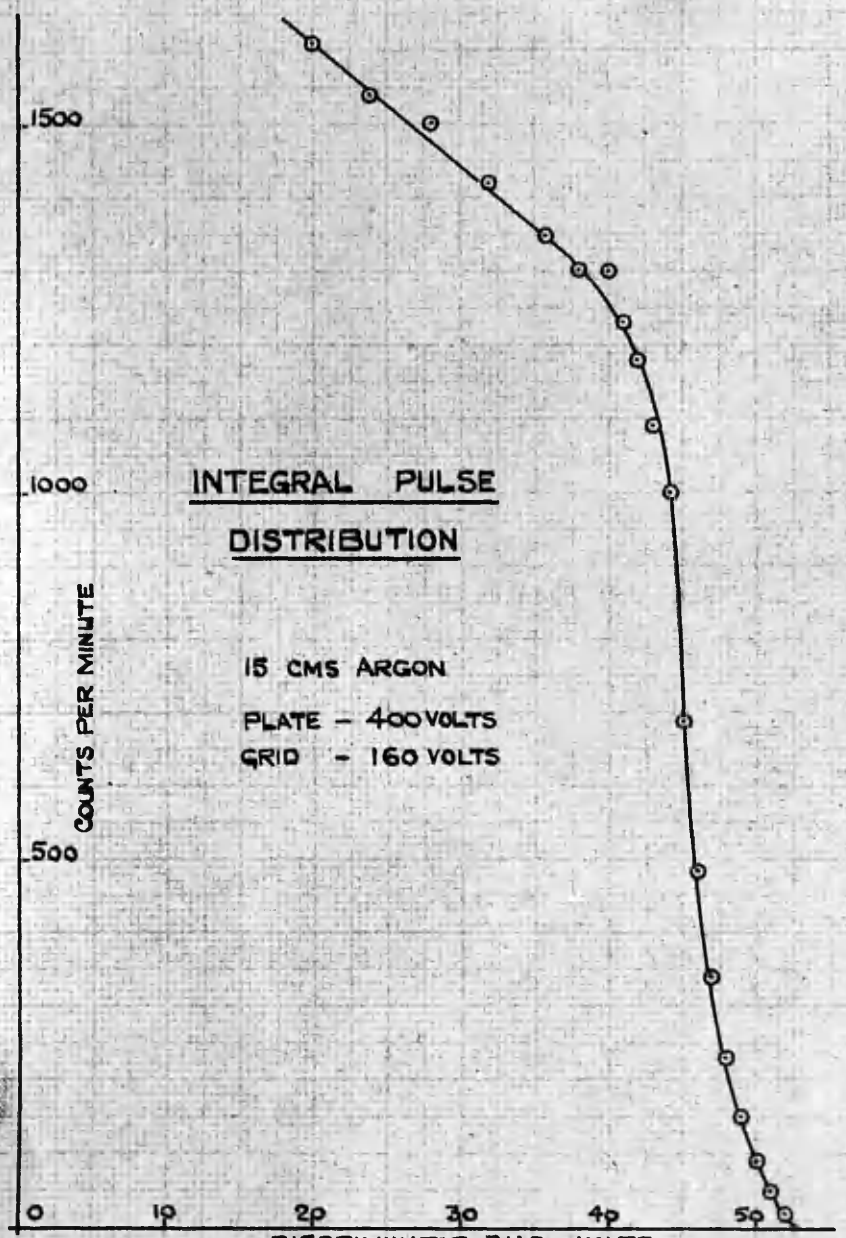
EXPERIMENTAL RESULTS.

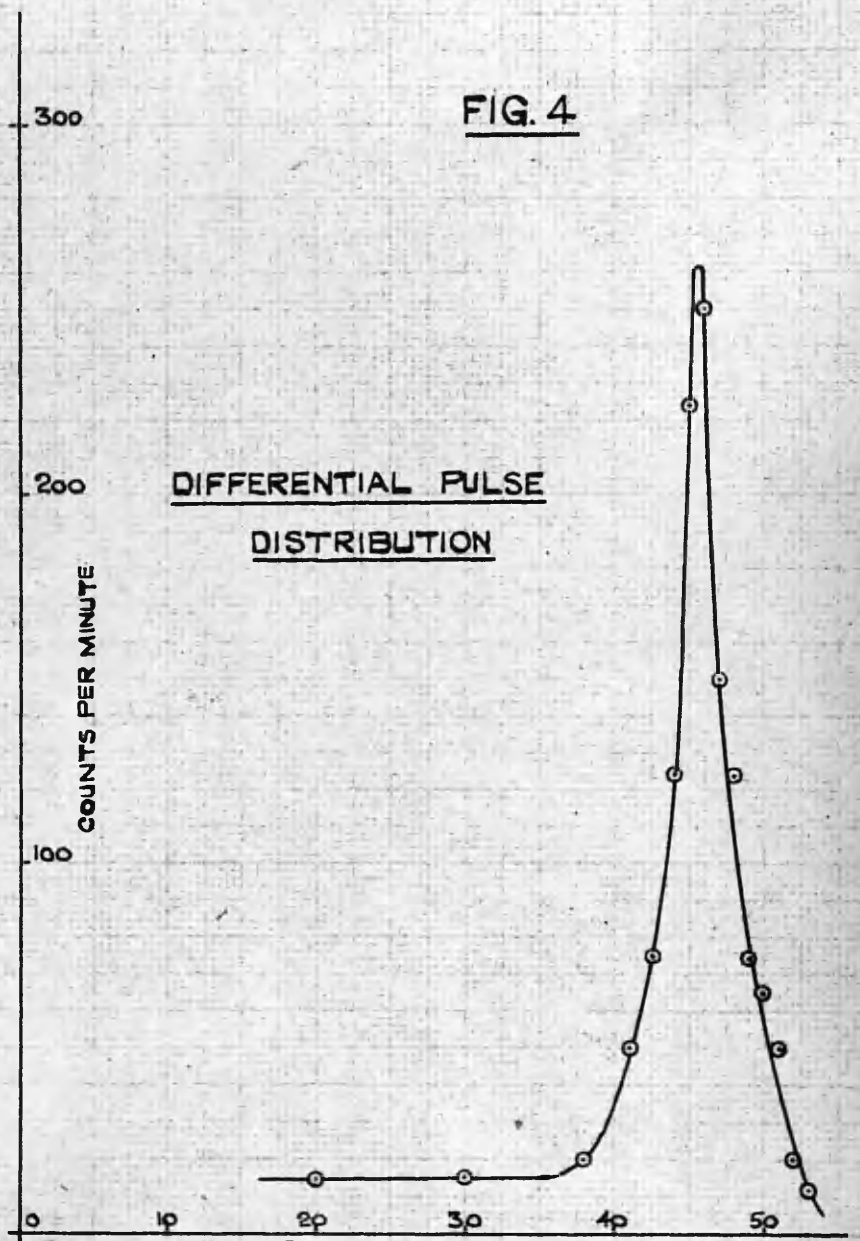
Three series of investigations were carried out with the arrangement described in the previous sections. In the first of these the ionization chamber was filled with argon at a pressure of 15 cm. Hg, and the pulse distributions obtained for a large number of settings of the voltages of the grid and deflecting electrode. The average pulse size was then calculated and related to the values of these voltages. This investigation was later extended to lower pressures in order to obtain the best working conditions for the second series of studies. The latter consisted of the measurement of the average pulse sizes obtained at various pressures using a fixed ratio of the voltages on the grid and deflector; these sizes were correlated with the number of ion pairs calculated from the Bragg curve, and a calibration of the instrument thereby obtained. The final studies consisted of the measurement of the collection efficiency of the chamber as a function of voltage for argon, nitrogen and air.

A. Variation of the Average Pulse Size with the Voltage of the Plate and the Grid.

The first measurements were carried out at a pressure of 15 cms. of argon; a typical integral distribution is shown in Figure 4, together with the pulse distribution obtained

FIG. 4





TO FOLLOW PAGE 23

by differentiation. In the first set of runs the voltage of deflecting plate was set to 400 volts and the grid potential was varied from 0 to 300 volts in steps of 20 volts. The average pulse size was determined for each setting and the curve shown in Figure 5 was obtained. The collection efficiencies were calculated on the assumption that the maximum value corresponded to 100 percent collection; this assumption appears to be justified by the consistency of the pulse sizes obtained for the various settings. This process was repeated for a number of other plate voltages between 50 volts and 800 volts, and three of these curves are shown in Figure 5. The results of a similar set of experiments carried out at a pressure of 2 cms. are given in Figure 6. The plate voltage for each curve is given, and the results have been plotted in terms of the ratio of the grid voltage to the plate voltage. While the number of points on these curves is somewhat less than that for the 400 volt curve, the variation of collection efficiency with grid potential is clearly represented.

The salient features of these curves may be described as follows. Firstly, for each curve the efficiency of collection increases rapidly with the grid voltage, up to a value of between 0.2 and 0.3 times the plate voltage. This is clearly due to the gradual decrease of the number of lines of force ending on the grid as its voltage is raised. Secondly, when the voltage ratio becomes equal to, or greater than 0.3,

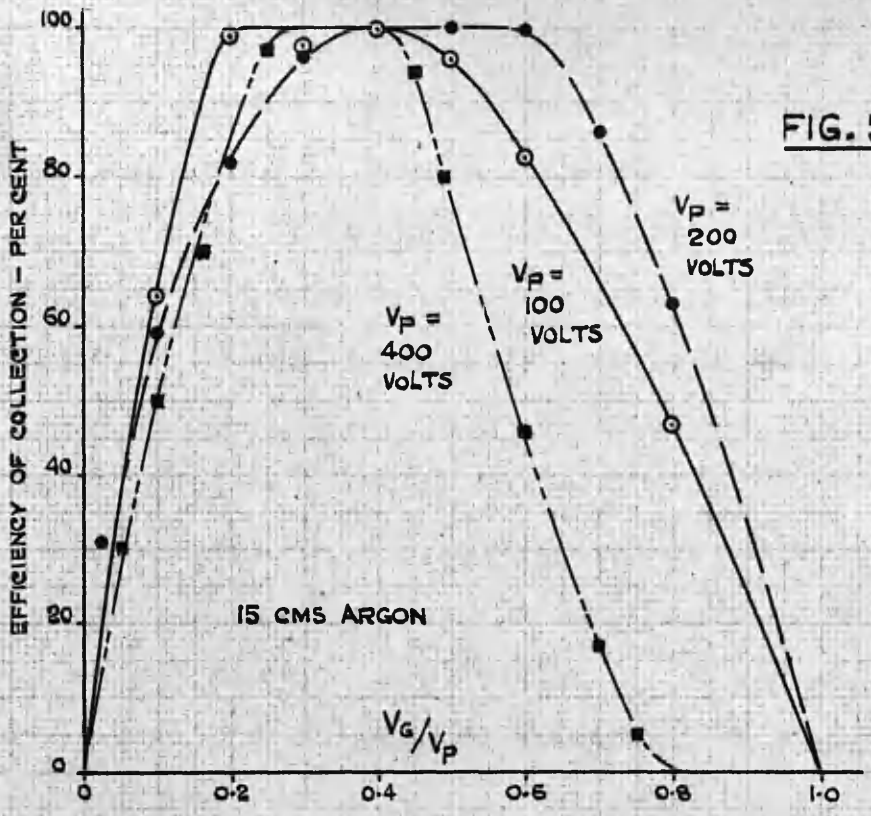

EFFICIENCY OF COLLECTION – PER CENT
100
80
60
40
20
0
0
0·2
0·4
0·6
0·8
1·0
V_G/V_P
V_P = 400 VOLTS
V_P = 100 VOLTS
V_P = 200 VOLTS
15 CMS ARGON

FIG. 5

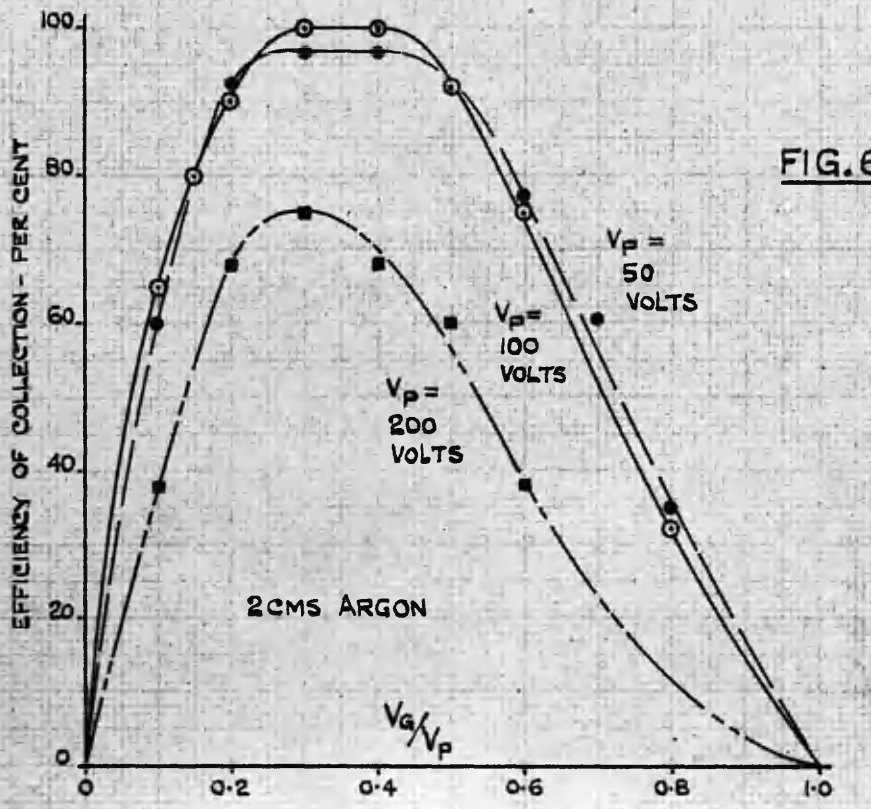

EFFICIENCY OF COLLECTION - PER CENT
100
80
60
40
20
0
0
0·2
0·4
0·6
0·8
1·0
V_G/V_P
V_P = 50 VOLTS
V_P = 100 VOLTS
V_P = 200 VOLTS
2CMS ARGON

FIG. 6

that is, when the field ratio E_g/E_p is equal to 1.8, the efficiency reaches a maximum value. If the field in the collecting volume is sufficiently high to overcome the recombination effect, this value will be 100 percent; as the grid voltage is raised further the collecting field is gradually reduced, and the efficiency will fall to zero. In the majority of the curves there is a reasonably broad plateau between these two phases.

If the overall voltage is not sufficiently high to overcome recombination, the maximum number of ions measured will be less than the number formed. A more unexpected effect giving rise to a decrease in pulse size is observed at higher plate voltages. Examples of this are given in the 400-volt curve of Figure 5 and the 200-volt curve of Figure 6. Here the pulse size decreases rapidly for grid voltages higher than 160 at the higher pressure and 50 at the lower pressure; a similar decrease appeared at higher voltages in each case. It appears that this effect can only be explained in terms of the velocity of the electrons passing across the space between the grid and the collecting electrode. If this velocity is very high the rise time of the pulse may be so short that it is decreased in amplitude by the high-frequency filter in the amplifier. From a consideration of the frequency response curve of the amplifier a pulse with a rise time of 2.5×10^{-6} sec. would be reduced to approximately one-half size, and it is

calculated that the electron velocities in the upper part of the 400-volt curve are of the order of 2 - 5 x 10^5 cm/sec.

The results of these studies showed that maximum efficiencies of collection could be obtained with grid voltages between 0.3 and 0.5 times the plate voltage in the majority of cases, and that the plate voltage should be of the order of 25 volts for every centimetre of pressure used.

B. Variation of Pulse Size with Pressure.

The results of the experiments described in the previous section showed that the maximum loss of ions for a grid voltage to plate voltage ratio of 0.3 would be 4 or 5 per-cent of the number collected, provided that the plate voltage was not unduly high or low for operation at the pressures chosen. An investigation was therefore carried out in which the pressure of argon in the chamber was varied between 1 cm. Hg and 20 cm. Hg, and the average pulse size was measured at each pressure. Throughout this investigation the grid voltage to plate voltage ratio was maintained at 0.3, and the plate voltage was increased by 25 volts for each centimetre of pressure added.

Typical curves showing the pulse distributions obtained at pressures of 4 cm., 6 cm. and 8 cm. are given in Figure 7. These were obtained with the gain of the amplifier equal to one half of its maximum value, and the spread of the three peaks is almost constant. The value of the full width

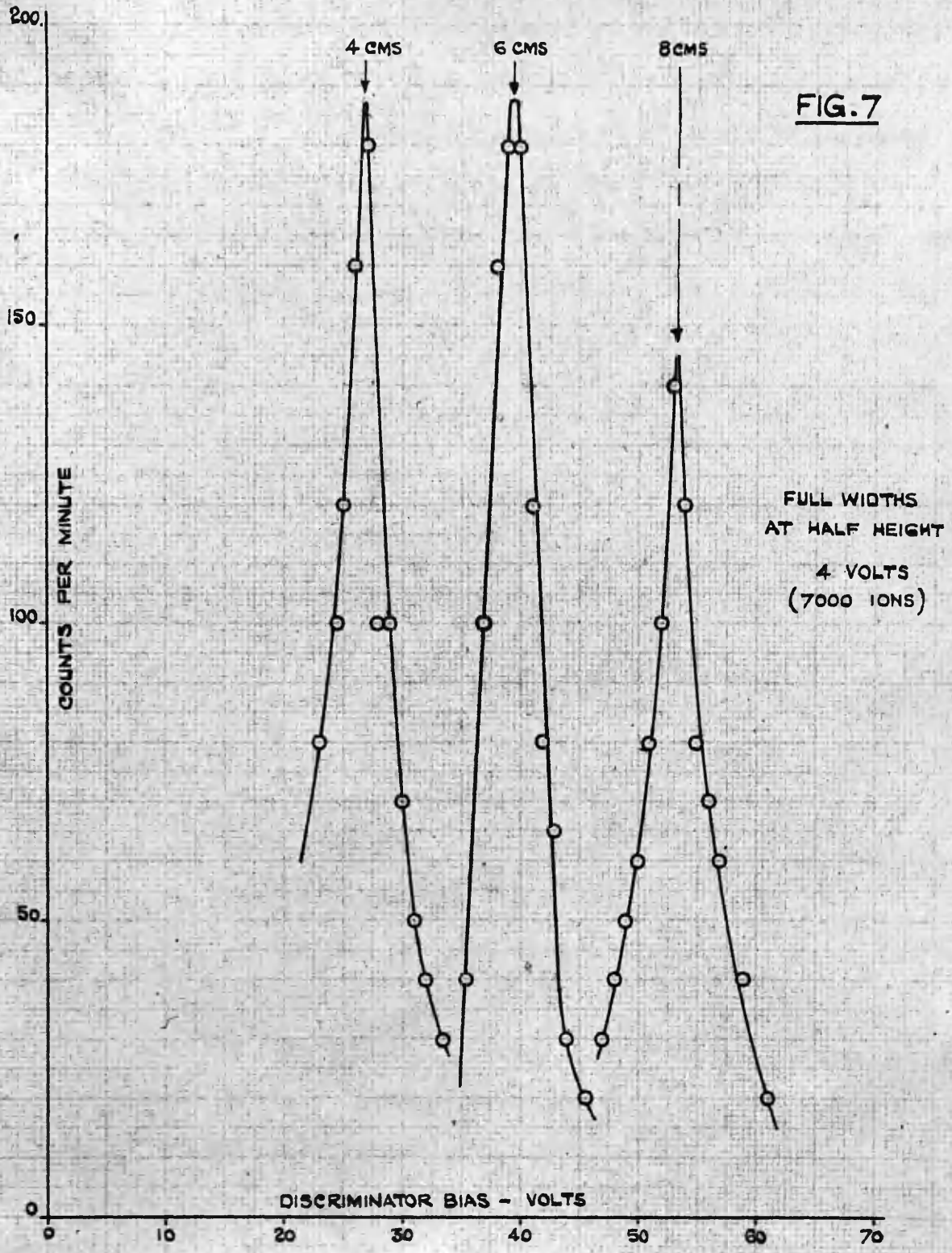


FIG. 7

FULL WIDTHS
AT HALF HEIGHT

4 VOLTS
(7000 IONS)

at half height is equal to 4 volts, and this was found by calculation to correspond to a spread of 7,000 ion pairs, that is, an energy of 200 kev. For full amplifier gain an analysis of the curves for lower pressures showed a spread also equal to 4 volts; this corresponded to an uncertainty of 3,000 ion pairs, that is, an energy of about 100 kev. For a gain setting of one quarter the spread was between 3 and 3.5 volts, and the uncertainty corresponded to 12,000 ion pairs. These results indicated that the major part of the spread was arising from the noise in the amplifier and uncertainties in the setting of the discriminator. Only a very small part could be attributed to the spread introduced by variations in the position of formation of the ions in the chamber.

The average pulse size is plotted as a function of the pressure in Figure 8. The curve increases with increasing slope to the point where the tracks of the particles end in the active volume. The pressure at which this occurs is 11.5 cm., and agrees well with the range of these particles. For pressures greater than this the pulse size falls as an increasing proportion of the ions are formed in the space between the source and the electrodes.

The average pulse size is plotted as a function of the number of ions formed in Figure 9. The latter quantity has been calculated from the Bragg curve by considering the number of ions produced in the track length falling within the active

FIG. 8

VARIATION OF PULSE SIZE WITH PRESSURE

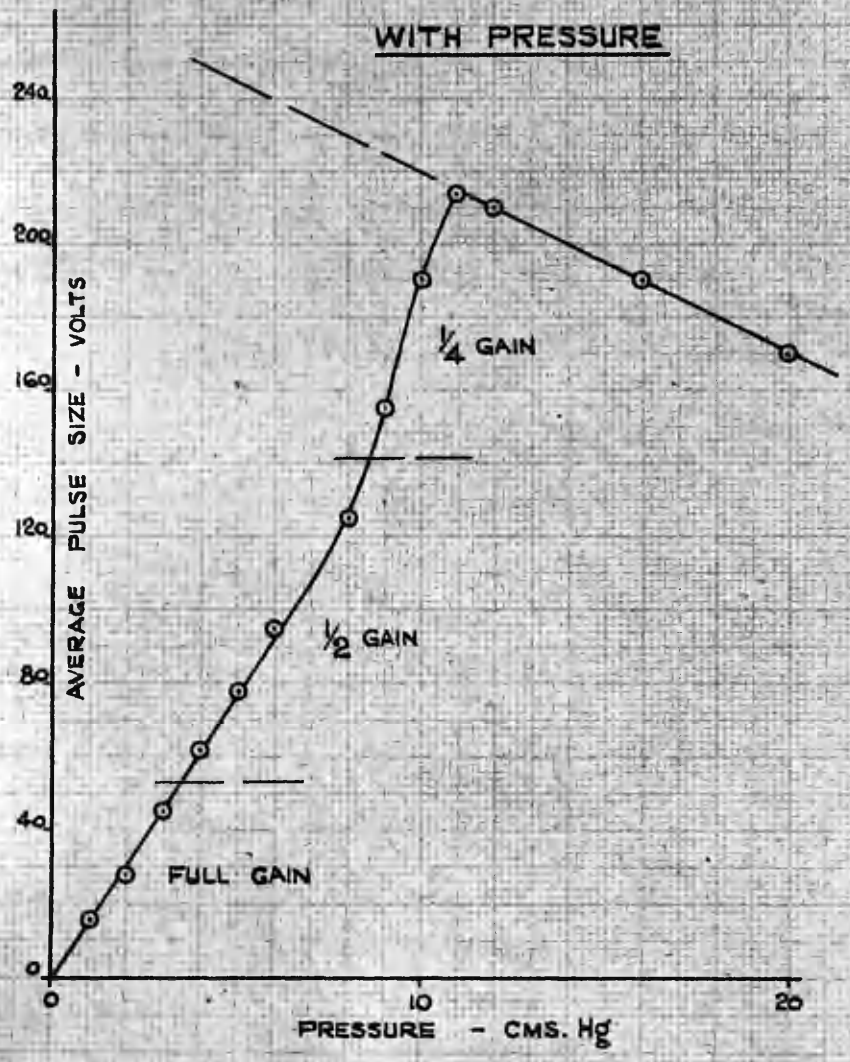
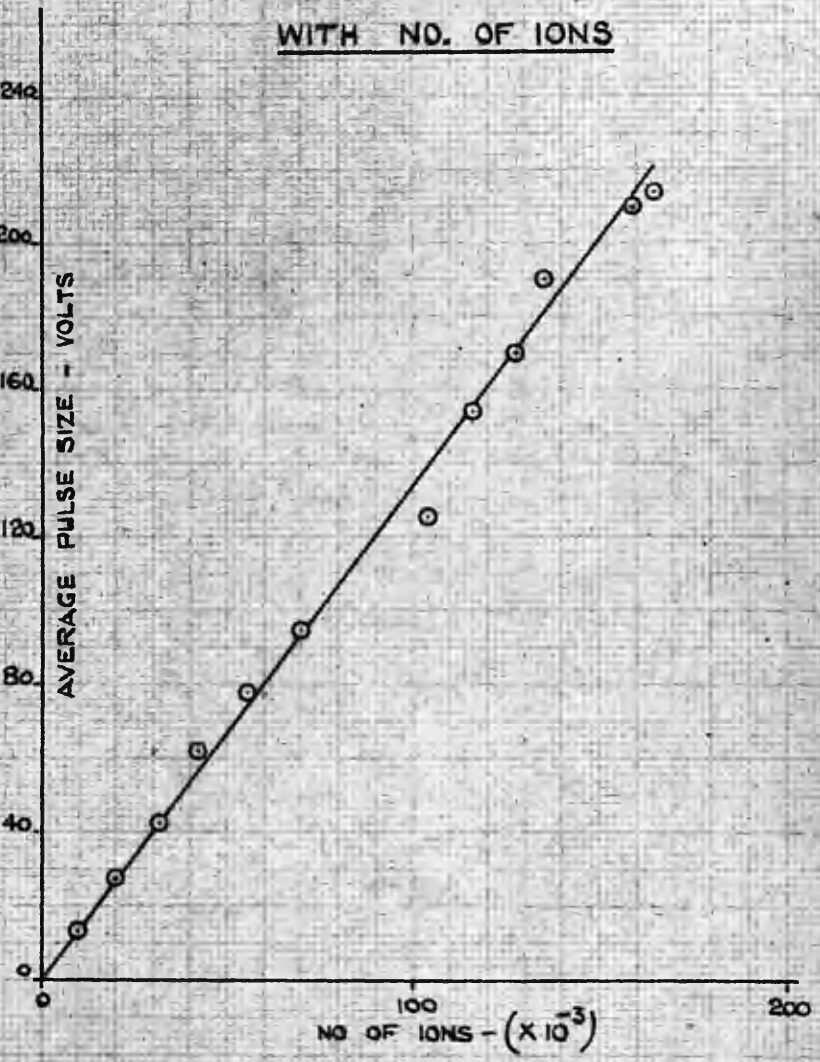


FIG. 9

VARIATION OF PULSE SIZE WITH NO. OF IONS



TO FOLLOW PAGE 27

volume of the chamber. This curve shows that the pulse size is directly proportional to the number of ions, the accuracy of each point being better than ten percent.

C. Variation of Collection Efficiency with Voltage.

A short survey was made of the variation of pulse size with plate voltage for argon, nitrogen and air; in these experiments the grid voltage to plate voltage ratio was again maintained at a constant value of 0.3. For each gas the efficiency of collection was calculated, in the case of air the absolute values of the efficiencies being calculated from the observed size of the pulse at maximum voltage and the number of ions being formed. The results, which are given in Figure 10, show that for both nitrogen at a pressure of 2 cm. and argon at a pressure of 20 cm. the efficiency of collection is very close to 100 percent for a plate potential of 50 volts. This corresponds to a collecting field of 35/4 volts per centimetre. Almost complete collection may therefore be obtained for fields greater than 33 volts per cm. per atmosphere in argon, and fields greater than 330 volts per cm. per atmosphere in nitrogen.

The difficulties associated with the efficiency of collection in air are well illustrated by the results given in Figure 10. From these it is clear that fields considerably greater than 1500 volts per cm. per atmosphere would be necessary to ensure collection efficiencies approaching 100 percent.

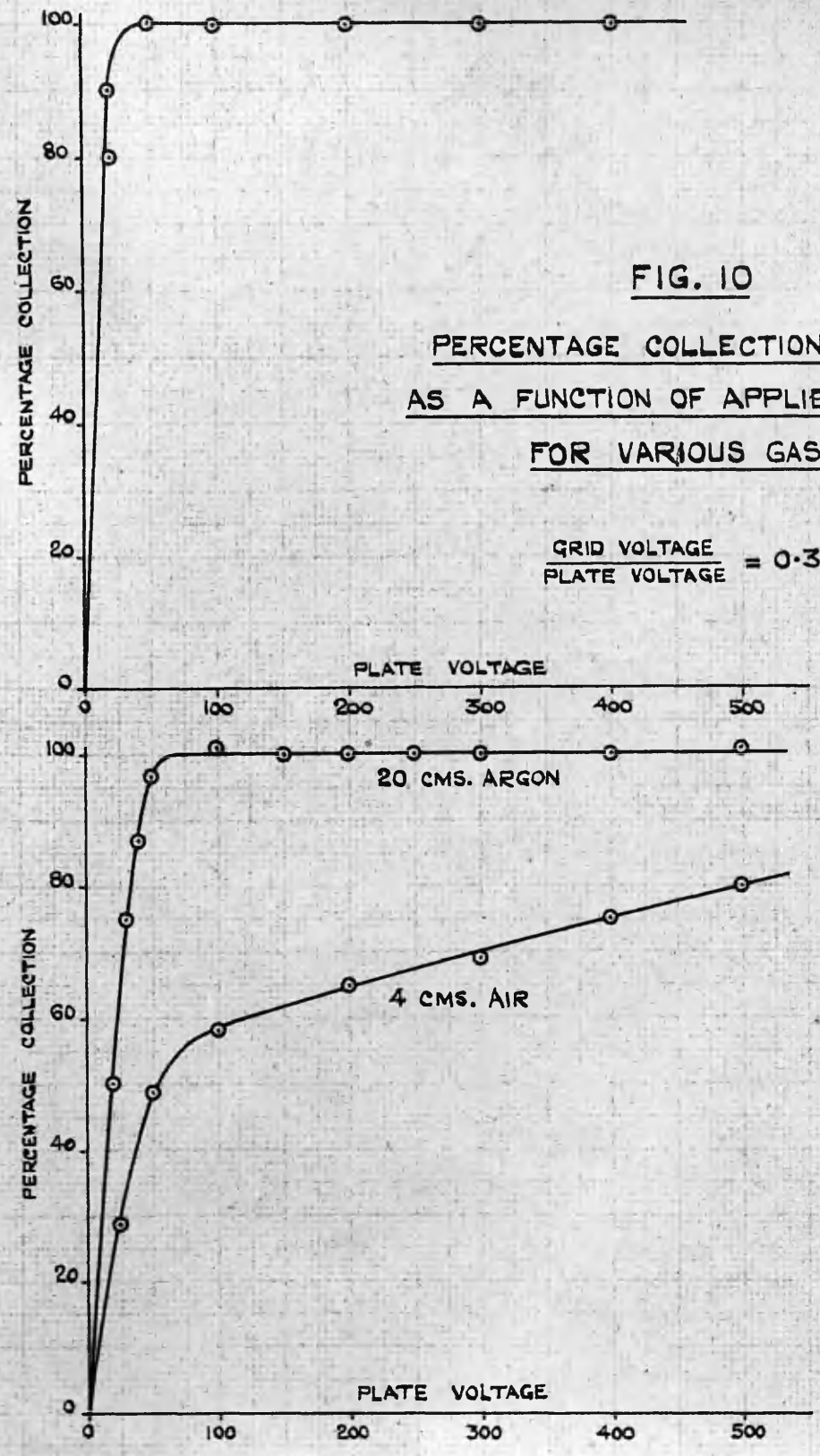


FIG. 10

PERCENTAGE COLLECTION OF IONS AS A FUNCTION OF APPLIED VOLTAGE FOR VARIOUS GASES

$\frac{\text{GRID VOLTAGE}}{\text{PLATE VOLTAGE}} = 0{\cdot}30$

SECTION 5.

DISCUSSION OF RESULTS.

The results of these investigations showed that the gridded chamber was capable of measuring numbers of ions greater than 12,000 with a reasonable degree of accuracy. The shapes of the pulse distributions obtained for various amounts of ionization indicated that the spread was due principally to the noise of the amplifier, and that the spread introduced by the chamber was of smaller magnitude. Thus the gridded ionization chamber provides a means of overcoming the distortion caused by variations in the position of formation of the charge and allows high frequency amplification to be used.

When suitable potentials have been obtained for the grid and the deflecting plate the chamber shows a good degree of linearity. For the ionization chamber described here the maximum collection efficiency was obtained when the grid voltage was between 0.3 and 0.4 times the plate voltage; the optimum plate potential was 25 volts per centimetre pressure used. With this arrangement an accuracy of a few percent may be obtained in the estimation of the ionization produced by a single particle, except for small amounts of ionization, for which the accuracy is considerably reduced by the noise of the amplifier.

For the measurement of the ionization of electrons

and mesons of high energy it would therefore be desirable if the number of ions formed was greater than 20,000. Assuming Dunlap's value of 67 ion pairs per cm. in argon, it would be necessary to pressurise the chamber to about 8 atmospheres, and this would require plate potentials larger than 10,000 volts. Measurements could readily be made with mesons if insulation difficulties were eliminated, but the very substantial walls made necessary by the pressure would make measurements on electrons very difficult. Moreover, the measurement of the variation of specific ionization with energy could not be carried out for energies less than several million electron volts, since the formation of a sufficient number of ions would require the expenditure of some 400 kev.

The initial stages of development of the proportional counters described in the second part of this thesis showed that these instruments possessed considerable advantages over a pressure chamber. For example, the pulse distributions obtained were considerably sharper than those observed here, although the number of ion pairs measured was very much smaller. It would be possible to investigate radiations of considerably lower energy, the amounts of ionization being some hundred times smaller. Investigations of electronic radiations and mesons could therefore be carried out without the difficulties associated with pressurisation. For example, the calibration of an ionization chamber would

require low pressure measurements and uncertainties would arise from the large pressure increase necessary. In the proportional counter, however, the measurements could be carried out at the same pressure as the calibration, and the calibration supplied by means of X-rays forming a number of ion pairs very close to those being measured. Specific ionization measurements could be carried out without involving the expenditure of a large energy and a more accurate determination of the energy dependence made. Measurements of this type are being carried out in this Department using the proportional counter technique.

For these reasons the investigations described in the following sections were carried out in preference to the continuation of the studies described here.

PART II.

STUDIES OF LOW ENERGY RADIATIONS

WITH PROPORTIONAL COUNTERS.

by

John Angus, M.Sc.

March, 1949.

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SECTION 1.

SUMMARY.

This thesis contains the results of a series of investigations of the properties and characteristics of proportional counters and their application to studies of the low energy radiations emitted by a number of radioactive isotopes. The study was initiated because it was felt that a considerable demand existed for an instrument which would allow rapid and accurate measurements to be made of the energy and intensity values of radiations of low energy. Such measurements had in several cases been rendered inaccurate and somewhat uncertain by the difficulties associated with the alternative methods available. In particular, it was considered that the development of proportional counting tubes for such studies would make possible the determination of the energy distribution of the beta-particles emitted by a source for which the maximum energy of the particles was small. For several sources of this type the energy distributions had not been measured with accuracies sufficiently high to allow significant deductions to be made regarding the agreement between the experimental results and the predictions of the various theories of beta-decay. Among these sources there existed at least two which possessed a very small number of constituent particles, and accurate determinations of the

shapes of the energy spectra of these isotopes would give results of considerable importance.

Cylindrical counters operating in the proportional region have recently been used for measuring the energies of protons and alpha-particles from the number of ion pairs which they produce. In the work described here the method was extended to measurements of the energies and intensities of electrons and electromagnetic radiations of both nuclear and atomic origin. The proportional counting tubes used varied in dimensions and construction, each being specially designed according to the nature of the radiation and the type of investigation proposed. These tubes were used in conjunction with a linear amplifier of high gain and low noise level, and the output of the instrument was recorded and analysed by means of a photographic film method. It was found that, with this system, energy and intensity measurements of relatively weak sources could be made with considerably greater ease than by other methods, and that the accuracy of the measurements was frequently much greater.

The energy distributions of the beta-particles emitted by tritium (H^3) and carbon (C^{14}) were investigated by this method, the sources being introduced into the gas mixtures of the counters. The results obtained from tritium gave an upper energy limit of 17.9 ± 0.3 kev; the shape of the energy distribution agreed with the predictions of the

Fermi theory to within 10%. From a detailed comparison of the shape of the spectrum near the end-point with theoretical predictions assuming various neutrino masses the rest energy of the neutrino was estimated as between 0 and 2 kev. The upper energy limit for carbon 14 was measured as 157.5 ± 5.0 kev; the shape of this spectrum for energies below 50 kev differs considerably from the predictions of the Fermi theory, the transition $C^{14} \rightarrow N^{14}$ being forbidden.

An investigation of the gamma radiation of radium D confirmed the presence of several gamma rays and a large intensity of L X-radiation emitted during the decay process. The energies of the principal gamma rays were determined as 46 kev, 25.8 kev and 7.8 kev.

SECTION 2

INTRODUCTION

The study of the properties of elementary particles and nuclei comprises one of the most important fields of investigation in modern physics. The object of this study is to obtain a comprehensive collection of experimental data which will reveal the structure of the nucleus and the nature of the processes which lead to its existence and to provide material for tests of present nuclear theories which are more rigorous than those afforded by information so far available.

Information regarding nuclear structure is obtained mainly by studies of the radiations which are emitted when changes take place in nuclei, either spontaneously, or as a result of their disturbance by collisions with other particles. In these transitions particles and electromagnetic radiations are emitted, and a study of the energies and other properties of these products reveals certain characteristics of the nuclei existing either before or after the transitions take place; in particular, the processes involved in transformations of radioactive nuclei yield information regarding the properties of both parent and product atoms. From studies of the products of radioactive disintegrations in various elements it is deduced that transitions occur between definite states of the nuclei,

these states possessing quantised energy values and representing the only configurations in which the nuclei exist; this was found to apply even to states in which the nuclei exist for times so short that a further transition follows within an interval too small to be measurable.

A radioactive nucleus undergoes a transition to a neighbouring nucleus by the emission of electrons or positrons, or, more infrequently, by the capture by the nucleus of an electron from an extra-nuclear orbit. Loss of energy by an excited nucleus occurs by means of transitions between the energy levels accompanied by the emission of quanta of electromagnetic radiation. Since in the majority of radioactive decays the first transition leaves the nucleus in an excited state, the electrons or positrons are accompanied by one or more groups of gamma rays emitted by the nucleus in the second type of transition while it is settling down to a configuration which is stable. Direct measurements of the properties of excited states and radioactive nuclei are extremely difficult and studies of these two types of transition form the only means of determining the properties of states other than the stable states.

Measurements of the energies and lifetimes of the transitions indicate that the levels possess, as well as a definite energy value, a further property which may be expressed in terms of one or more theoretical quantum

numbers; in the stable configurations the principal of these quantum numbers has been associated with the spin of the nucleus, and attempts have been made to determine the principal quantum numbers associated with the other levels by means of selection rules. The position is much more difficult than that for atomic spectra, however, and a study of the energy values of the nuclear levels and any further information from which the quantum numbers may be deduced with more certainty is therefore of prime importance in increasing the understanding of nuclear properties.

For those nuclei which decay by the emission of an electron or positron the particles are observed to be emitted with a continuous distribution of energies up to a maximum value. In view of the quantised nature of the energy levels it has been necessary to assume that the emission of an electron is accompanied by the emission of a second particle, with which the energy is shared. The maximum energy of the electron energy spectrum represents the energy difference between the two levels involved; since the second particle has not been observed experimentally, it is necessary to assume that it has no charge and a comparatively small mass. Various theories have been put forward to account for the shapes of the energy distributions of charged particles from different sources in terms of the properties of the levels of

parent and daughter nuclei and the properties, in particular, the mass, of the uncharged particle, the neutrino.

Experimental results of measurements of the energy distributions of electrons and positrons from various radioactive sources are gradually being obtained and improved. In many cases, however, the accuracy of the measurements is still not sufficiently high to allow detailed comparison with the predictions of the various theories to be made: in other cases the section of the distribution investigated has not been that in which the theories differ very greatly, a principal example being the lack of information available regarding the shapes of the distributions in the low energy region for all but a very small number of isotopes; this has been dictated by the limitations of the experimental methods employed. Frequently, also, the nuclei have been of such complex nature, and the energies of the radiations so large, that valid tests of any theory have not been possible because of the large and uncertain corrections introduced. Apart from the reasonable agreement of several of the experimental distributions with the distributions calculated by means of the Fermi theory of beta-decay[1], an agreement which extends principally over the upper part of the distribution, very few conclusions can be drawn. The number of cases in which disagreement exists between the experimental distribution and that calculated with the simple Fermi theory, indicates that

transitions of several types occur; this indication is supported by theoretical discrepancies which exist between the half-life and upper energy limit of a number of transitions.

Among the points of interest on which information has been most urgently required in this field are, firstly, the energy distributions of beta-particles from sources which possess a very small upper energy limit, secondly, the energy distributions obtained from nuclei which contain only a small number of constituent nucleons, and, thirdly, an estimation of the mass of the neutrino. It will be shown that these deficiencies have arisen principally as a result of the difficulties associated with the normal methods of investigation in the region of low energies. The new experimental method of measurement described here allows such determinations to be made in a number of cases. During the time that this work was being carried out extensions have been made in the normal methods towards these low energies, but it is considered from the results of this study that the method described here is considerably more accurate than any alternative method at present available for the study of low-energy beta-spectra. This work has also allowed a determination of the mass of the neutrino to be made within limits of accuracy much closer than those afforded by any other technique.

For those nuclei which decay by the emission of both a charged particle and electromagnetic radiation, and in nuclear processes involving the de-excitation of a nucleus by the same means, the gamma rays are observed to emerge with one or more definite quantum energies. It is important to know, firstly, the energy of each group of gamma rays being emitted, and, secondly, the order or arrangement in which the transitions occur if more than one gamma ray is observed.

Energy determinations are carried out by several methods involving various degrees of accuracy and each applicable only to a particular range of energies. For high energy gamma rays, for example, measurements are made from studies of the absorption properties of the radiation, or by an analysis of the Compton electrons which it produces; for radiation of lower energy measurements are made from studies of the photoelectrons which it releases, or by X-ray crystal reflection technique. Both methods are severely limited by considerations of efficiency of detection and the length of time required to obtain good accuracy.

The determination of the de-excitation decay scheme of a nucleus involves also the measurement of the coincidence of occurrence of two gamma rays of different energies. The normal method employed involves the use of Geiger counters and absorbing materials. The efficiency of detection of gamma rays of high energy with the conventional

Geiger tube is of the order of one percent, and this considerably limits the uses to which the method may be applied. For example, with such an efficiency the number of coincidences observed is only one ten thousandth of the number which would be detected if the efficiency of each counter was one hundred percent. For various sources it is therefore exceedingly difficult to obtain accurate measurements of coincidence rates because of relatively high background, counting rates and in the majority of cases the estimation of triple events involving more than two counters is impossible, due to the high rate of random events leading to the recording of spurious coincidences.

During the period in which this work was being carried out investigations have been made elsewhere of the properties of scintillation counters[2] for the detection of gamma radiation. The greatly increased sensitivities afforded by counters of this type have made possible experiments on effects which are of considerable theoretical importance; such studies as those reported by Deutsch[3] on the angular correlation of successive gamma rays and their direction of polarisation, should give information regarding the quantum numbers of levels and the order of multipolarity of the radiation. With the increasing development of these counters more information should become available regarding these properties. The increased sensitivities afforded by

the counters to be described here, even though the energies for which this sensitivity is high are limited to less than 200 kev, should be of considerable value in extending these types of measurements to low energies.

A most important field of gamma ray studies is the investigation of low energy radiation, that is, radiation with energy less than 200 kev. Interest centres on this field for several reasons. Firstly, there is every indication that low energy gamma rays may be emitted in several disintegrations, and that these have so far not been observed due to limitations of the methods available for their detection. Secondly, several radioactive transitions give rise to low energy gamma radiations which do not appear to fit any recognised decay scheme, a particular example being that of radium D. Thirdly, in the majority of cases in which nuclei decay by the emission of gamma radiation of low energy, the gamma rays may appear outside the atom as photons or as secondary electrons; in the second case the energy of the ray is transferred to one of the electrons surrounding the nucleus. While most of these low energy gamma rays are accompanied by a measurable number of secondary electrons, the probability of secondary electron production shows considerable variations between different sources. This probability has been shown theoretically to depend on the energy of the radiation, the atomic number Z of the product

element, the atomic shell from which the electron is ejected, and the multipole order of the radiation; the latter arises from a theoretical consideration in which the nucleus emitting the radiation is regarded as an electric or magnetic oscillator with the characteristics of a dipole, quadrupole, or multipole. The order of polarity is connected theoretically with the quantum numbers of the energy levels in the following manner; a transition between two levels differing in quantum number by a value l should give rise to a gamma ray with an order of polarity of 2^l. Since, as has already been pointed out, the variation of the probability of internal conversion with the order of polarity is considerable, an estimation of the probability may allow a definite determination of the order of polarity. The internal conversion coefficient, defined as the ratio of the number of secondary electrons to the number of gamma rays, has been calculated as a function of the energy and the nuclear charge for the various orders of polarity[4]. For example, the internal conversion coefficient of a gamma ray of energy E in the K-shell of an atom with charge Z is given by:-

$$\alpha_K = \left(\frac{l}{l+1}\right) \alpha^4 Z^3 \left(2mc^2/E\right)^{l+5/2}$$

where $\alpha = 1/137$. Corresponding expressions exist for the other shells.

The results of experimental determinations may in

the main be explained by these expressions, assuming a value for ℓ. Nevertheless, some anomalies exist. For example, among the naturally radioactive elements there are several radiations for which the ratio is either too great or too small. A study of anomalies such as these would be of great value.

The measurement of the number of X-rays accompanying the transition also permits a determination of the ratio to be made, since the ejection of an electron from an atomic shell is followed by a rearrangement of the atom. This method could with advantage be enlarged, since cases exist in which the intensities of X-rays and secondary electrons do not agree.

The identification of a K-capture decay depends upon the identification of the X-rays resulting from the removal of an extra-nuclear electron. While the critical absorption method of analysis is adequate to reveal the presence of the characteristic radiation, and identify the nuclear charge, if no other low-energy radiation is present, it would be of considerable advantage if the technique could be extended to allow coincidence measurements to be made with more ease, for example between the X-rays and gamma rays. Since the nucleus and the K-electron must be considered to form a single system it is possible that special effects might appear if angular correlations were sought between X-ray and

gamma ray.

The discovery of isomerism and the existence in quite a number of nuclei of meta-stable states has also led to investigations involving low-energy gamma radiation. In these isotopes a nucleus may exist for a very appreciable length of time in a state other than the ground state, and finally decay by the emission of a low energy gamma ray transition which usually exhibits a high internal conversion coefficient. The lifetime of a gamma ray transition, like the conversion coefficient, is related to the energy and the multipole order of the radiation[5]. This relation may be expressed as follows:-

$$\log \lambda = 20.3 - 2 \log(1.3.5 \cdots 2\ell - 1) - (2\ell + 1)(1.3 - \log E) - 2\ell(0.84 - 1/3 \log A)$$

where λ is the decay constant (0.68/lifetime)

E is the energy of the gamma ray

and A is the atomic weight of the element. Cases exist where measurably long lifetimes are observed; these occur when the energy of the gamma ray is small and the multipole order high, and explanations of both isomerism and the existence of meta-stable states are based on this consideration. Here again estimations of the internal conversion coefficients provide useful cross-checks of the theoretical interpretation; in some cases, indeed, the discovery of the process has depended on the identification of the charge of

the product nucleus by means of the X-rays emitted.

While some of the effects discussed here had been observed in the naturally radioactive elements, the recent discovery that many radioactive isotopes of the other elements could be produced by artificial methods has considerably extended the frequency of occurrence of transitions of every type, and the variety of radioactive sources available has also extended the range of the phenomena to be investigated. Among the most notable of these extensions are the increase in the range of energy of the decay products, the increase in the range of lifetimes, in particular the inclusion of several excited states with lifetimes between 1 second and 1 micro-second, and the discovery of transitions with a high degree of complexity. This rapid expansion in the number of sources available, and the demand for more detailed information regarding the other radiations emitted, must certainly involve the development of improved experimental methods of investigation. Vigorous efforts are being made to improve the existing methods and to devise new techniques to meet these demands.

Among the transitions of exceptional interest there are a sufficient number involving radiations with energies less than 200 kev to constitute a considerable demand for an instrument which in this energy region would permit rapid and

accurate measurements of energy and intensity to be made. As has been pointed out these radiations include primary and secondary electrons, gamma rays and X-rays; in a suitable gas, however, the electromagnetic radiations may readily be converted into photo-electrons, so that the problem then resolves into one of the measurement of the number and energies of electrons of fairly low energy. The two methods used for such determinations are the measurement either of the range or number of ion pairs produced by individual particles in a cloud chamber or of the curvature in a magnetic spectrograph or spectrometer. In the latter instruments the source must be of fairly strong intensity and must be confined to an area as small as practicable; this means that, with the solid sources used, considerable uncertainties arise due to the thickness of the source mounting (giving rise to reflection effects) and the thickness of the source itself (giving rise to absorption of the electrons which originate some depth below the surface). It is clear that these effects are most serious for electrons of low energy. By using thinner sources and source mountings together with Geiger counter detection these effects may be reduced, but even so results obtained below 30 - 40 kev are usually open to some doubt, and it is much more desirable to use a method which involves a gaseous source. The cloud chamber method possesses this advantage but the drop-counting of the tracks

is extremely slow and laborious; it is therefore not surprising to find that the number of tracks measured is usually so small that statistical fluctuations are considerable and only the barest outline of the distribution obtained. While the alternative method, namely the measurement of the ranges of the particles, is less tedious and a larger number of tracks may therefore be measured, the energy estimations depend on a range-energy curve which at present does not appear to be very accurate at very low energies. However, the recent development of a cloud chamber method of energy measurement of photoelectrons from low-energy radiations by T. San-Tsiang et. al.[6] has shown the capabilities of such an instrument; while the accuracy is fairly good compared with previous methods, it is felt that the instrument is not adequate to deal with most of the problems outstanding in the low-energy region.

As a result of a preliminary investigation of the properties of a cylindrical counter operating in the region of proportional gas amplification it was considered that a method could readily be developed to allow measurements of energy and intensity to be made in the region described above. This method involved the use of a proportional counter with a linear amplifier of high gain and low noise in a manner similar to the more normal ionization chamber and linear amplifier technique. The latter is of course not directly

applicable to such energies since the number of ion pairs formed by an electron with an energy of, for example, 10 kev is only 300, and this number is much too small to give a pulse which could be amplified successfully in view of the variations present in the first valve of the amplifier. These variations, which arise from thermionic disturbances in the emission of the filament and in the grid circuit, may be lowered by a suitable selection of band-width in the subsequent stages of amplification, but it is not possible at present to reduce them to such a stage that a pulse of 300 ions would be observed. A pulse of ten times this number could be detected, but, if the pulse is to be considerably greater in amplitude than the average noise level of the amplifier, considerably more ion pairs would be necessary; it is therefore impossible to make accurate measurements with an ionization chamber if the energy of the radiation is less than 150-200 kev.

By the use of suitable multiplication in the proportional counter, however, the level of the pulse could be raised by a factor which would allow easier multiplication in the amplifier, and the instrument could be applied to radiations of very small energy, in fact, down to energies for which only one or two ion pairs are formed. In order to test that this technique was capable of energy and intensity measurements of high accuracy the investigations reported in

the following sections were carried out. A variety of proportional counters were used and the results have in every way confirmed the original considerations which indicated that counters of this type are exceedingly useful for measurements in this range of energy.

A proportional counter is essentially a cylindrical ionization chamber in which the ions produced by individual particles are multiplied by collision processes in the gas on their passage to the central electrode. This multiplication is carefully controlled and limited in such a manner that the pulse obtained at the collecting electrode is directly proportional to the number of ions produced in the initial process. The greatest multiplication used is much smaller than the multiplication used in Geiger counters, and the proportional counter has characteristics and properties lying between those of an ionization chamber and those of a Geiger counter.

Historically the development of the proportional counter by Geiger and Rutherford[7] preceded that of the Geiger-Muller counter; proportional counters have, however, been used most frequently for counting particles in a similar manner to the latter, since they may be adapted to discriminate between ionization produced by a heavy particle and that produced by electrons and electromagnetic radiations. These

counters are most frequently used to separate ionization events involving large numbers of ions from an intense background of smaller ionization events, and considerable attention has been paid to their performance in such conditions.

A further use of the proportional counter is the measurement of the number of ions produced by an ionizing particle or radiation, as described above; if the complete track of the particle is confined within the counter this measurement allows a determination of the energy to be made, and the method is essentially the same as the ionization chamber method, differing only in the relative number of ions collected. The smaller amount of attention which has been paid to this use may be attributed, firstly, to the reasonable adequacy of the ionization chamber for measuring the energies of heavy particles, and, secondly, to the lack of a satisfactory understanding of the mechanism of proportional gas multiplication.

Since the recent work of Korff and others, and the development by them of a theoretical explanation of the gas multiplication process, several proportional counters have been designed for energy measurements of heavily ionizing particles. These counters have been shown to have some advantages over the ionization chamber, and the results of the work described here, in which proportional counters are

applied to the ionization produced by electrons and electromagnetic radiations of much smaller energy, indicate that these satisfactory properties may be retained also in this energy region. The energy range of the instrument is confined to energies below 150 kev, a limitation imposed by the dimensions of the counters, and the following characteristics are noted in this range:-

(a) Strict linearity of the output pulse with the energy of the radiation for a number of gas fillings,

(b) Good resolution of photoelectron lines of uniform energy, allowing accurate measurements of energy to be made,

(c) High sensitivity to gamma radiation, and

(d) The existence of counting plateaux of several hundred volts extent in which the rate of variation of counting rate with voltage is remarkably small.

These results have been obtained from a study which consisted of the measurement of the variations of output pulse size with the energy of the radiation, the voltage and the gas mixture; in the majority of these investigations photoelectrons from the fluorescence X-rays of various elements have been found to provide a source of radiation of suitable energy, and studies have also been made of the distribution of pulses from which the widths of the X-ray lines and the resolving power of the instrument could be estimated. These

measurements also provide a set of data by means of which a check may be made of the theory proposed by Korff, and from which it is possible to determine the optimum working conditions for any specific purpose.

This proportional counter may therefore be seen to provide a relatively simple means of measuring the energies and intensities of nuclear and atomic radiations. Accordingly an investigation of several sources emitting low energy radiations is being carried out; the results which have been obtained with the first three of these sources are contained in the following sections. The sources are tritium ($_1H^3$), carbon ($_6C^{14}$) and radium D ($_{82}Pb^{210}$). While further details regarding each source will be found in the appropriate sections, the reasons for which they were chosen will be given here.

The investigations of the beta-spectra of tritium and carbon form part of a series of investigations of the radiations of a group of isotopes emitting low-energy beta particles, the others being sulphur 35, nickel 59, and radium D. Tritium is undoubtedly of considerable importance in view of the very low energy of the end-point (18 kev) and the very simple nature of the nucleus; as has been pointed out already, these both enhance the value of any results to be compared with a theoretical prediction. Carbon 14 has been chosen principally because of its low mass value and the "forbidden"

nature of the transition; it may also be readily converted into a gaseous source. With an upper energy limit of 155 kev, it has recently come within the scope of the magnetic spectrographic method, and the results obtained here are compared with those obtained with the latter method. An investigation of the spectrum of sulphur 35 is at present being carried out; this has an upper energy limit of the same order of energy and has also been investigated with the spectrometer, so that comparisons should again be possible. It is clear from the very great differences in shape which appear between the two spectra already studied that the completion of these studies should give considerable information regarding the simplest of the beta-spectra with various orders of "forbidden-ness."

The investigation of the gamma radiation of radium D has been carried out for two reasons. Firstly, this source is included among the low-energy beta-emitters to be studied, and a clear picture must first be obtained of the low energy gamma radiation and the electron conversion lines to be expected. Secondly, it forms the first of a series of studies on the low-energy gamma radiations of various elements which show anomalous decay schemes; for these gamma ray studies it is not necessary to have a gaseous source, and a search may also be made of a large number of sources for low energy gamma rays and X-rays.

The methods described here are well suited to the measurement of numbers of ions between 10^2 and 10^3, and a determination of the specific ionization of the meson may be made. This development appears to require only the construction of a satisfactory method of coincidence selection of those tracks which are of approximately equal length; the number of such tracks seems to be adequately large for such a measurement. This, if carried out in conjunction with a measurement of the range or curvature, would allow the mass of the meson to be calculated, although the accuracy of such a calculation is not particularly high.

The various properties of these counters should prove of considerable value in future work. It is felt that the results given here illustrate the types of problem to which the technique may be applied; while the number of such problems is large, the method may also be applied to other problems beyond the scope of this investigation.

Prior to these studies no proportional counter measurements of the energies of electrons or gamma rays had been reported. During the course of this work two reports have appeared. The first[8] describes the application of a methane-filled proportional counter to such measurements, though the gas gains indicated seem to be very high; no energy measurements from which the performance of the counters

may be estimated have so far been given. The second report[9] describes the study of the K X-rays emitted by radioactive argon, the method being essentially similar to that employed here; the use of a much less selective method of pulse recording has, however, failed to show the high degree of resolution obtained in these studies.

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SECTION 3

PREVIOUS WORK ON PROPORTIONAL COUNTERS

Theoretical.

Korff[10] proposed a theory of the process of gas amplification taking place in a proportional counter which may be described as follows. Consider a cylindrical counter having a central wire of radius r_1 and a cathode of radius r_2 maintained at a voltage V. Gas amplification will take place when an electron during its passage towards the central wire picks up from the field sufficient energy in one mean free path length to ionize an atom of the gas. Suppose that this occurs when the field strength reaches a value equal to or greater than $E_{crit.}$ and that the field strength reaches this value at radius r; then, on the average, the number of electrons moving towards the wire will be doubled in each mean free path length between r and r_1. The relation between r and $E_{crit.}$ is:-

$$E_{crit.} = V/r(\log. {}^{r_2}/_{r_1}).$$

As the voltage on the counter is raised, gas amplification will start at a value $V_{crit.}$ where r is greater than r_1 by one mean free path length and, since this difference is very small, being of the order of 10^{-3} cms at one atmosphere, we may write:-

$$V_{crit.} = E_{crit.} (r_1. \log {}^{r_2}/_{r_1}).$$

and hence:- $r/r_1 = V/V_{crit}$.

Now if on the average dN ion pairs are formed by N electrons in a distance dr, we may write $dN = \alpha N\, dr$; α will depend on the field strength and may therefore be written as $\alpha(r)$. If N_o is the number of ion pairs initially produced, the number of ion pairs observed in the counter when a voltage V is applied is therefore given by:-

$$N = N_o\, e^{\int_{r_1}^{r} \alpha(r)dr}$$

that is, $$N_o\, e^{\int_{r_1}^{V/V_1 r_1} \alpha(r)dr}$$

The value of α has been determined by Korff to be $(a\, N_m\, C\, V / r)^{\frac{1}{2}}$, where a is the ionization cross-section of the electron divided by its energy, N_m is the number of gas atoms or molecules per unit volume, and C is the capacity of the counter. The gas amplification is defined to be the ratio of the charge collected to the charge produced and is therefore given by:-

$$A = N/N_o \quad e^{2(aN_mCV)^{\frac{1}{2}}.r_1^{\frac{1}{2}}\left[(V/V_{crit})^{\frac{1}{2}} - 1\right]}$$

where $V > V_{crit.}$ and equal to unity for $V < V_{crit.}$ If $V = V_{crit.} + \delta V$ this expression becomes:-

$$\log A = k.\ \delta V$$

and therefore the gas amplification would be expected to increase exponentially with voltage beyond the threshold value.

Experimental.

The assumptions on which this theory is based are:-

(1) That no electrons are lost by recombination with gas molecules and ions,

and (2) That no electrons are produced by other processes such as bombardment of the cathode by positive ions or photoelectric emission.

Rose and Korff[11] investigated the variations of A with V for various gas mixtures and pressures and found good agreement with this theory; departures occurred in conditions where assumption (2) could not be applied, that is, where no attempt was made to absorb the photoelectrons. By the addition of a sufficient amount of a polyatomic gas to prevent the spread of photons these departures were removed; this practice was adopted in the work at higher pressures described here and has been found to give very satisfactory results.

Rose and Ramsey[12] carried out similar experiments for values of A between 1 and 100 and observed the predicted linearity between log A and V, except for small values of A. Near the threshold voltage the onset of gas multiplication was less abrupt than that predicted on the simple theory; since certain quantities inserted in the equations given above represent average values, the treatment is statistical in nature and such departures are likely to be observed, particularly where either the number of ions produced initially or the number of mean free path lengths producing multiplication is small.

The statistical nature of the process also gives rise to fluctuations in the pulse sizes obtained. An analysis by Snyder[13] of this fluctuation, in which the number of ion pairs produced by the ionization radiation is assumed to be $n \pm \sqrt{n}$, gives a mean square deviation:-

$$\Delta h = \sqrt{2/n} \times h,$$

where h is the final pulse size. It will be shown that this equation is not applicable to the results given here, since:-

(1) The variations of the number of ion pairs produced by an electron stopping in the gas is considerably less than $\sqrt{n}$, and

(2) A simple analysis of the problem shows that the position is more complex and that the result of the analysis depends on the assumptions made.

Coon and Noble[14] describe the application of argon-filled proportional counters to measurements of the energy of neutrons by means of proton recoils. They chose proportional counters as they wished to observe radiations with energies below 100 kev. The counters, which had a cathode radius of 2 in. and a wire diameter of 0.005 in., were operated at gas gains between 5 and 50. The pulse size appeared to be accurately proportional to the energy of the recoiling protons, the variation of gas gain with voltage was found to be relatively slow (increasing by a factor of 10 in 400 volts), and the counters were found to remain accurate to

10% over a period of a week.

Koontz and Hall[15] describe the construction of a cylindrical counter which could be used either as an ionization chamber or as a proportional counter. This counter had a cathode diameter of 1 in. and a wire diameter of 0.0025 in., and was filled with argon - nitrogen mixtures to a total pressure of several atmospheres. Comparison of the pulse distributions obtained from protons of 0.6 Mev energy produced in the n,p reaction in nitrogen showed that the width of the proton group was smaller when gas amplification was used. This was explained as follows: in a cylindrical ionization chamber the size of a pulse obtained from a proton track of fixed energy depends upon the average radius at which the track is formed, and this gives rise to a considerable distortion of the pulse distribution; for the proportional counter, however, this distortion is absent since an overwhelming proportion of the ion pairs collected is produced within a few mean free path lengths of the wire.

The operation of this counter in experiments on the scattering of neutrons in helium is described by the same authors[16]. The curves obtained in the two conditions of operation again indicated that in each case the pulse size was directly proportional to the energy.

Since it is proposed to discuss in a later section the counting plateaux obtained with the low energy proportional

counters reference may be made here to the work of Simpson[17]. A description is given of the properties of a proportional counter used for counting alpha-particles in the presence of large amounts of beta-activity. The counter used was filled with methane and had a plateau of about 300 volts (½% variation in 200 volts); although the presence of a beta-source of high strength (5×10^9 disintegrations per minute) reduced this plateau to 150 volts, satisfactory operation could be obtained. This paper is a good example of the type of results which can be obtained when proportional counters are used solely for the registering of particles.

Borkowski and Fairstein[8] give a brief report of the use of proportional counters for counting X-rays and soft electrons. The counters, which had a wire diameter of 0.005 in. and were filled with methane at a pressure of one atmosphere, were stated to allow very rapid counting and to have plateaux of about 400 volts with a slope of 1% per 100 volts. While interested principally in the counting properties of these tubes, they point out that energy measurements may be made with them, since they are operating in the proportional region.

The extension of proportional counters to energy measurements of radiations which are otherwise difficult to study, and the high sensitivities which they offer for the detection of soft gamma rays were considered to be of great

importance. A study was therefore carried out of the radiations of a number of artificial radioactive isotopes known to emit beta-particles of low energy, and this study was extended to sources emitting gamma rays and X-rays of low energy. Several properties of the counters were also investigated and the results of the latter study indicate the advantages which are afforded by such a technique and support the explanations of the mechanism of operation put forward by Korff.

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SECTION 4

DESCRIPTION OF APPARATUS.

The method consisted of the use of a proportional counter in conjunction with a linear amplifier of high sensitivity, the output pulses from this amplifier being recorded by a photographic method. A description is given in this section of the various parts of the apparatus and the reasons for which this method was adopted.

In the course of this work a large number of counting tubes of various constructions were used depending on the type of radiation to be investigated. These counters varied considerably in diameter and length and were constructed in either metal or glass. This variety of sizes gave rise to various problems, but it was found that with reasonable care the counters could be made to operate satisfactorily and remain stable in characteristics over quite long periods.

It is interesting to observe that this technique extends to very low energies the ionization chamber type of investigation, the energy of a particle being determined from the number of ion pairs which it produces. These investigations have so far been limited by the signal-to-noise ratio of the amplifier to about 5000 ion pairs, or 150 kev. By using gas amplification factors up to 10^3 this value may be lowered to a few hundred electron volts, and it is considered from the results of this investigation that the fluctuations

introduced are reasonably small.

Proportional Counting Tubes.

Tubes with Glass Envelopes.

Figure 1 shows the type of counter used in the initial experiments. The envelope, which was $2\frac{1}{2}$ inches in diameter and about 10 inches long, was constructed in Pyrex glass 2 - 3 mm. thick. The cathode consisted of a thin sheet of aluminium with polished surface, held in position by light pressure against the glass; connection to the high voltage supply was provided by a wire contact led out through a seal in the side arm of the counter. The collecting wire, tungsten of diameter 0.002 or 0.004 in., was held taut by a spring inserted as shown into one of the tubes sealed into the ends of the counter. These tubes served as central mountings for the wire and had a bore of a few millimetres; they were coated on the outside surface both inside and outside the counter with a deposit of "Aquadag". They therefore served as guard-rings and were earthed through the two small seals so that they also prevented the central wire from direct electrical exposure to any of the insulating surfaces which might become charged. This is of especial importance since any accumulation or rearrangement of charge distribution on these surfaces when the voltage is applied would give rise to spurious pulses and thereby increase the noise level of the

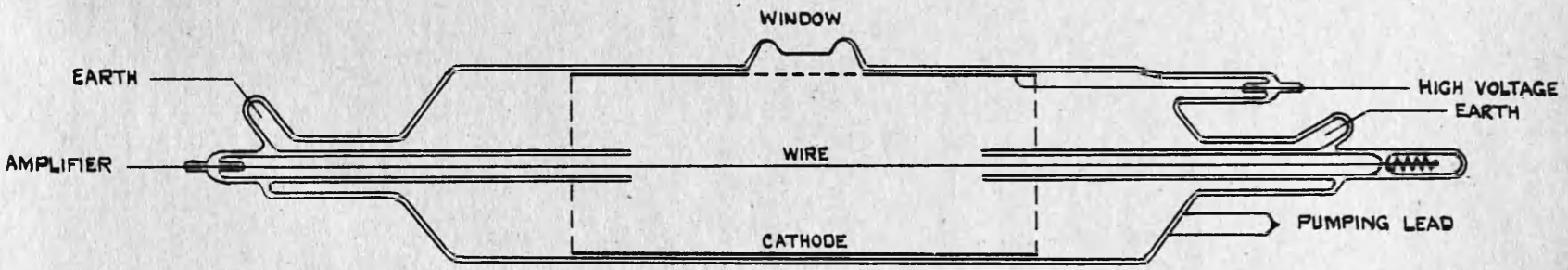


FIG. 1

GLASS PROPORTIONAL COUNTER

TO FOLLOW PAGE 34

amplifier. It was found that with the arrangement shown the number of spurious pulses was reduced to an almost negligible value.

The contact between the wire and the amplifier was screened by a short length of metal tubing connected to the case of the head amplifier to prevent picking-up of external signals. Since high frequency signals would be passed across the space between the cathode and wire, it was necessary to screen the whole counter: this was effected either by enclosing the counter in a metal box, or, where this was inconvenient, by coating the outside of the glass at each end with "Aquadag", extending the coatings to overlap the ends of the cathode. Either method was entirely satisfactory.

Several of the counters were very microphonic due to movement of the central wire; this difficulty was overcome by mounting them on shock-absorbers weighted with several pounds of lead; in the majority of cases, however, no such mounting was necessary.

Finally, a window about a tenth of a millimetre thick was blown in the side of the counter to minimise the absorption of low energy radiations at the wall. A hole of corresponding diameter (3/4 in.) was cut in the cathode; this hole was covered by one or two thin wires to maintain the uniformity of field, and it appeared that no appreciable distortion resulted from this small discontinuity of the

surface of the cathode.

Metal Tubes.

As the range of energies measured was increased it became necessary to use counters of larger dimensions and to arrange for their pressurisation to several atmospheres. A series of counters was therefore designed for construction in copper. A copper cylinder and its end-plates provide the vacuum case and also act as the cathode; the wire supports, which are shown in Figure 2, were insulated with ebonite from the case, which carries the high voltage. The ebonite tubes screwed centrally into each end-plate and the metal guard-rings were fixed to the ebonite by the locking nuts at one end and by the flat plate at the other. The guard-ring had an external diameter of 3/16 in. and could be adjusted so as to extend into the counter by any desired length. The ebonite plug which fitted into the recess in the guard-ring tube provided an accurately centred mounting for the wire; at one end of the counter it housed the spring as shown, while at the other end the wire passed out through a fine hole to the amplifier.

On completion of the assembly the ends of the counter were made vacuum tight by coating the outside with wax; thus, for both the glass and metal counters, no waxed surfaces were exposed to the gas inside the counter. The counters were

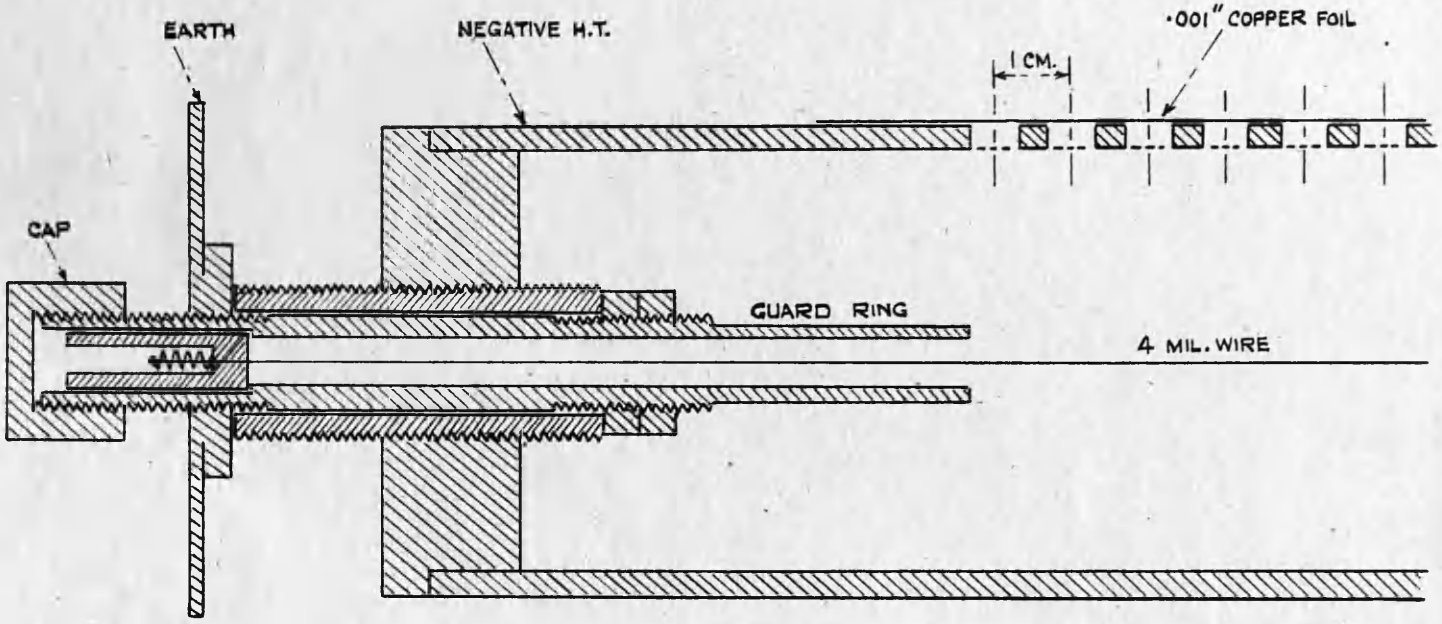


FIG. 2

DIAGRAM OF END OF COUNTER

[SHOWING INSULATED BUSHES]

TO FOLLOW PAGE 36

evacuated and filled through a side tube attached to the main casing and closed by a small glass stop-cock. In the counters which were used for high pressures the vessel was sealed off by means of a thin copper tube which could be bent back on itself and soldered after cutting; this was found to make a very good vacuum seal. Other constructional details of the high pressure counters will be described in the relevant sections. However, it may be noted here that the ebonite mountings were sufficiently strong to allow pressures up to $5\frac{1}{2}$ atmospheres to be applied to the inside of the counters, and that the insulation of these materials behaved satisfactorily at potentials up to 6,000 volts. The developments of technique associated with the use of the beaded counter will be found in Section 8.

The windows of the metal counters were made by drilling holes in the copper wall and covering them with thin foils of copper, aluminium, mica or cellophane. This has several advantages over the glass windows, since any desired thickness of foil could be used, and the absorption properties of the windows could be calculated before they were inserted.

It is unfortunate that the major part of the surface carried the high voltage. The large condensers connected across the high voltage line to provide additional stability enhanced this danger, but the addition of earthed screens was impossible, particularly on the larger counters.

The Linear Amplifier.

Several amplifiers were used in this work; each consisted of a head amplifier and a main amplifier designed by Mr. A. L. Cockcroft. These amplifiers had a very high signal-to-noise ratio, being designed for ionization chamber measurements, and played a very important part in the success of this work. A pulse of 1500 ion pairs arriving at the grid gives a signal which appears above the maximum noise level. Assuming this maximum level to be 1000 ion pairs, the root mean square noise level is 300 ion pairs. This noise is caused principally by fluctuations in grid potential resulting from small grid currents and therefore varies with the input capacity in a similar manner to the signal. For an input capacity of 10$\mu\mu$F, for example, the signal is 20 microvolts and the noise (root mean square) is 4 - 5 microvolts.

A circuit diagram of the head amplifier is given in Figure 3; it consists of a single stage of amplification and a cathode follower stage to enable the pulses to be transmitted to the main amplifier in a low impedance cable. The high voltage lines are stabilised and the first two valves are heated by three 2-volt accumulators. The highest signal-to-noise ratio is obtained when no resistance is inserted in the grid circuit, but the amplifier was usually employed with a 100 megohm grid leak, since, for counting rates greater than 10^4 per minute, the first valve tended to bias-off; the grid

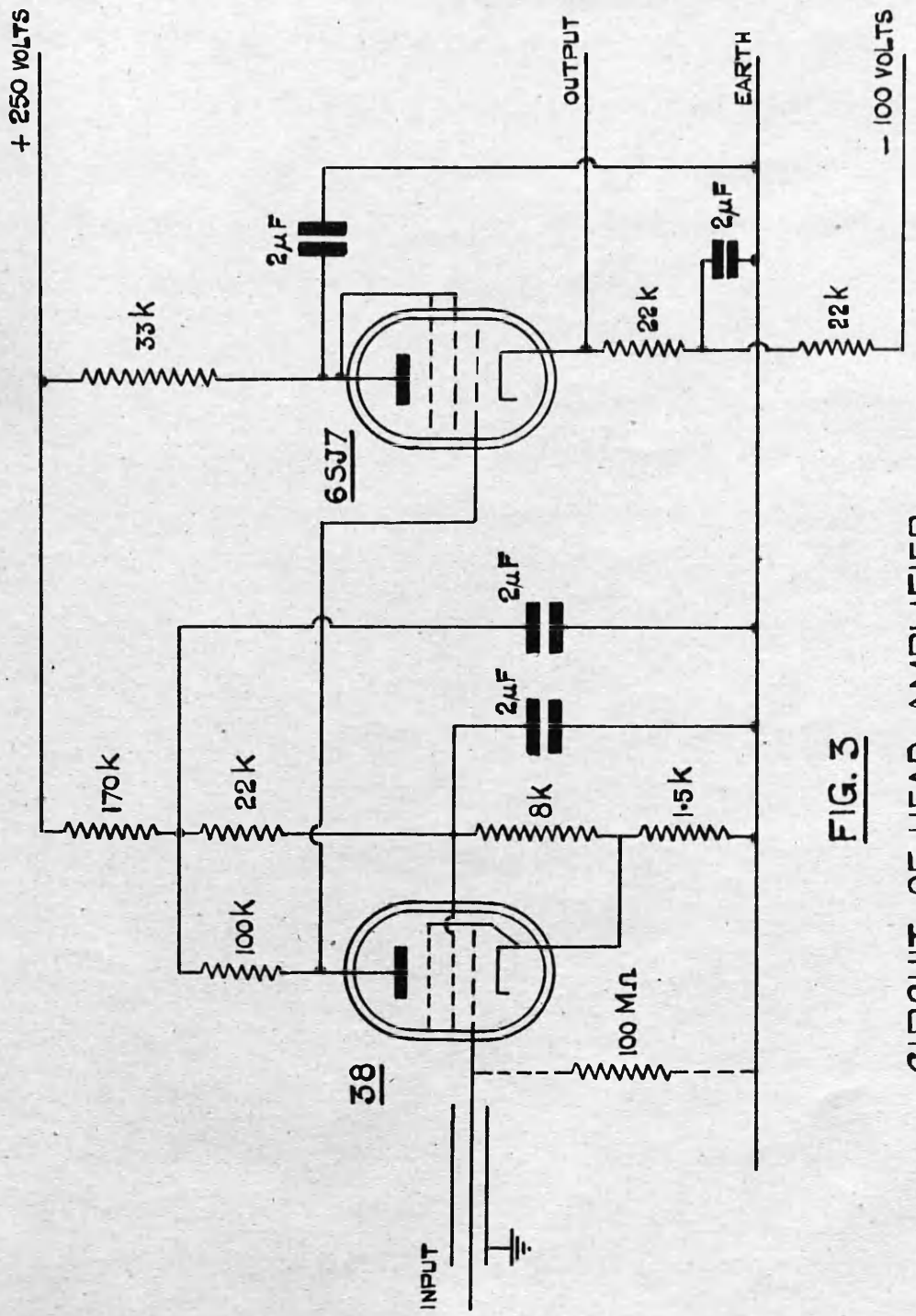


FIG. 3

CIRCUIT OF HEAD AMPLIFIER

leak, while preventing this trouble, slightly reduced the sensitivity of the instrument.

The main amplifier consisted of three stages of push-pull amplification of conventional design with a cathode follower output to the oscillograph. A maximum output pulse of about 100 volts was obtained, and the overall amplification of the system was between 10^6 and 10^7.

Recording and Analysis of Pulses.

The most usual methods of recording the amplitude distribution of the pulses are either

(a) To count in a fixed time interval all pulses larger than a given size, using an electronic discriminating circuit, or

(b) To display the pulses on an oscilloscope and record them on a moving photographic film.

In the first method the counting must be repeated at a large number of discriminator bias settings, when the integral size distribution may be obtained. In the second method the film is analysed to give directly the size distribution of the pulses obtained. Electronic multi-channel pulse analysers have recently been developed, but the number of channels available is too small to allow accurate resolution.

The photographic method was chosen in this work because

(a) Pulses of all sizes could be recorded together

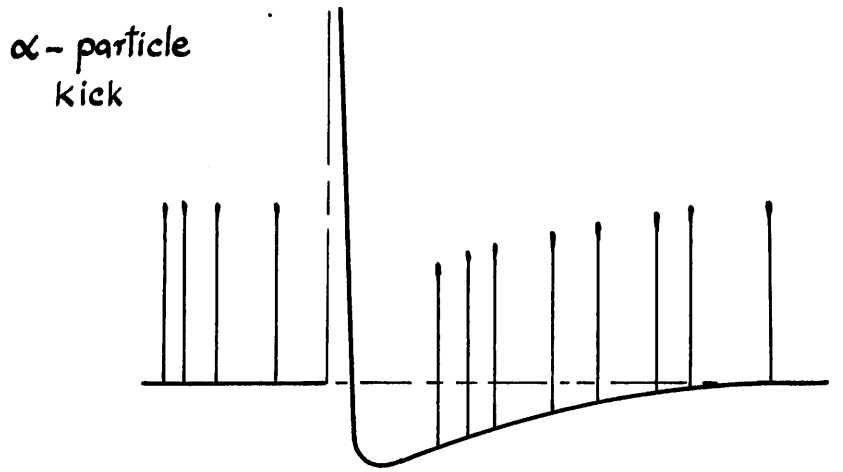
and a sufficient number for analysis could be obtained in a single run of short duration,

(b) The pulses may subsequently be divided into any number of size intervals up to at least 100, whereas, with a discriminator method, corresponding resolution can only be obtained if the counting rates at 100 settings are measured, and if the accuracy of the discriminator settings is correspondingly high.

(c) The short time required for obtaining full film data, even for very weak sources, considerably relaxed the stability requirements of both the high voltage supply and the amplifier gain. For example, a source giving 10,000 pulses per minute could be recorded in four minutes (on about 50 ft. of film) and would give an average of 400 pulses in each energy interval; with a discriminating circuit, using the method of differences, the corresponding accuracy would involve very much greater counting rates or a prohibitively long running time.

(d) The presence of occasional large pulses caused by contamination of the walls of the counters with α-particle emitters causes a momentary depression of the base-line during the time when the amplifier is recovering. If pulses from the source under investigation appear during this time they will be depressed in amplitude as shown in the sketch. Such pulses, which would be erroneously recorded in

an electronic discriminator, may be rejected on inspection during the analysis of the film.



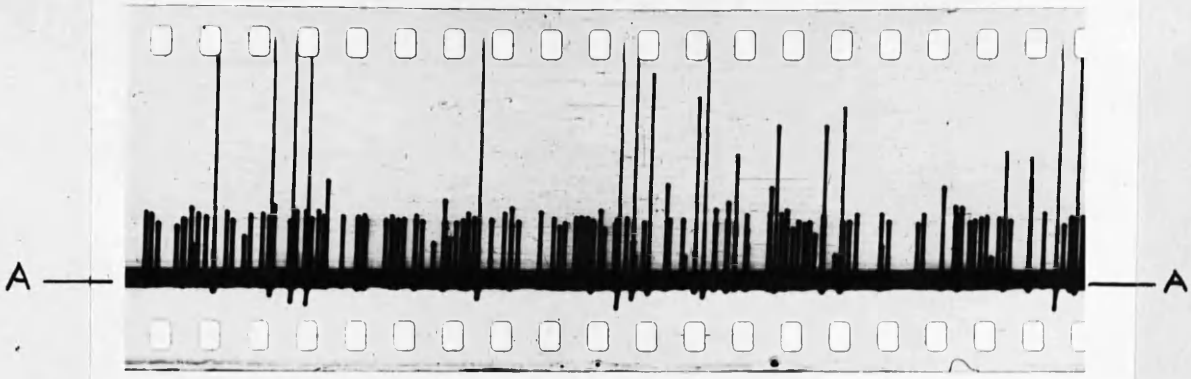
(e) Selected portions of the distribution may be obtained by screening the major portion of the tube face, allowing an analysis to be made of a small number of pulses from an intense distribution, as, for example, in the end-point film shown in Figure 4.

The cathode ray tube spot was here deflected vertically by the output pulse of the amplifier while the film (35 mm. recording film) was moved through the camera at a constant speed horizontally. A high voltage oscilloscope was used, together with a lens of aperture f/1.8; several typical traces are shown in Figure 4. In each the undeflected position of the spot is marked by the line AA; the second line, which appears above this, is caused by halo which spread beyond the semi-transparent mask over the spot.

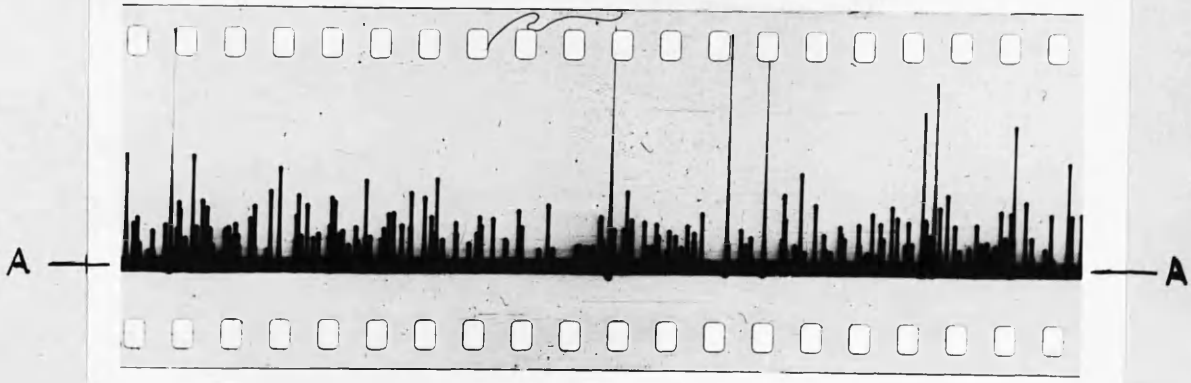
FIG. 4

TO FOLLOW PAGE 41

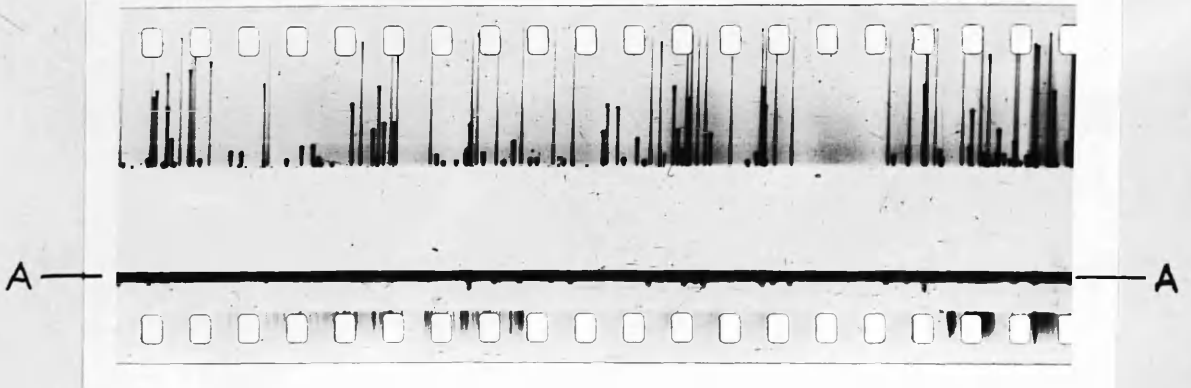
Cu X-RAYS



C^{14}



H^3 END POINT



The films were analysed by projecting them on to a screen which had 120 lines ruled on it at a spacing of $\frac{1}{2}$ cm. Each pulse is recorded in the energy interval into which it falls; while this analysis occupied a large amount of the working time, it was felt that this was justified by the high degree of accuracy which could thereby be obtained.

Throughout this work the negative high voltage supply for the counters was obtained from four-kilovolt stabilised power units designed by T.R.E. For voltages higher than 4 kV two supply units were connected in series, the transformer windings of one of them being insulated from earth by a pair of high-voltage transformers placed back-to-back. The output of these units was additionally smoothed by two or three 0.1-microfarad condensers with 2-mehohm series resistances, and provided an adequately stable and smooth supply throughout the work.

SECTION 5

PROPERTIES AND CHARACTERISTICS OF THE COUNTERS

Sensitivity to X-Rays and Gamma Rays.

These counters were designed to have a sensitivity as high as possible to electromagnetic radiation. For X-rays and gamma rays sensitivity is of prime importance and a high sensitivity value for the instrument would enhance its value considerably. In the usual Geiger tubes, sensitivities of the order of one percent make intensity measurements with weak sources difficult, and the usual alternative method employed is that of the crystal spectrometer. Both the proportional counter and the crystal spectrometer may be applied only to a limited range of energies, and, for these energies, the high sensitivity obtained from the proportional counters enables a larger amount of information to be obtained from weak sources than may have been obtained from a crystal spectrometer. A high sensitivity should also give much more accurate results in coincidence measurements.

This higher efficiency of detection is obtained by making use of the high photoelectric absorption coefficients of various gases below 150 kev, and by operating the counters at pressures around atmospheric pressure. By choosing gases or vapours which have high absorption coefficients in the range of energies involved the absorption in the gas was made as high as possible. Where possible the gamma rays or X-rays

were introduced through a thin window, and, further, the absorption of the walls of the counter was made as small as possible by using a light cathode material, such as aluminium or graphite, or by coating the copper walls of a metal counter with these materials.

The gas mixtures most frequently used contained argon at a pressure of 55 - 60 cms. Hg, together with a quenching gas; these mixtures were found to be suitable for energies between 0 and 50 kev. For example, at 10 kev, this gas has an absorption coefficient of 62, so that in a path length of 7 cms, over 40% of the quanta will produce photoelectrons. Since each photoelectron will be detected, this represents a very high sensitivity.

For quantum energies around 100 kev good sensitivity may be obtained by using gases of higher atomic number and by increasing the counter dimensions; efficiencies up to 15% can be obtained by the use of krypton and xenon in a ten cm. diameter counter. An increase in the size of the counters was also demanded by the following considerations. The photoelectrons released in the gas must have a high probability of completing their ionization process inside the counter if an accurate determination of their energy and that of the quantum is to be possible; therefore the dimensions of the counter must be large with respect to their range. Also, since the photoelectrons produced in the wall will emerge with a variety

of reduced energies, it is desirable that their effects should be small; clearly, therefore, the gas absorption must be made as high as possible if accurate energy measurements are to be made.

As a rough working rule the size of the counter was never less than twice the range of the photoelectrons under study and was usually as large as practicable.

Uniformity of Multiplication.

For accurate energy measurements it is necessary to make the variations in pulse size as small as possible. Since the gas multiplication process depends critically on the value of the field strength close to the surface of the wire, good uniformity could be obtained only if wires of uniform radius were used and if they were accurately centred along the axis of the tube. Care was taken to use wires of low tolerance and to avoid careless alignment; microscopic analysis of the wires showed that they were free from prominences.

At the ends of the active volume of the counters the electric field varied in magnitude and direction due to the close proximity of the earthed shield. This resulted in a non-uniformity of gas multiplication in these regions, and this effect was studied in the specially designed counter, part of which is shown in Figure 2. This counter had a series of thin windows at intervals of one centimetre along its length through which the fluorescence radiations of copper were

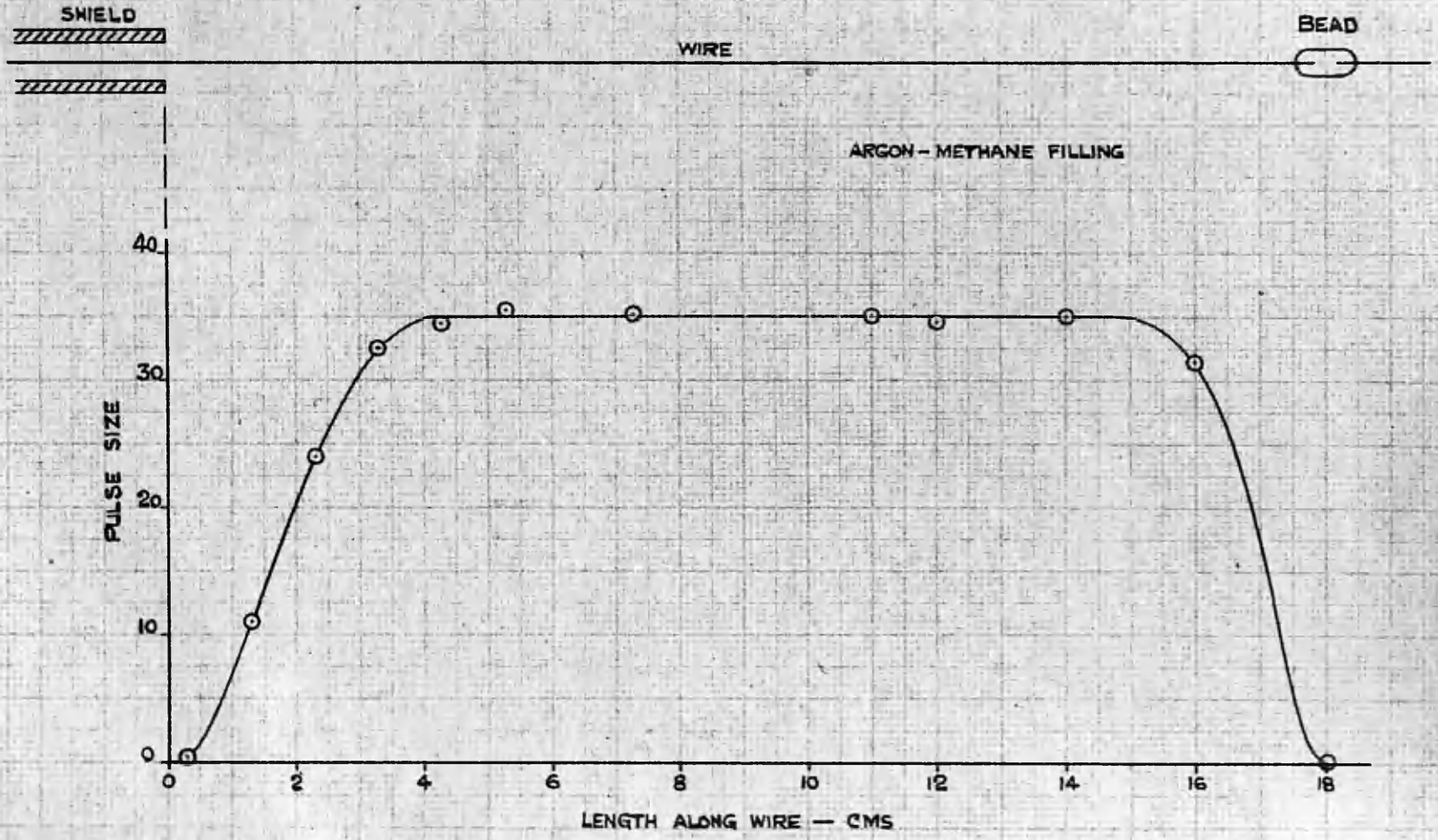
admitted. The average pulse height of these X-rays was obtained at various positions and these are shown in Figure 5. Since these were obtained in identical conditions they may be considered to represent the values of the gas multiplication, and it will be seen that they fall off very rapidly close to the end of the wire. Since this distance is determined by the radius of the screening electrode and, since most of the metal counters had the same radius, the end-effect was considered to be the same to the first order. The effect could probably be reduced by using a narrower screening electrode, but this is difficult.

It is important to note, however, that in this method the windows, and therefore the widths of the X-ray beams are finite: this may cause some smoothing of the end-variation which may consequently be slightly sharper than that indicated here.

From the studies of the end-effects of Geiger-Muller tubes published by other authors[18] it would appear that the variations are similar to those observed here. In the Geiger tubes the counting efficiency is reduced and at the same time there is a dependence of the counting efficiency on the voltage. For these tubes, however, the effect may be neglected in the measurement of X-rays and gamma rays from an external source, since the photoelectrons will be confined to a region near the window of the counter, that is, in a region

FIG. 5

VARIATION OF PULSE SIZE ALONG WIRE



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where multiplication is uniform. For work with beta rays using an internal source, this end-effect must be known, and it is important to use counters which are as long as practicable. Alternatively, beaded counters may be used and these will be described in the relevant sections.

The wide variation in field strength in the counters between the central wire and the cathode surface necessitates some check upon the uniformity of multiplication of ionization produced at various radii. In every experiment in which fluorescence X-rays are used for calibration purposes these X-rays form photoelectrons with approximately equal probability across a complete diameter of the counter. If these X-rays produce a good homogeneous group of pulses, as is observed in the majority of cases, the radial variation of multiplication may be assumed to be very small. Those cases in which inhomogeneity of X-ray pulses may not be explained by range or gas mixture considerations indicate a failure to collect the full number of ions near the circumference. It is then necessary to adjust the working conditions until such uniformity is observed.

Dependence of the Output Pulse Size on the Energy of the Radiation.

The average output pulse sizes for radiations of various energies were investigated by the following method for counters filled with various gas mixtures and operating at

various gas amplification values. An industrial A.C. X-ray tube used to give a beam of white X-rays of 50 kev maximum energy; this beam was allowed to fall on to a thin scattering foil and the scattered radiation was collimated to enter the window of the counter at right angles to the direction of the initial beam. This scattered radiation was known to consist principally of the fluorescence X-rays of the scatterer and, by using foils of various materials from calcium to barium, groups of X-rays could be admitted with accurately known energies between $\sim$ 3 kev and 40 kev. It was found that the K_α and K_β radiations could be resolved and it was therefore possible to examine the variation of pulse size with X-ray energy to a high degree of accuracy in this region. The histograms for manganese chloride, calcium and copper are given in Figures 6 and 7; these show a clear resolution between the K_α and K_β lines of each element. The appearance in Figure 6 of the K-radiations of both manganese and chlorine is of particular interest, since a calibration may thereby be made below the critical absorption edge of argon (3.2 kev). The relative intensities of the lines differ from those expected theoretically due to differential variations of absorption in the window and detection in the counter gas for the different energies; this effect does not, however, prevent the determination of the average output pulse sizes of the characteristic radiations.

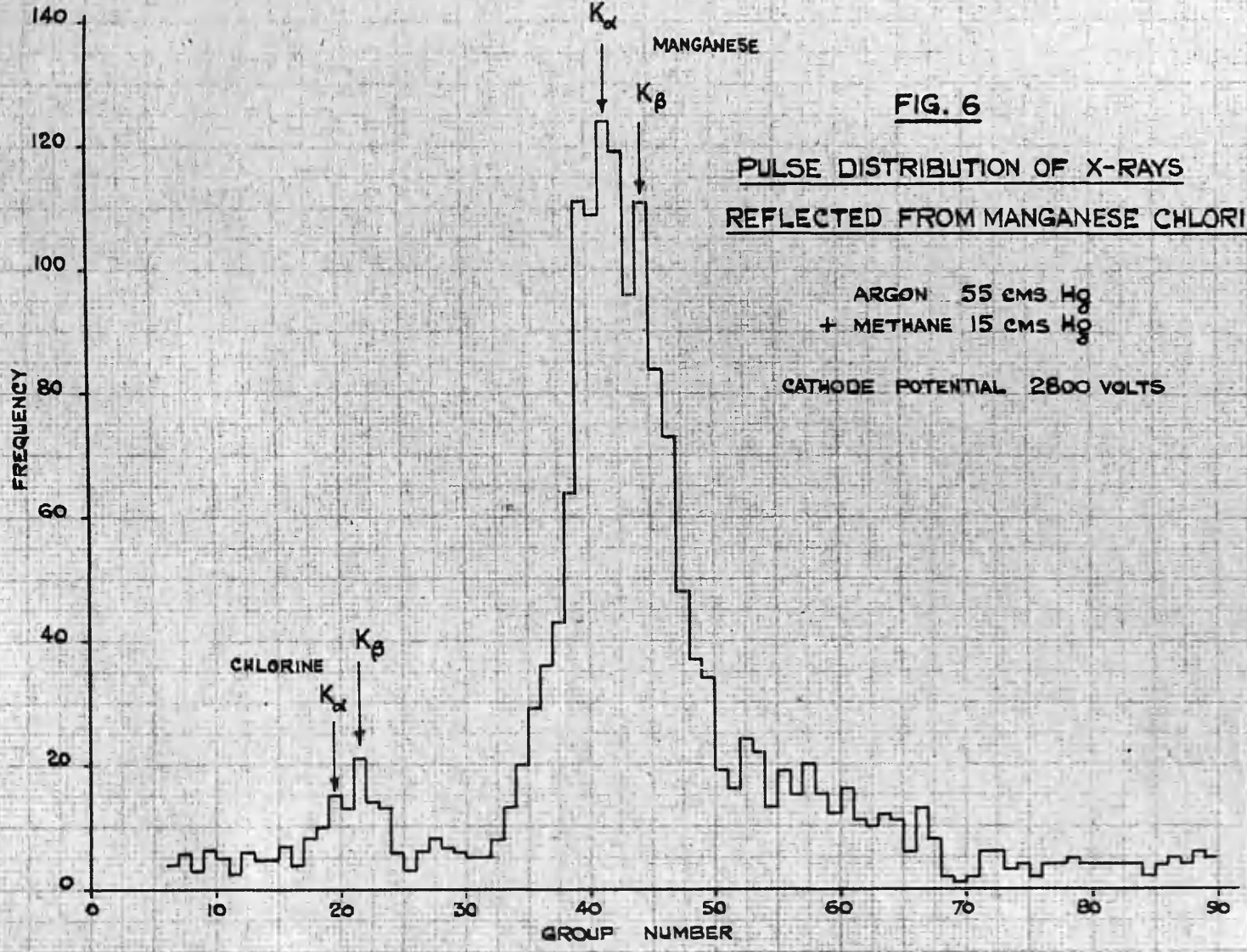

FIG. 6

PULSE DISTRIBUTION OF X-RAYS

REFLECTED FROM MANGANESE CHLORIDE

ARGON 55 cms Hg
+ METHANE 15 cms Hg

CATHODE POTENTIAL 2800 VOLTS

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FIG. 7

PULSE DISTRIBUTION OF X-RAYS

REFLECTED FROM CALCIUM AND COPPER

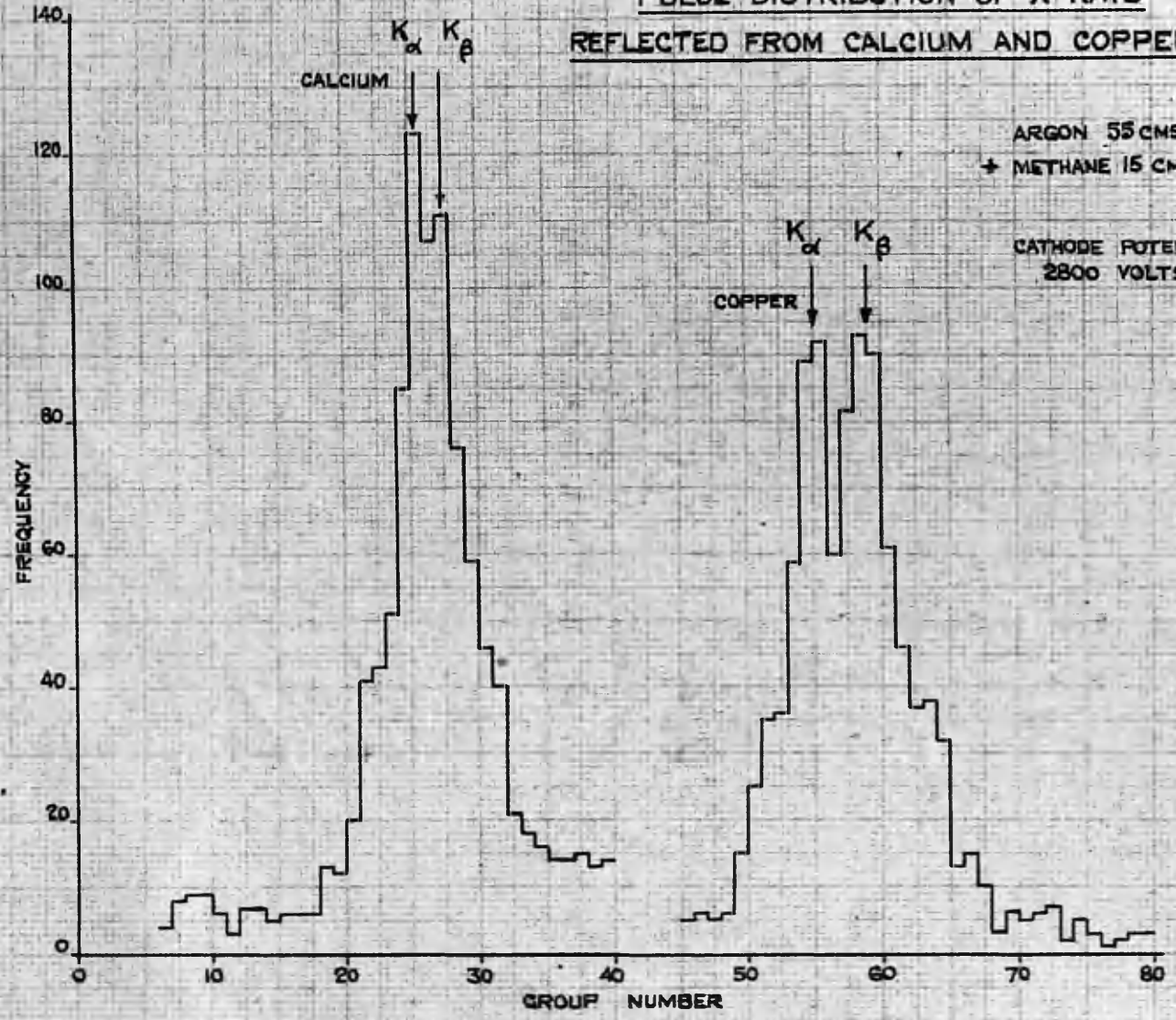

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Figure 8 gives a plot of the average pulse size against the energy of the X-rays of a number of elements for the argon - methane mixture most frequently used (60 cms argon and 15 cms methane), and it will be seen that the points lie close to a straight line passing through the origin. Since, for these energies, the majority of the quanta are absorbed in the K-shell of argon, they produce initially an electron with an energy 3 kev less than the energy of the quantum; the remaining energy, which might appear as secondary radiations, must be converted into further photoelectrons in almost every case. The Auger effect in argon is 93%[19] so that a very high proportion of the absorption events lead to the release of a number of photoelectrons with a total energy very close to the energy of the quantum; in the remaining cases, where a K X-ray of argon is emitted, they are rapidly converted in the neighbouring atoms of the gas, due to their high absorption coefficient in the M-shell. Therefore very few of the ionization events are expected to give rise to photoelectrons which do not sum up in energy to very nearly the energy of the quantum.

The results obtained for the radiations below 10 kev in energy are given in Figure 9. These show a good degree of linearity.

A similar investigation was carried out in a mixture consisting of nitrogen (60 cms Hg), methane (7 cms Hg) and

FIG. 8

DEPENDENCE OF PULSE SIZE ON ENERGY FOR NORMAL GAS FILLING

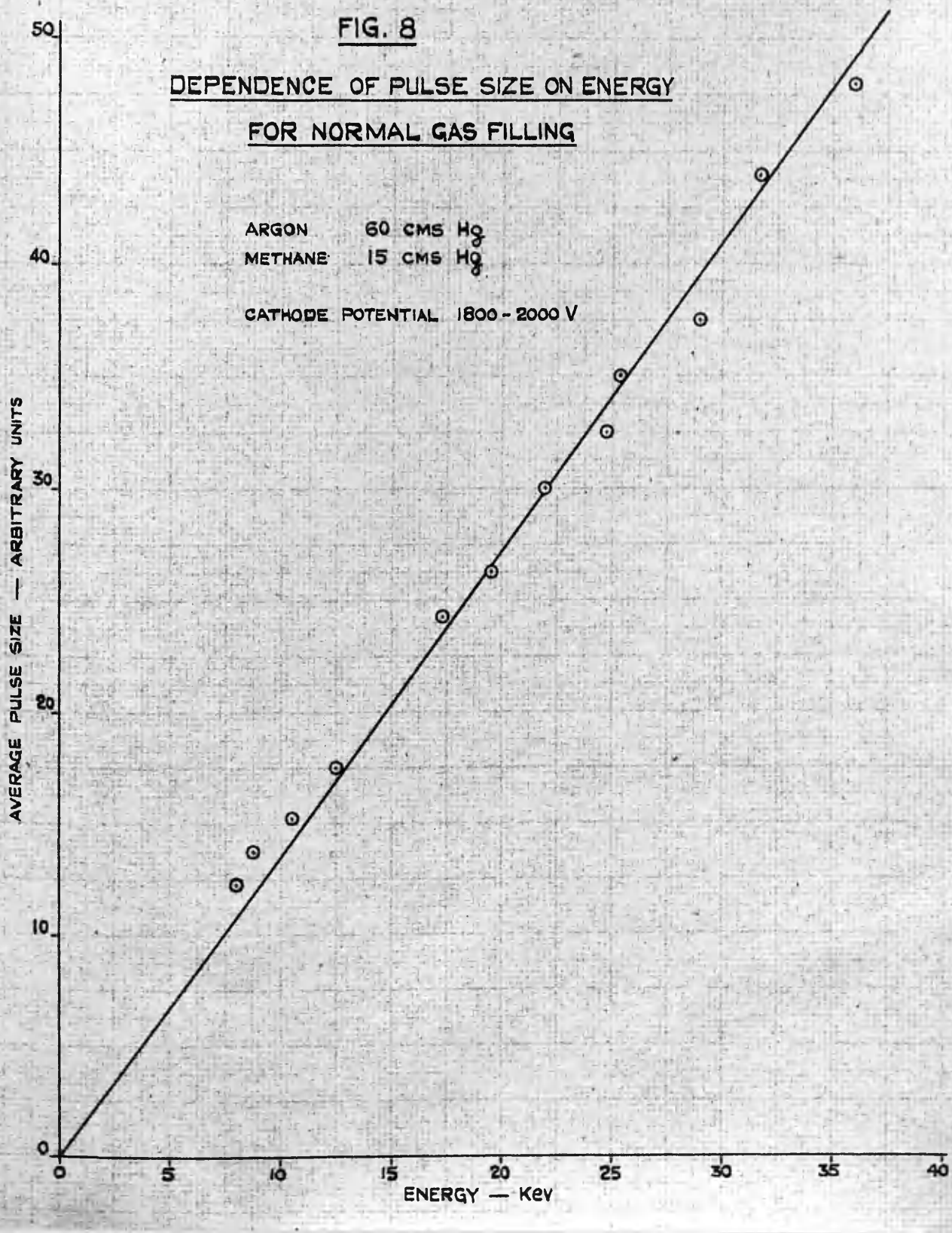
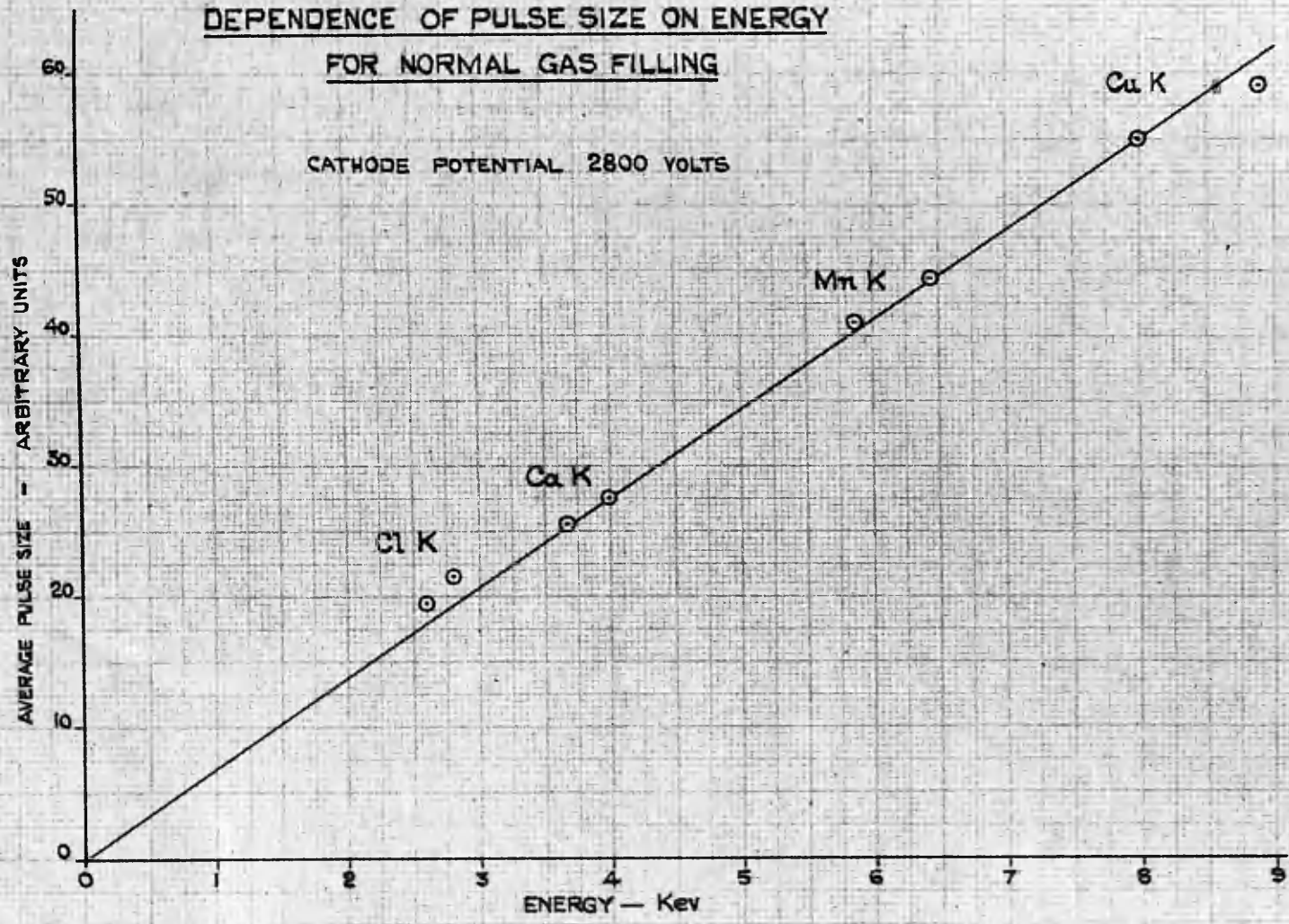


FIG. 9

DEPENDENCE OF PULSE SIZE ON ENERGY
FOR NORMAL GAS FILLING



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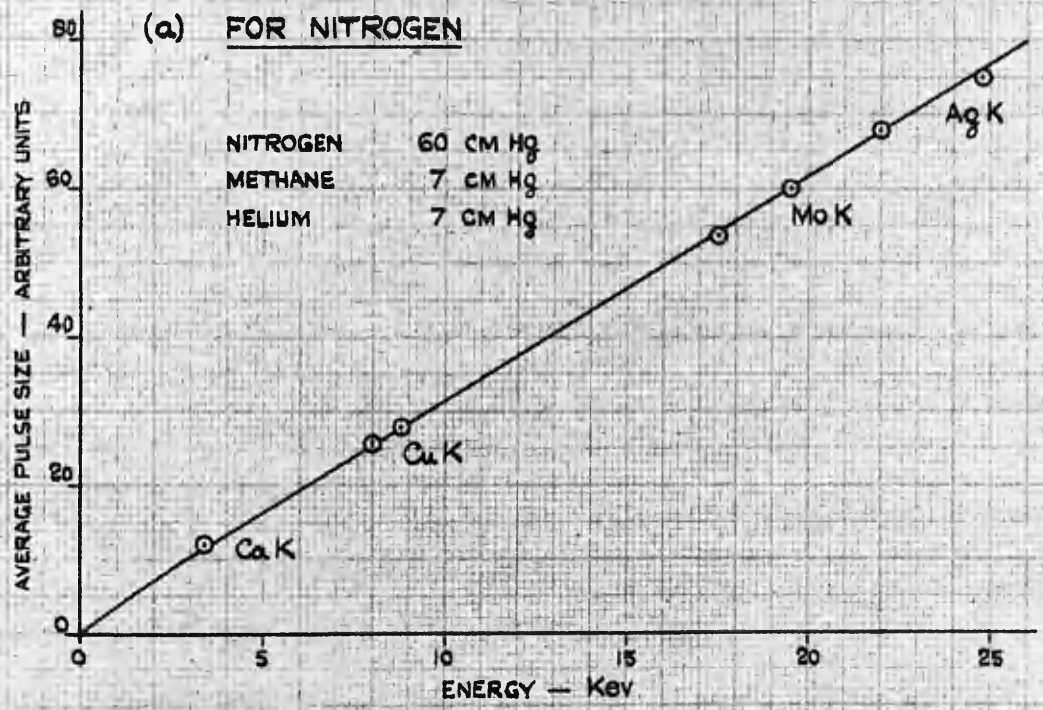
helium (7 cms Hg) and the results are shown in Figure 10. This also shows an almost direct proportionality between pulse size and energy similar to that of Figure 8; the shape of the individual distributions indicated that the detection efficiency was somewhat smaller in this case, as there were considerable pulses of smaller energies due to wall effect. An investigation was also carried out with a mixture containing methane (63 cms Hg) and helium (10 cms Hg), and the results are also given in Figure 10; this shows that the relation is somewhat curved. The X-ray absorption properties of methane seem to be very poor since the pulse distributions obtained from this mixture failed to show the clear groups obtained with argon. For example, the distribution obtained with molybdenum shown in Figure 11, includes a considerable background of pulses of all energies up to K_α and K_β peaks, now much more indistinct. While this may be caused by a relatively larger wall effect, indirect evidence appears to indicate that non-uniform distributions are obtained in this gas; this may be a severe limitation of the proportional counters already reported to have been used with this filling.

A point of more fundamental importance arises in the consideration of Figures 8, 9 and 10. The linearity of the relations indicates that the energy expenditure per ion pair in argon and nitrogen is sensibly constant over the range involved here. Since these measurements involved photo-

FIG. 10

DEPENDENCE OF PULSE SIZE ON ENERGY



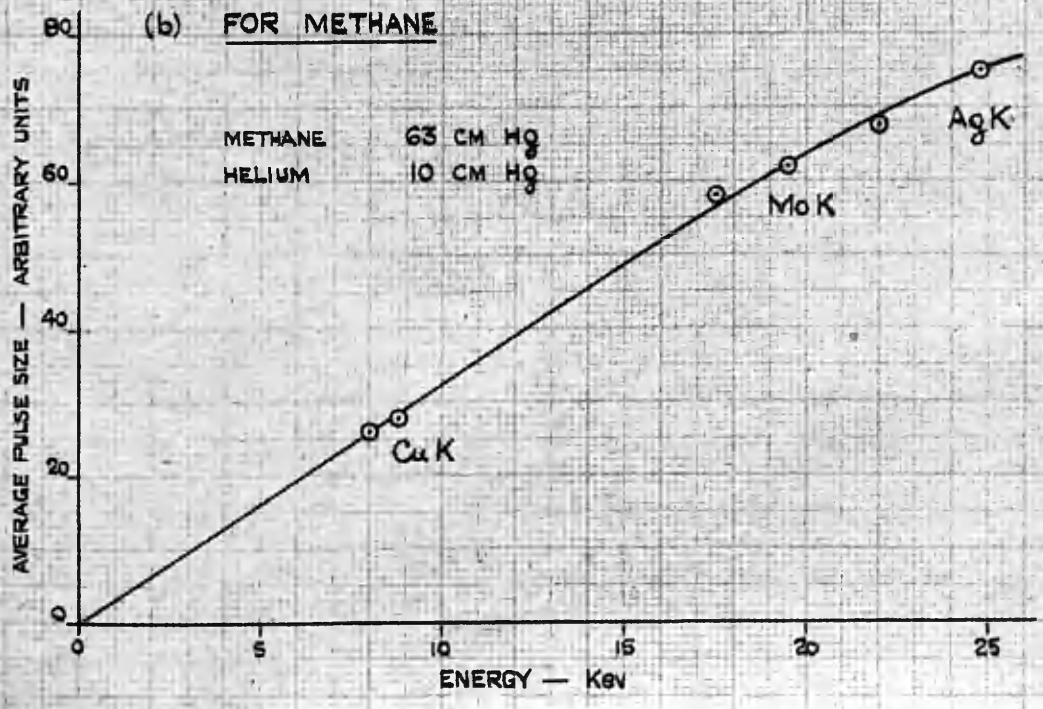
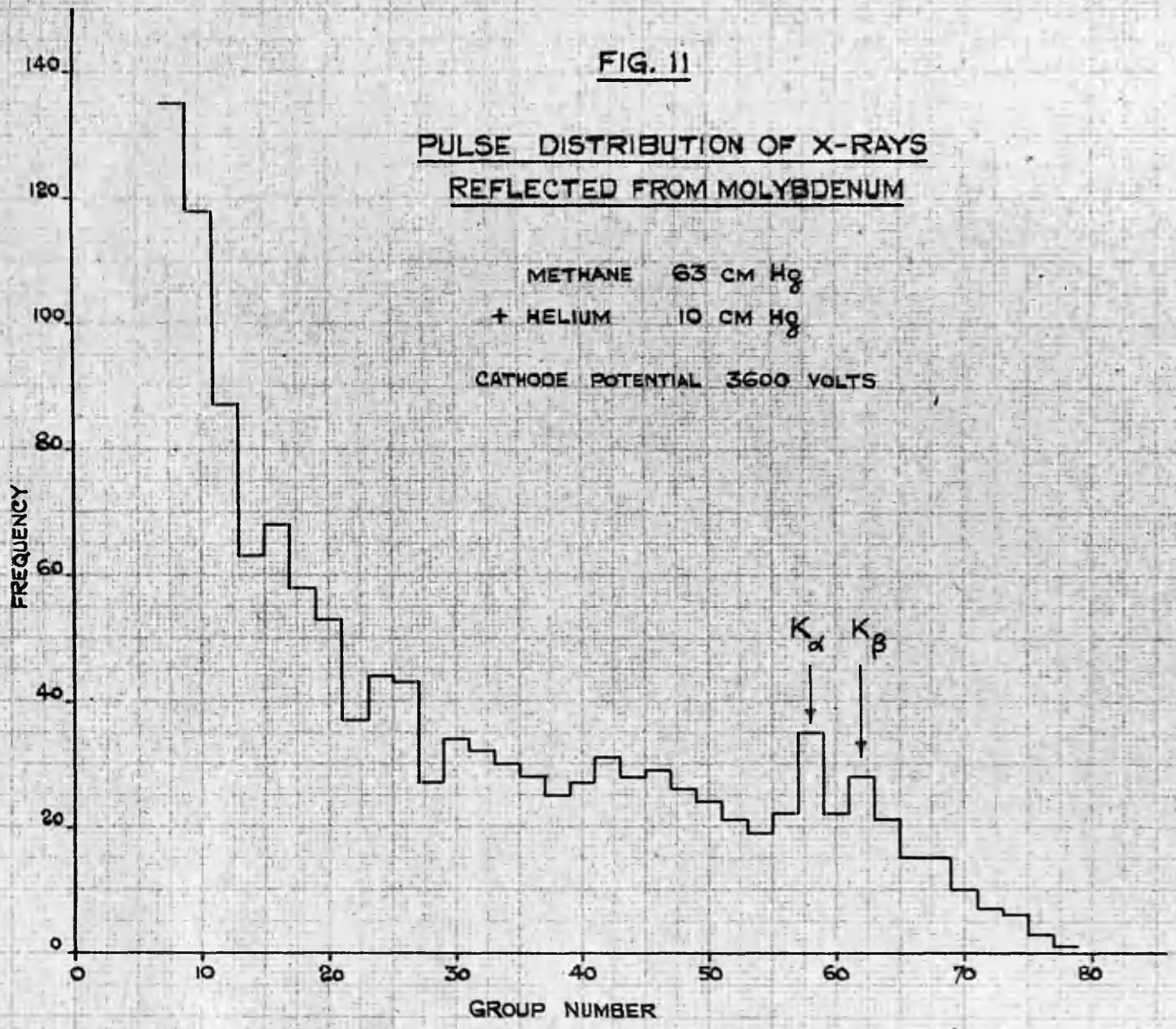


FIG. 11

PULSE DISTRIBUTION OF X-RAYS
REFLECTED FROM MOLYBDENUM

METHANE 63 cm Hg
+ HELIUM 10 cm Hg

CATHODE POTENTIAL 3600 VOLTS



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electrons from a few hundred volts to about 40 kev in energy, strong evidence is afforded of the linearity of energy and the number of ion pairs produced over the whole of this region. This supports the indirect evidence put forward by Gray[20] for the constancy of energy expenditure in the noble gases. The linearity in nitrogen indicates that the variation of energy expenditure in air observed by various investigators[20] is due almost entirely to the oxygen content.

The non-linearity of methane is not unexpected; in polyatomic gases electrons will lose energy very frequently in the excitation of vibrational and rotational states, and these losses may vary considerably with energy.

The method used here could readily be adapted to a systematic study of the energy expenditure per ion pair in a large number of gases, particularly with quanta of even lower energies.

Identification of Radiation.

As an illustration of the discrimination available in this counter a study was made of the fluorescence X-rays of copper (Z = 29) and nickel (Z = 28). The pulse distributions for these are given in the top two curves of Figure 12, the lowest curve being the pulse distribution of the electromagnetic radiation of radioactive copper. The latter decays by three alternative processes:-

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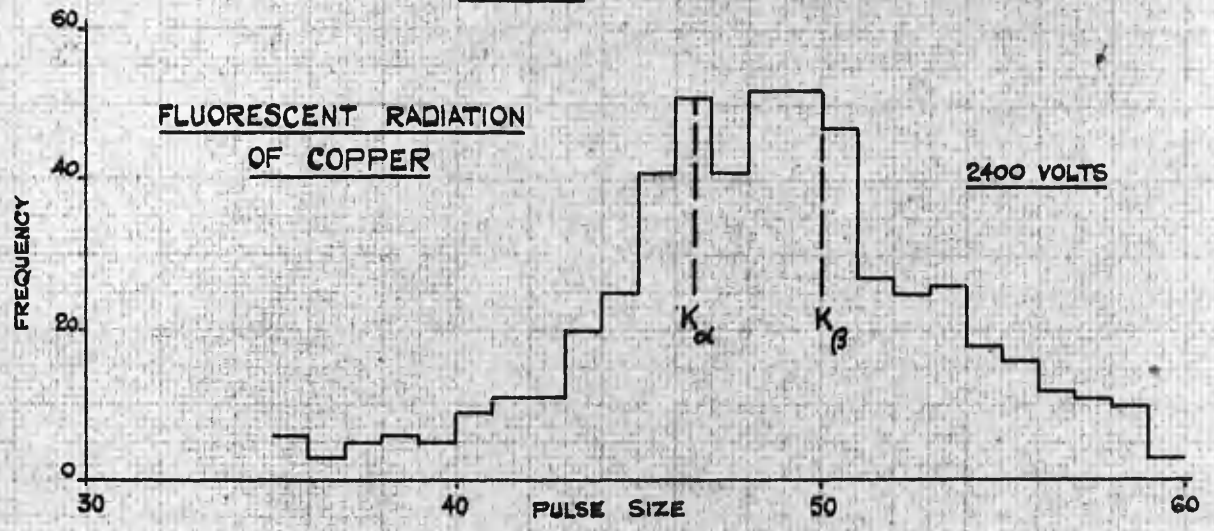
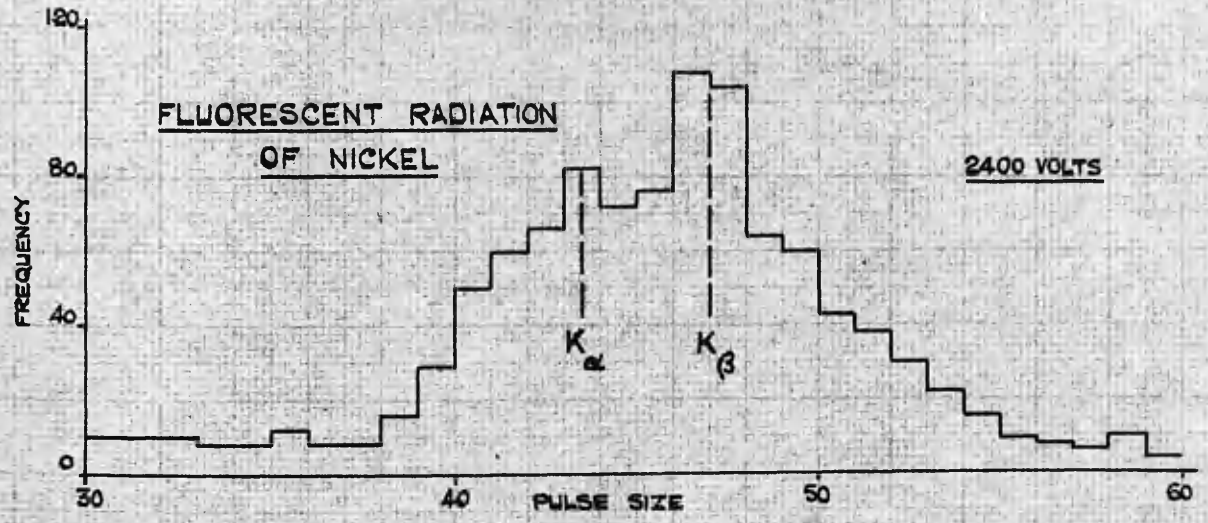
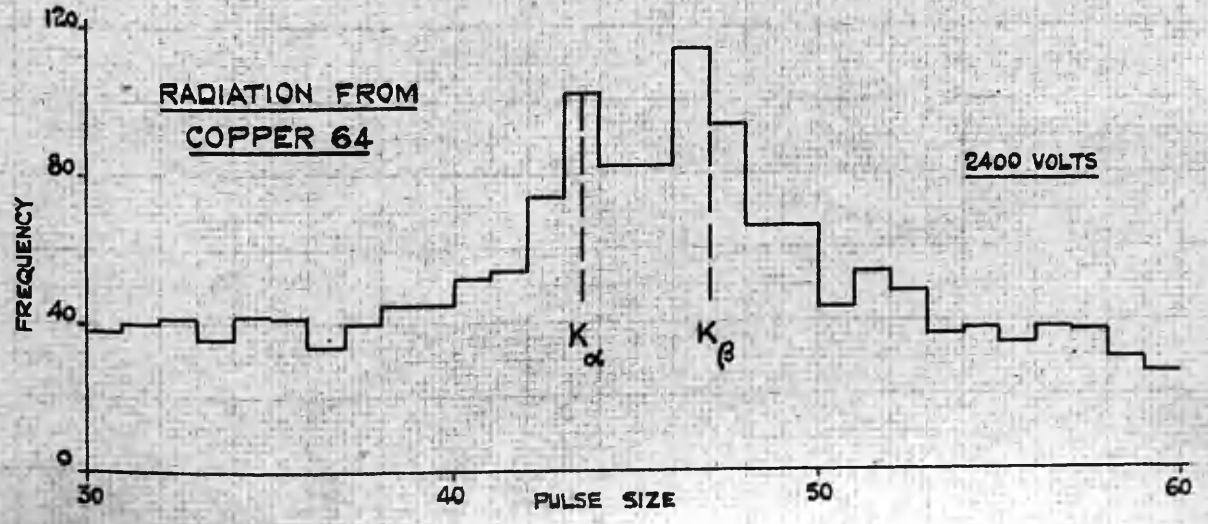
FIG 12







$$Cu^{64} \rightarrow \begin{cases} Ni^{64} + e^{+} \\ Zn^{64} + e^{-} \\ Ni^{64} + \text{K-capture} \end{cases} + \text{gamma rays}$$

The radioactive source was covered with polythene of sufficient thickness to absorb all the electrons and positrons; the peak due to the X-radiation resulting from K-capture in the source is therefore superimposed on the pulses due to:-

(1) Annihilation quanta of 0.51 Mev,

(2) Gamma quanta of energy 1.3 Mev,

and (3) The natural background of the tube.

A very cursory analysis shows, however, that the radiation corresponds exactly with the K X-rays of nickel, and that the resolving power of the instrument is quite adequate to separate it from that of copper. In a similar manner it is possible to differentiate between L X-rays of neighbouring elements, as was done in the case of radium D which will be described later.

Resolving Power.

It has been shown that the resolving power of the instrument was adequate to differentiate between the X-rays of neighbouring elements. A more rigorous analysis involves consideration of the experimental widths of the lines measured, and their correlation with the statistical fluctuation of the number of ion pairs formed in the gas and of the gas amplifi-

cation.

It is concluded from the histograms of various lines that the full width at half amplitude of a distribution due to homogeneous radiation producing an average initial number of ion pairs, n, which may be expressed as α $A\sqrt{n}$, where A is the gas amplification, and α has values between 1 and 2. Since the average pulse size is An, the experimental variation may be expressed as:-

$$h = \alpha \sqrt{1/n}.\ h,$$

where h is the full width at half amplitude. As this variation is some three times larger than the root mean square value in definition, it is clear that the experimental variation is considerably smaller than the value given by Snyder's analysis, which assumes that the initial fluctuation is $\pm \sqrt{n}$. It is shown in Appendix 1 that the variation produced by the gas amplification is small and it is therefore necessary to assume that the initial variation is considerably less than $\pm \sqrt{n}$. This is not surprising since in most of the gases used all the energy of the particles will be absorbed in ionizing processes and dissociation processes. The probability that an electron will form an ion pair is therefore an involved expression of the energy in each gas, and in many gases the only process by which an electron may lose an appreciable amount of energy is the production of ion pairs. It cannot therefore be assumed that, if n is the

number of ion pairs produced on the average by an electron of energy E, i.e., if $n = E/E_0$, where E_0 is the energy required to produce one ion pair, the probable variation will be $\pm \sqrt{n}$, but rather that it will depend on the fluctuation in E_0; in many cases this may result in a much smaller variation.

The fluctuation in amplification will depend on the nature of the gas used and the value of the gas gain A, since these will control the number of mean free paths and their variation. It will also be related to the number of ion pairs produced, since this determines the final number of ion pairs collected.

The role of the various factors may be studied in detail with this apparatus.

Variation of Gas Gain with Voltage.

Various experimenters have reported investigations of the variation of gas amplification with cathode potential for different gases. A short investigation was carried out here with the usual argon-methane mixture and with nitrogen, in order to ascertain the approximate values of the gas gains being employed in the initial experiments. The counter used had a cathode diameter of 6 cm and a wire diameter of 0.004 in; the average pulse size at each voltage setting was determined (a) for the 47 kev line of radium D at low voltages and (b) for copper X-rays at higher voltages. It was thereby

possible to cover a considerable voltage range without altering the gain of the amplifier. Figures 13 and 14 give the variation of log. A with potential for argon and nitrogen; for the latter only a relative scale is used; for the former the gas amplification at 1400 volts was assumed to be unity since no appreciable decrease of the pulse size was observed as the voltage was lowered below this value. It is clear that the gas gain increases exponentially for both gases, in agreement with the theory, except for a small region near the threshold.

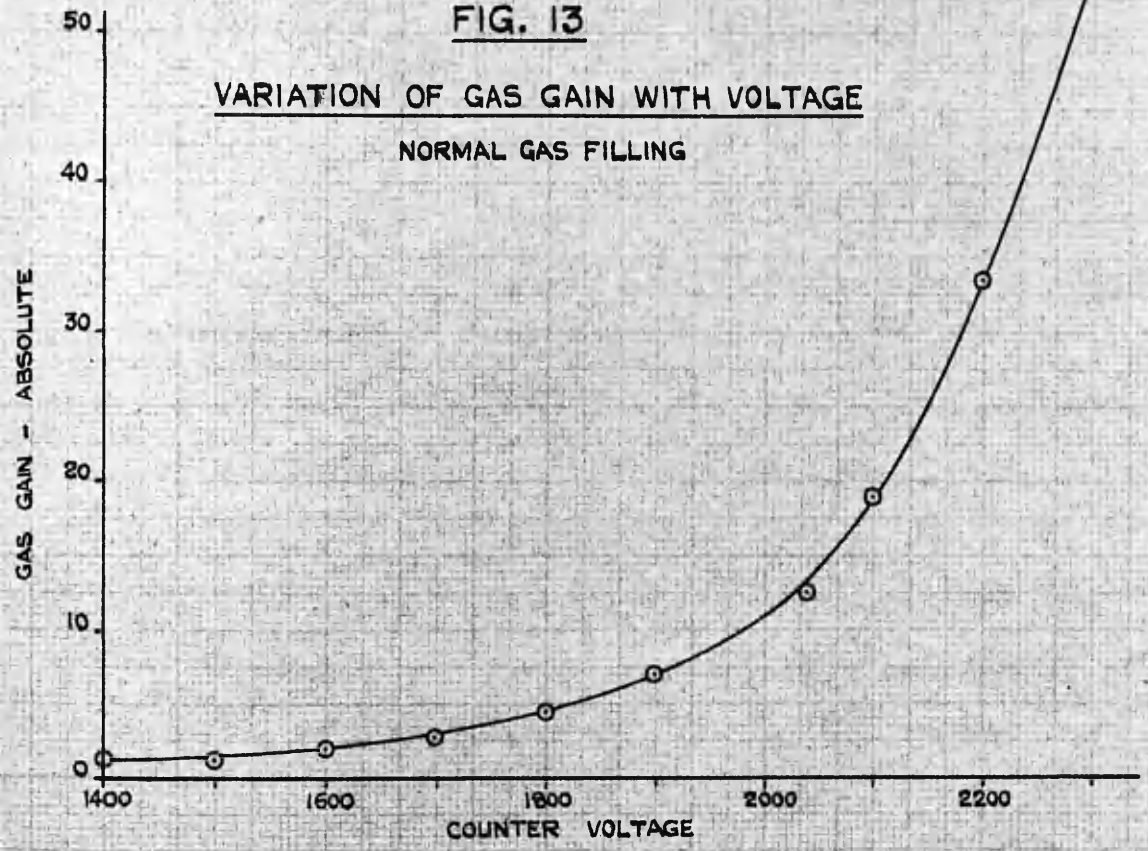
Because of the considerations regarding statistical fluctuations already discussed the gas gains used in the analyses of β - and γ -spectra were normally greater than 15 - 20, while, for considerations of linearity, they were usually less than 100; exceptions, principally the investigations of very low energy components, are noted in the text.

(18) K. Greisen and N. Nereson, Phys. Rev., 62, 316, (1942)
(19) A.H. Compton and S.K. Allison, "X-Rays" (Macmillan, 1936)
(20) L.H. Gray, Proc. Camb. Phil. Soc., 40, 72, (1944)

FIG. 13

VARIATION OF GAS GAIN WITH VOLTAGE

NORMAL GAS FILLING



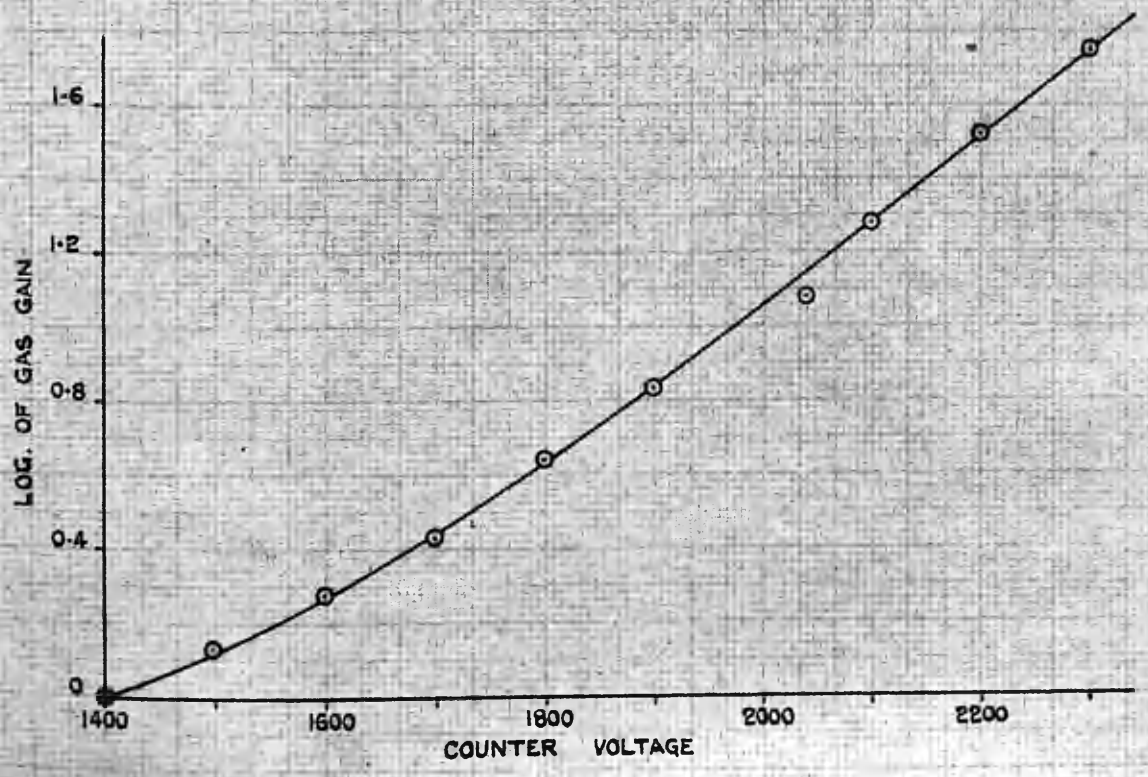
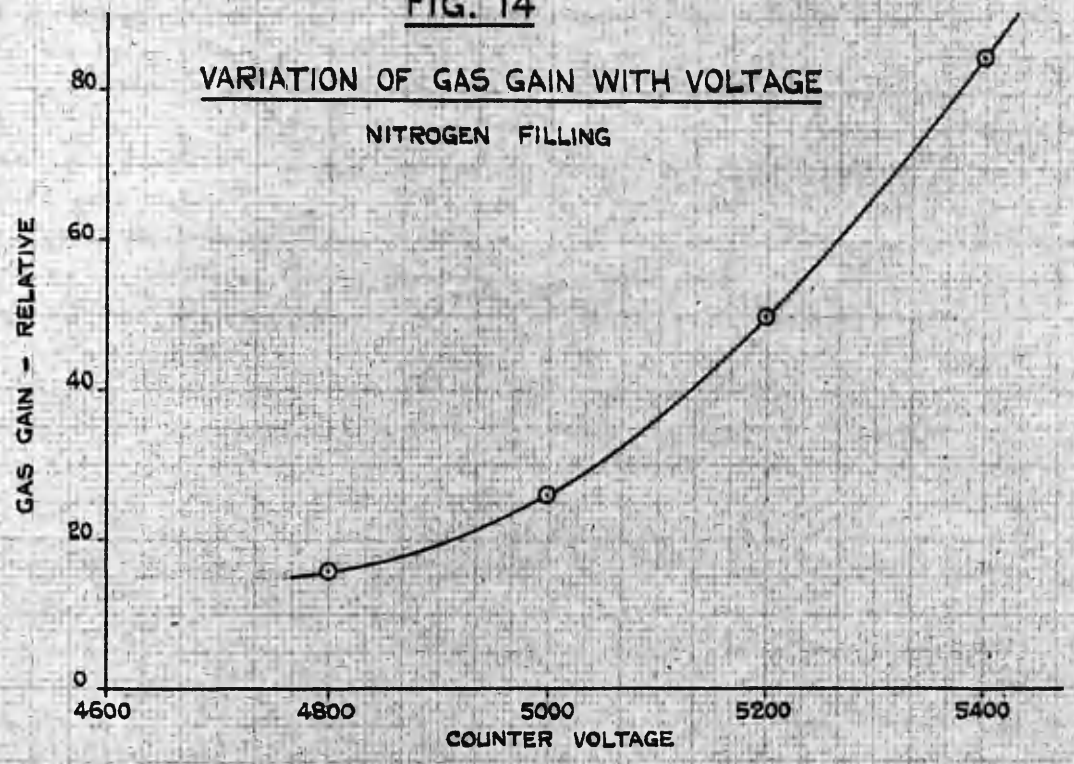
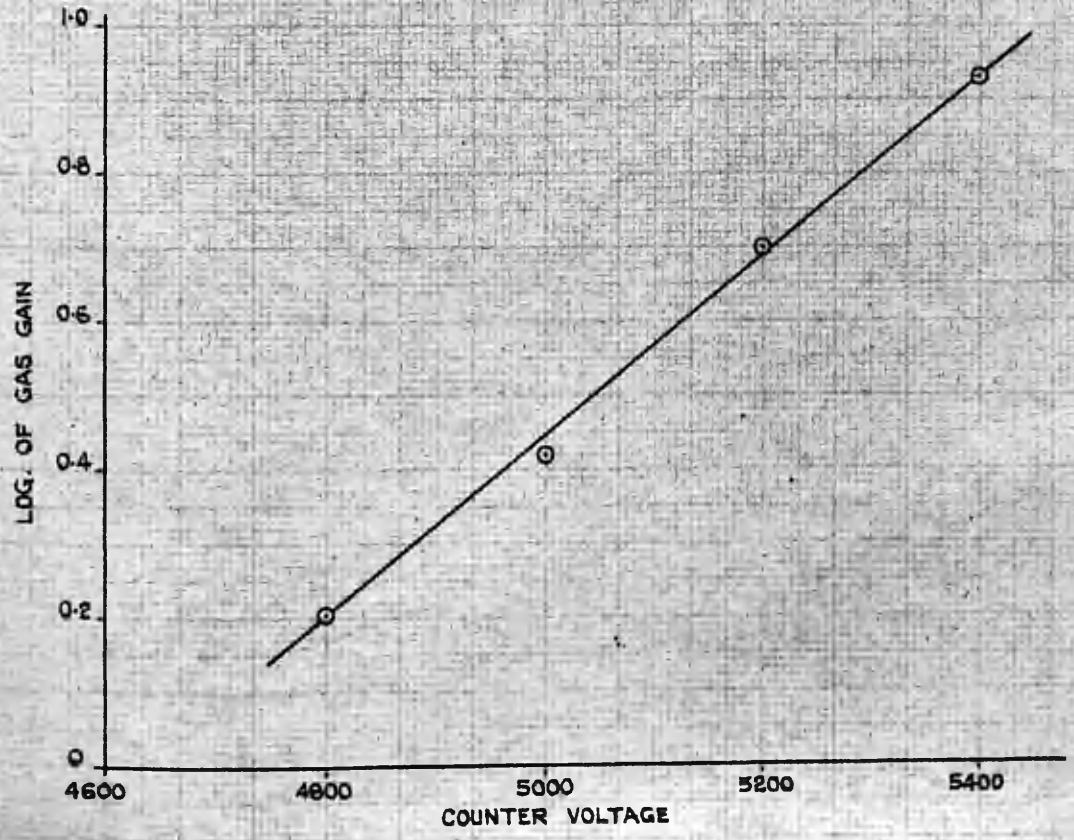


FIG. 14

VARIATION OF GAS GAIN WITH VOLTAGE

NITROGEN FILLING





SECTION 6

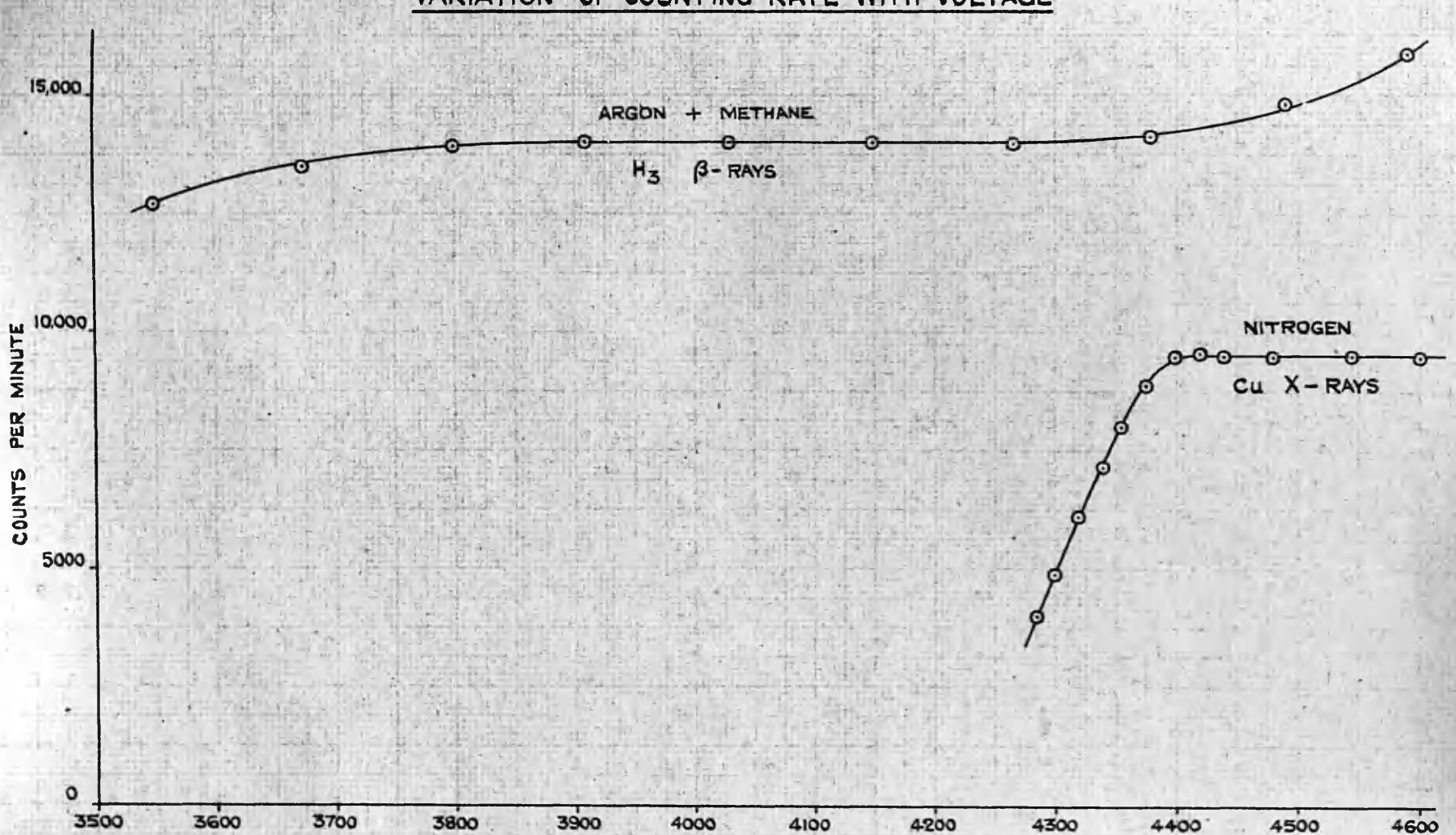
QUANTITATIVE DETECTION OF RADIATION.

It has been shown[17] that proportional counters exhibit a remarkably uniform plateau of several hundred volts for the detection of heavily ionizing particles, and they are frequently used for quantitative studies of source radiations. A study was therefore made of the plateau obtained in these counters for several gas mixtures, using lightly ionizing radiations. It has been shown that the formation of a few ion pairs in one of these counters operating at a gas gain of 10^3 gives a pulse sufficiently large to allow it to be registered above the noise of the amplifier. A counter of 40 cm. active length and 6 cm. diameter was filled with argon (60 cm. Hg) and methane (15 cm. Hg) together with a mixture of hydrogen and tritium to a pressure of 5 mm. Hg. This counter had been used for one of the measurements on tritium and the counting rates were determined with a scaler for different voltages, all of which were within the strictly proportional region. The results of this experiment are shown in Figure 15; it is clear that the plateau is extensive and the slope (0.005% per volt) is much smaller than that of a Geiger counter. Since the tritium spectrum consists of electrons of all energies this provides a very satisfactory test of the counting properties of this arrangement.

The plateau is limited on the low voltage side by

FIG. 15

VARIATION OF COUNTING RATE WITH VOLTAGE



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loss of counts of particles of low energies, and on the upper side by the appearance of extraneous effects, such as the liberation of photoelectrons at the wall or in the gas.

This experiment was repeated with nitrogen, in this case using X-rays of copper; an appreciable number of the photoelectrons will be released at the walls and will therefore have energies varying from a few electron volts to 8 kev. The results of this investigation, also shown in Figure 15, again indicate a plateau of very nearly zero slope and extending over several hundred volts. These results show:-

(1) That if a radiation produces more than 100 ion pairs complete detection will be obtained,

(2) That proportional counters may readily be used for the quantitative measurement of soft radiations in a manner exactly similar to the Geiger tube, and the application already discussed to high energy radiations,

and (3) That nitrogen filled proportional counters may be used with advantage to replace Geiger counters in the detection of cosmic rays and fast beta-particles, since these will, in a path length of several centimetres, give sufficient ion pairs to ensure complete detection; a nitrogen filled counter would be free from all the difficulties associated with the finite counting life of the conventional Geiger mixtures of argon and organic vapours.

Applications of these properties will be described

in later sections, and extensions of the investigations described in the preceding section discussed.

SECTION 7

THE BETA SPECTRUM OF TRITIUM

Tritium (H^3) was the first radioactive source to be investigated in the series of experiments on the low-energy beta-emitters. Such an investigation is ideally suited to the proportional counter technique since (a) the maximum energy of the beta-particles is sufficiently low to allow counters of quite small dimensions to be used, and (b) the source may be introduced as a gas into the mixture of the counter. With regard to (a) it is possible to neglect the effect of the limitation of the counter dimensions if diameters greater than 5 cm. are used; with regard to (b) the introduction of the source into the counter as a gas overcomes the difficulties associated with the more normal spectrographic technique. These difficulties are, firstly, the finite thickness of the source and its backing, and, secondly, the absorption of the thin window dividing the main body of the spectrograph from the detector. These at present produce unsurmountable difficulties with a source having a maximum energy less than 20 kev.

The spectrum of tritium is of considerable importance for several reasons:-

(1) The relatively simple structure of the ${}_1H^3$ nucleus and its undoubted assignment to an allowed transition makes possible a crucial test of the Fermi theory of beta-decay.

(2) The unusually low energy of the spectrum makes possible an accurate estimation of the mass of the neutrino,

(3) The mass difference between $_1H^3$ and $_2He^3$ may be determined with great accuracy.

Previous Results.

Previous determinations of the end point of the spectrum (E_o) by various methods gave values ranging from 9.5 kev to 18 kev.

Libby and Lee[21], using a magnetic deflection method, obtained a value of 13 ± 5 kev.

Alvarez and Cornog[22], using an ionization chamber, measured the ionization per unit volume and obtained a value of 18 kev.

Brown[23] measured the range of the beta particles in helium as 0.23 mg/cm^2, from which he calculated a value of 9.5 kev. O'Neal and Goldhaber[24] measured the range of the beta-particles in an argon-alcohol mixture with a screen-walled counter as 0.46 ± 0.05 mg/cm^2, from which they calculated an energy of 15 ± 3 kev; it was later shown[25] that the difference between these last two measurements was due almost entirely to differences between the range-energy relations used, and that the second result was more accurate.

Watts and Williams[26] used a method in which the beta-particles were accelerated through the window of a counter; by first accelerating thermionic electrons through

the window its stopping power was obtained; the value which was obtained for the end-point was 11 ± 2 kev.

In each of these methods, solid sources were used and the shape of the spectrum could not therefore be determined. Neilsen[27], using a drop count method in a cloud chamber containing a tritium gas source, obtained an average energy value of 6.5 kev and an end-point of 14.5 kev. As Neilsen pointed out, these values were in disagreement with the shape of a Fermi distribution, and it is unfortunate that this determination included only 108 tracks.

Novick[28] reported a recent measurement of the half-life of $_1H^3$ in which the amount of helium produced by a known amount of tritium in some 200 days was determined quantitatively. This measurement, which was confirmed by the ionization chamber results of Goldblatt[29], left little doubt that the half-life was close to 12 years, and indicated that the transition was allowed.

Konopinski[30] has shown that a theoretical discrepancy exists between the measured half-life and end point energy for $_1H^3$ when they are compared with the corresponding values for the nearest electron emitting nucleus, $_2He^6$. The calculations indicated that, if neither of these quantities had been underestimated, the discrepancy could only be resolved by assuming a neutrino with mass lying between 1/30 and 1/45 of the mass of the electron. This assumption

could not be proved or disproved since the shape of the spectrum was not known. The results of Cook, Langer and Price[31] on the energy distribution of the beta particles of S^{35} indicated a neutrino mass of less than 1/100 of the mass of the electron. This work was directed to give an accurate determination of the shape of the spectrum and as an attempt to resolve the difficulty by a more accurate determination of the end-point energy.

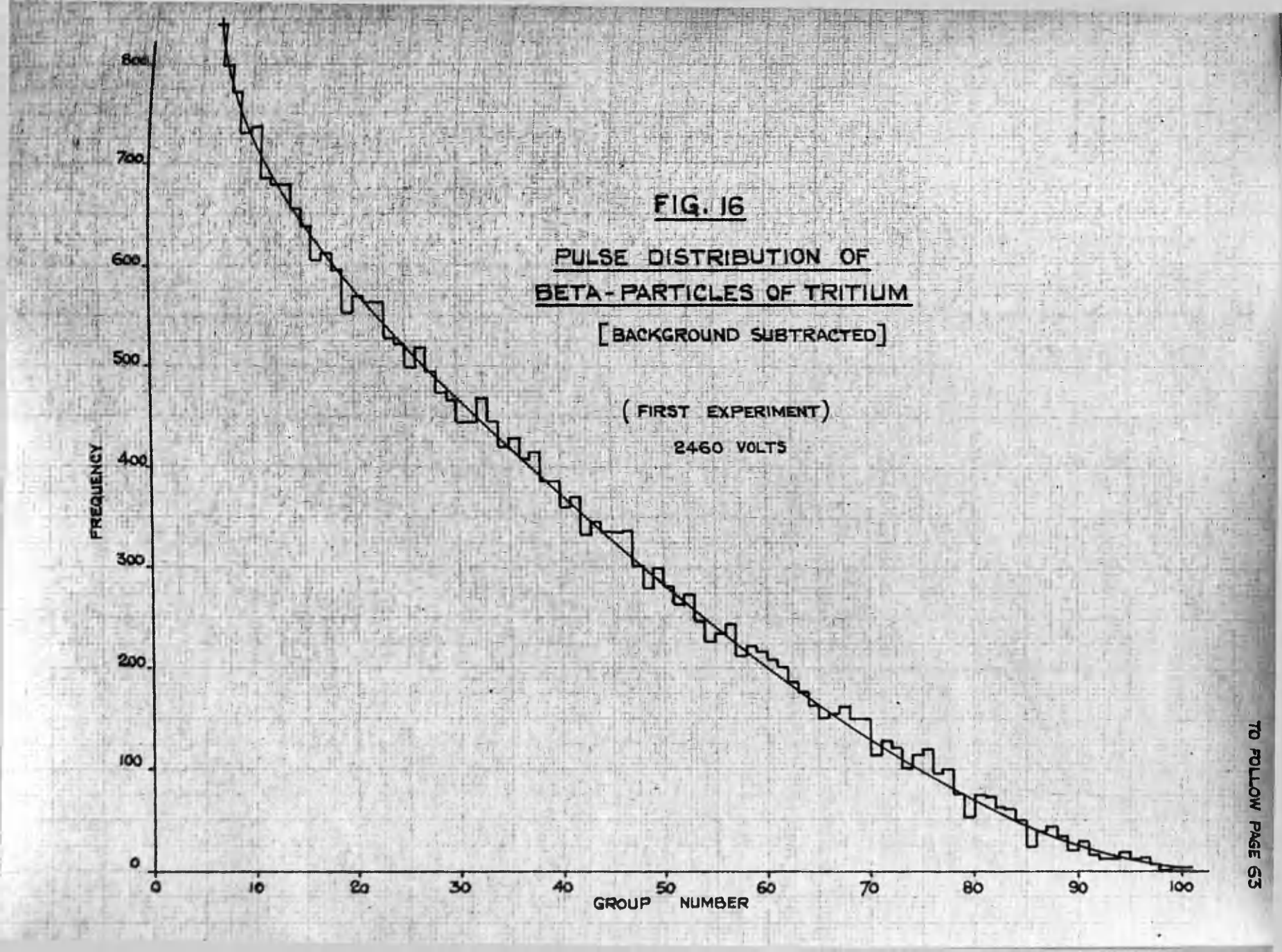
Experimental Results.[32, 33]

In these measurements, which were carried out with several counters of different dimensions, the tritium was introduced into the gas mixture of the counter, which contained also argon at a pressure of 60 cm. Hg and methane at a pressure of 15 cm. Hg. In each case the method of filling was the same; the counter was first evacuated and about 1 cm. of inactive hydrogen was introduced; this was allowed to stand for about 30 minutes in order that any absorption of hydrogen by the walls would be completed. A few grams of palladium containing the active hydrogen was then heated in a side tube connected to the system and the gas released diffused into the counter. Heating was continued for about 15 or 20 minutes and the side arm was sealed off. The normal argon-methane mixture was then added.

Counting rates of the order of 5,000 to 10,000 per min. were obtained; it was necessary, however, to check that

the active gas was not being absorbed in the walls or metal parts of the counters, since this would have resulted in considerable distortion of the spectrum. It was verified that the gas could readily be removed from the counters since the normal background counting rate was obtained after they had been evacuated and refilled with inactive gas. Further, it was found that the gas could easily be transferred to an evacuated counter, and that such sharing resulted in activities proportional to the active volumes of the counters and independent of the surface areas exposed.

The initial experiments were carried out with a glass counter of 6.7 cms. diameter and an effective counting length of 12 cm. Only one investigation will be discussed, since it was subsequently found that the counter was of such short effective length that the end-effect already discussed in section 5 resulted in a considerable distortion of the spectrum. An analysis of some 40,000 pulses from the tritium source was made under conditions which allowed energies between 900 ev and 18 kev to be covered and the results of this analysis are given in Figure 16. A further analysis was carried out at the same gas gain and amplifier gain in which the film was exposed at a slower speed in order to obtain a considerably larger number of pulses in the region between 13.5 kev and 18 kev. The results of these measurements are given on the right hand side of Figure 17. Photoelectrons from



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the fluorescence X-radiation of copper were superimposed on the beta spectra to provide an energy calibration, and the pulse distribution, which is also given in Figure 17, gave two quite distinct groups. The natural background of the counter, with energy calibration, had first been determined over a period of several minutes, and the pulse size distribution obtained; this distribution, which averaged 40 pulses per group, was subtracted from the experimental distribution before it was plotted in Figure 16.

It will be seen that the counting rates in the majority of the energy intervals are considerably higher than the background counting rates, except very close to the end-point where some lack of accuracy results. The spectrum obtained here increases continuously towards the low energy end, and is slightly concave throughout. Since this arises principally from the large end-effect no discussion of this figure will be given; a comparison of these results with those of later work, in which the end-effects were much smaller, demonstrates the appreciable distortions which may result from such an effect.

The upper energy limit of this spectrum was found to be 16.9 kev, and, because of the large background, the probable error was considered to be about $\pm$ 0.3 kev. Several later runs with these counters gave end-point values between 17.0 kev and 17.9 kev; the counters used were of similar length but

smaller diameter, and they consequently had smaller background counting rates.

The metal counter 40 cm. long and 6 cm. in diameter, which had been used for a study of the end-effect described in Section 5, was used for the second set of measurements. The experimental results obtained with this counter as described above are given in Figures 18 to 23; in each case the energy scale was determined from a calibration with the fluorescence X-rays of copper. Figure 18 gives the distribution of beta-particles for energies between 1.5 kev and 19 kev, and also gives the distribution of background pulses; Figure 19 the end-point distribution between 15 kev and 19 kev. The broken curve of Figure 20 was obtained by smoothing the experimental results and subtracting the background; the full curve represents the Fermi distribution for an end point of 18 kev and has been corrected for the end-effect; the two curves have been arranged to have the same total area. These corrections were made as follows; firstly, the theoretical Fermi distribution:-

$$F(W) = (W_0 - W)^2 . \text{ p.W.G.D.}$$

may be written as:-

$$S(E) = (E_0 - E)^2 \left[1 - e^{-\pi \sqrt{0.5369/E}} \right]^{-1} D,$$

where p is the momentum in units of mc

W is the total energy in units of mc^2

E is the non-relativistic energy in kev

FIG. 18

PULSE DISTRIBUTION OF BETA-PARTICLES OF TRITIUM

(SECOND EXPERIMENT)

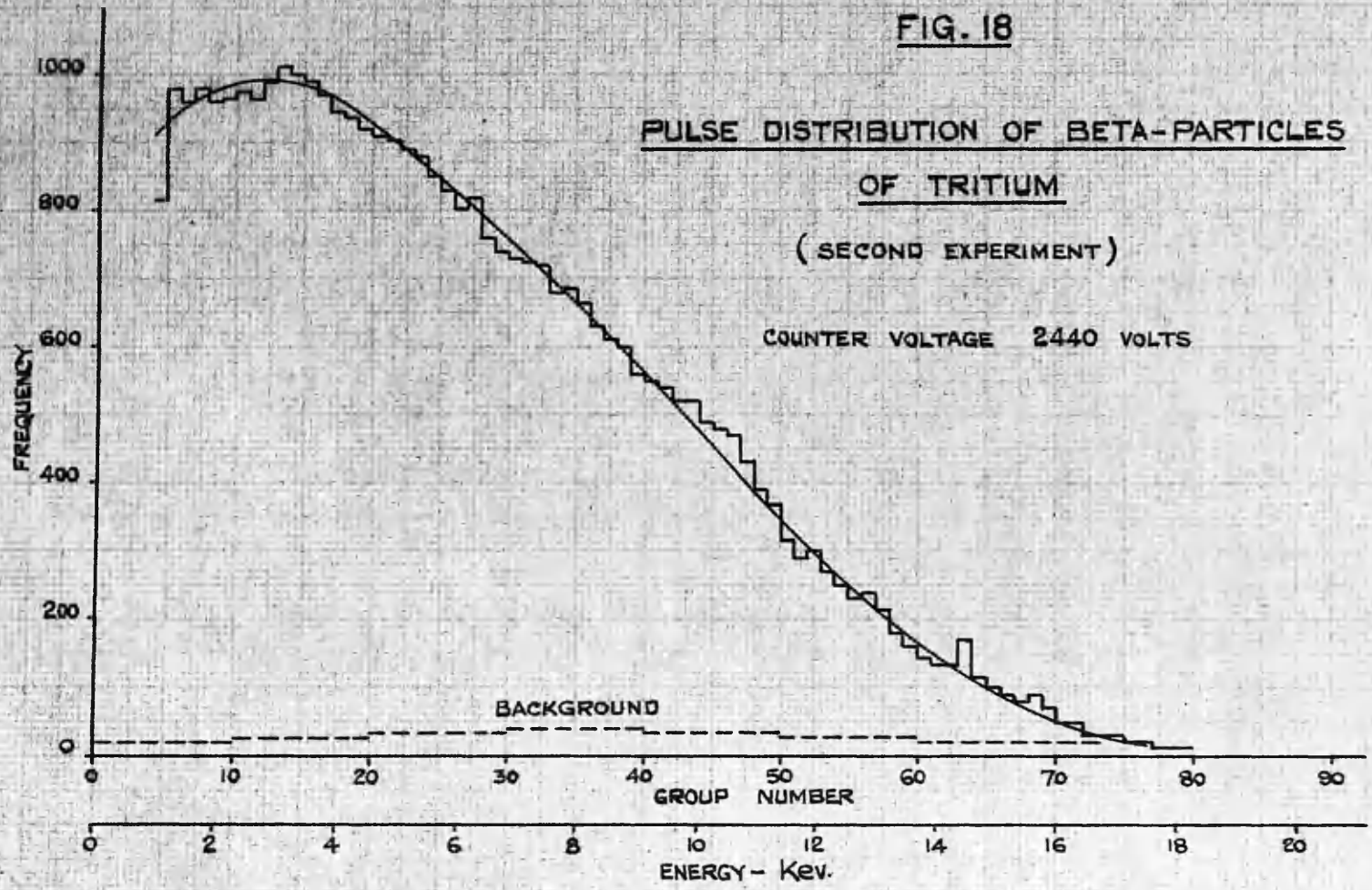

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FIG. 19

PULSE DISTRIBUTION OF BETA-PARTICLES OF TRITIUM NEAR END POINT

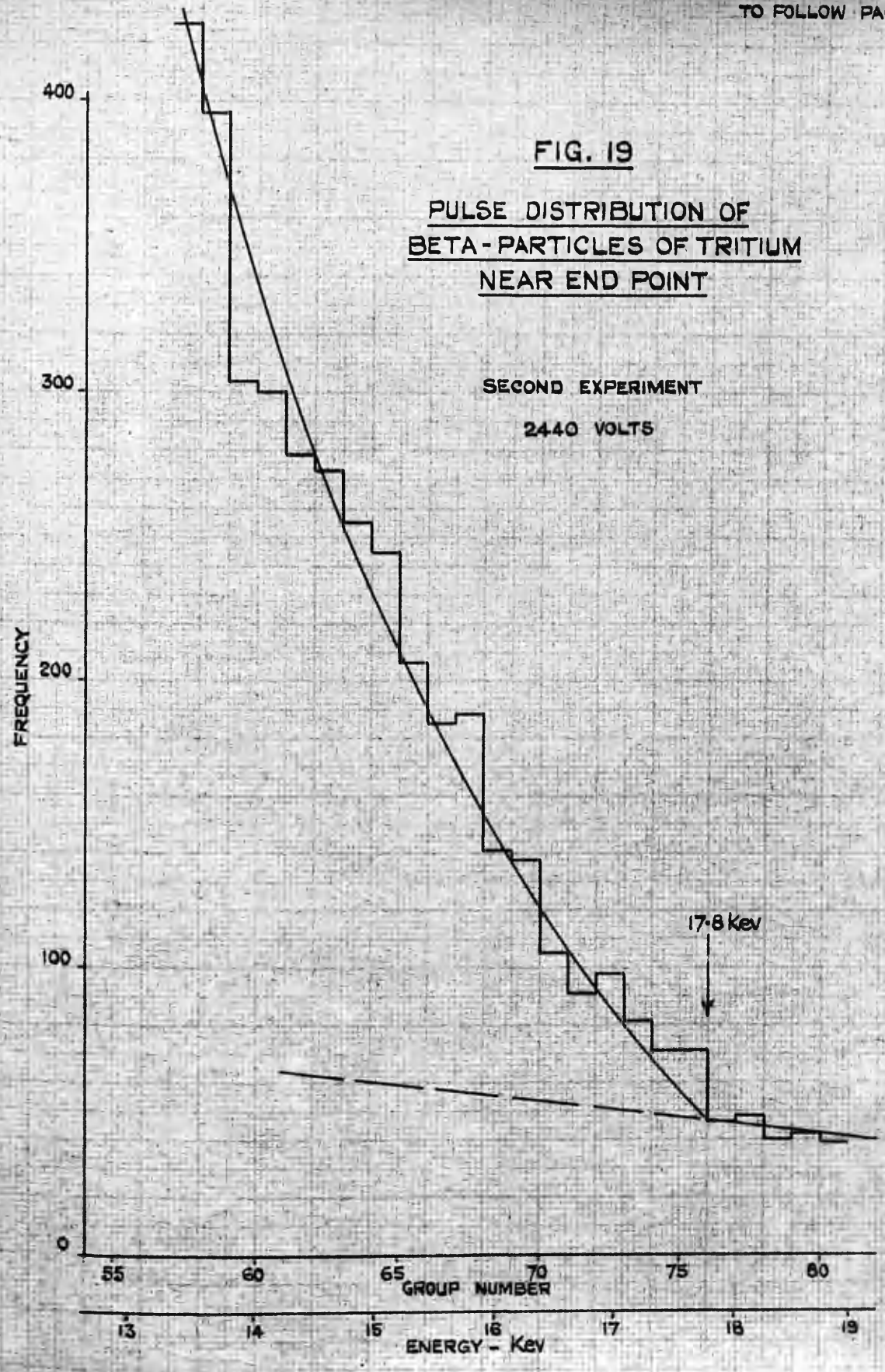
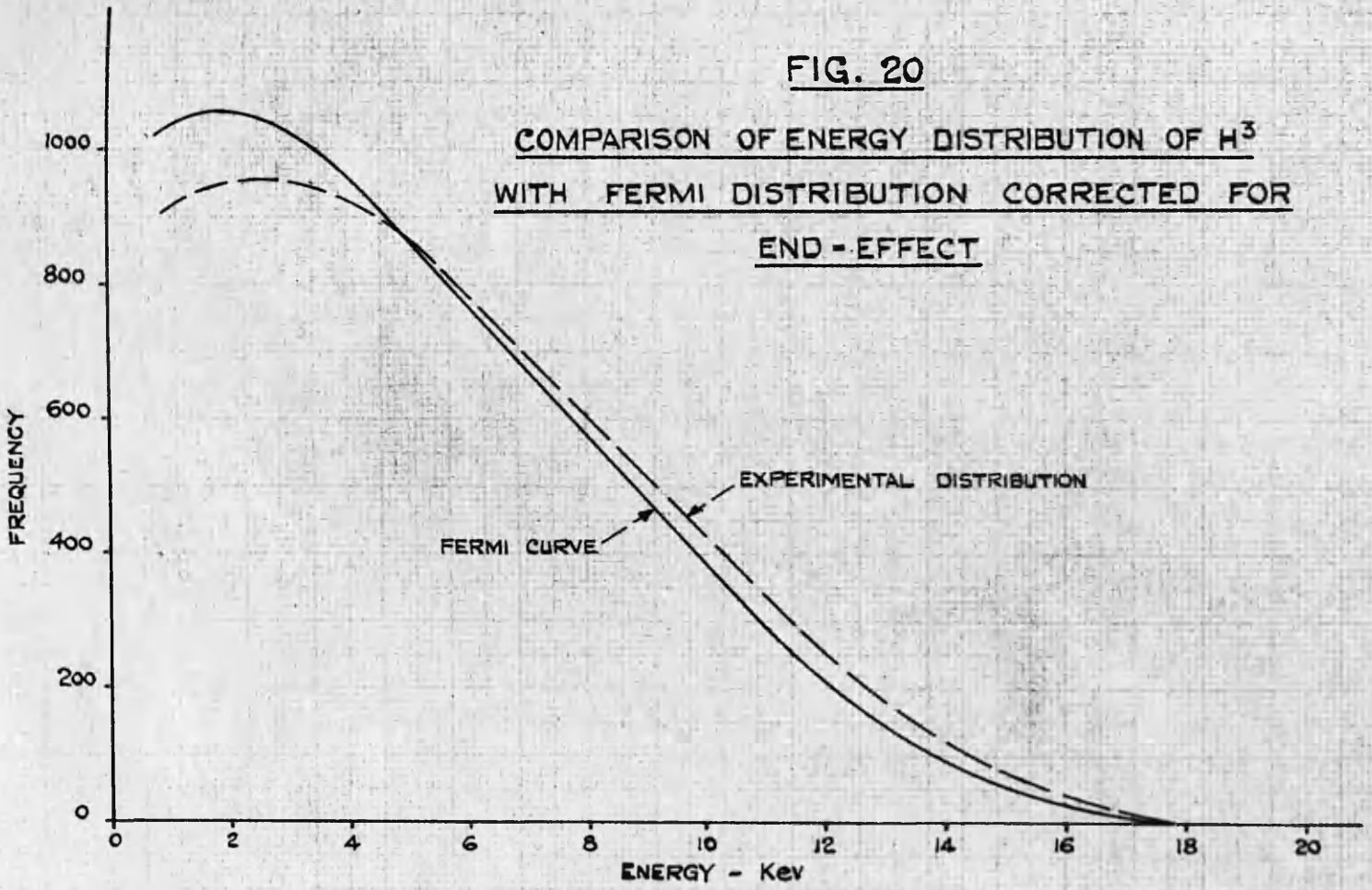


FIG. 20

COMPARISON OF ENERGY DISTRIBUTION OF H^3 WITH FERMI DISTRIBUTION CORRECTED FOR END-EFFECT



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Z is the charge of the product nucleus, and is equal to 2

G is the Coulomb factor,

and D is the Dirac factor, which is effectively constant for these low energies.

The end regions of the counter were considered to be divided into small sections between planes perpendicular to the wire and the gain of each section was known from the results given in Figure 4 as a fraction of the gain in the central section of the counter. Fermi distributions of reduced scale were calculated and added to the main distribution with relative intensities equal to the volumes of the sections chosen. The modification which resulted was very slight due to the greater length of the counter.

A comparison of the theoretical curve with the experimental results may therefore be made from Figure 20. It is seen that the trends of the curves are slightly different, the experimental maximum is less pronounced than that of the theoretical curve and slightly higher in energy. The sudden drop in the last energy interval is due to experimental limitations and it does not appear in the more detailed investigation of lower energies. Elsewhere, however, the departure from the theoretical curve is not very great, although the intensity appears to be systematically smaller than would be expected at the lower energies and greater at higher energies.

Since allowance has been made for the end-effect and since the distortion due to wall absorption effects must be very small at these higher energies, it is difficult to attribute these small discrepancies to any experimental limitations.

The end-point energy of this curve is 17.8 kev, and the estimation is considerably more accurate than that previously quoted, since the background effect is considerably less and may therefore be allowed for more accurately. A Fermi plot of the results is given in Figure 21. In this $\left[N(1 - e^{-\sqrt{2.1476/E}})\right]^{\frac{1}{2}}$ is plotted against E, where N is taken from the smooth curve of Figure 20, and it is seen that the points lie very close to a straight line with an intercept on the E-axis of 18.1 kev. This surprising agreement with the Fermi predictions must, however, be taken to indicate a somewhat insensitive property of the method of plotting. The final result is 17.9 kev and the accuracy has been estimated as $\pm$ 0.3 kev.

The results of a separate investigation of the spectrum at low energies are given in Figure 22; they cover an energy range from 600 ev to 6 kev, and were obtained by increasing the gas amplification in order to allow more accurate data to be obtained. This confirms the maximum at 3 kev and indicates an extrapolated intensity value at zero energy not less than 0.8 times the intensity at the maximum.

Although it was extremely unlikely in view of these

FIG. 21

FERMI PLOT OF PULSE DISTRIBUTION OF TRITIUM

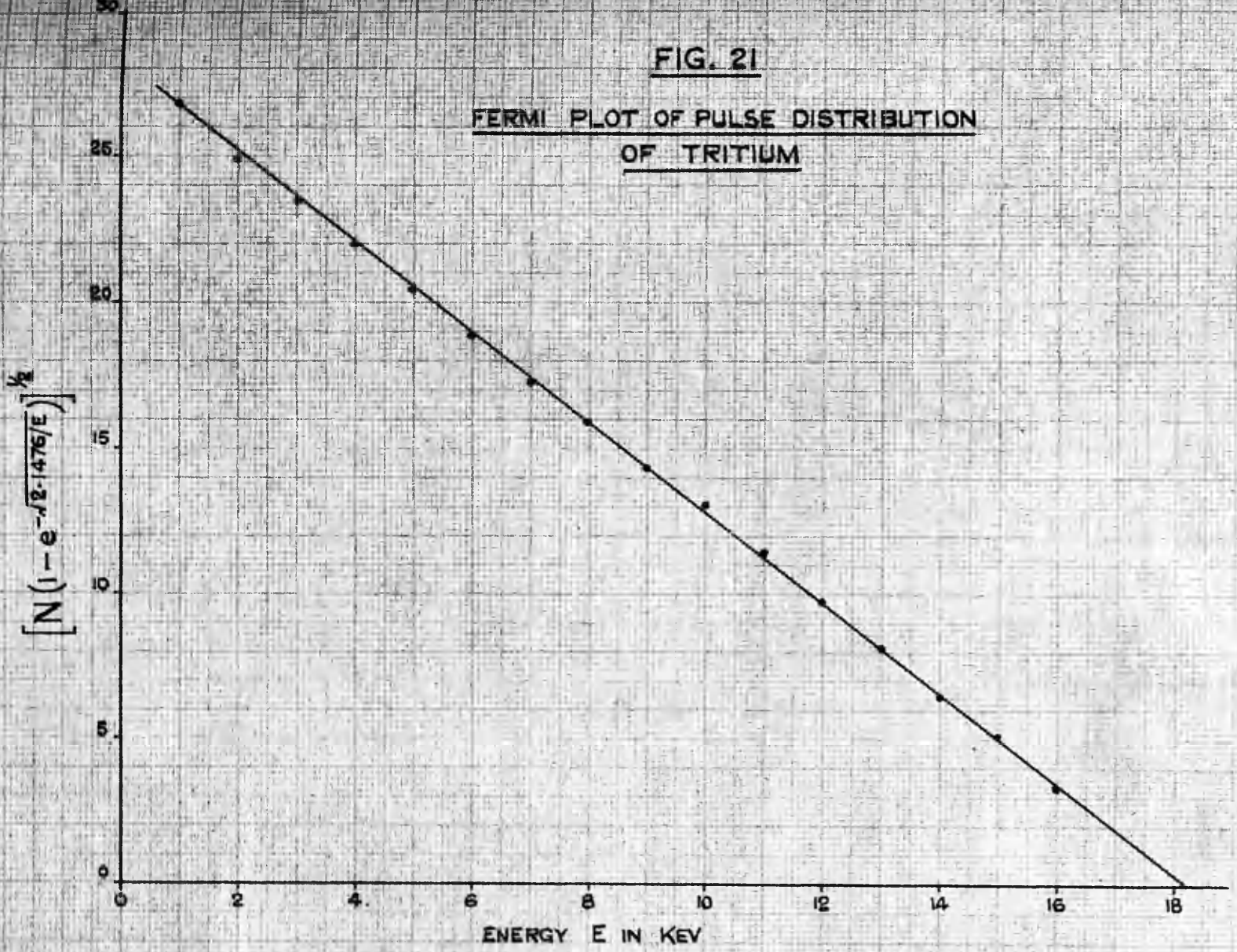

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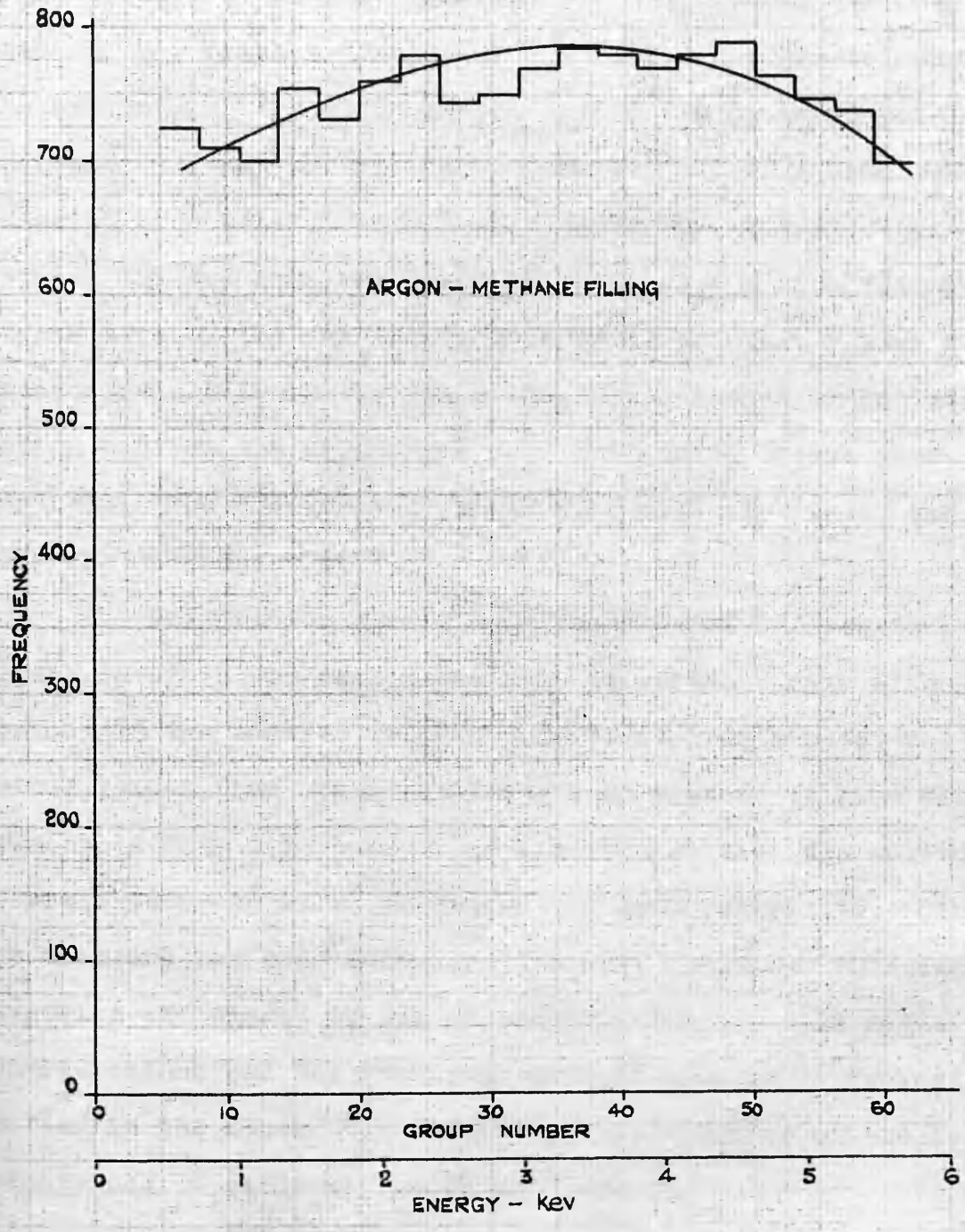
FIG. 22

PULSE DISTRIBUTION FROM TRITIUM IN LOW ENERGY REGION



results that the transition was complex, this was checked in the following manner. Tritium gas was introduced into an envelope containing two Geiger counters, as shown in Figure 23. A gas mixture of 74 cm. argon and 1.5 cm. alcohol was used so that, if any gamma radiation of low energy was emitted during the transition, the efficiency of detection would be not less than 25%. If any of the transitions results in a simultaneous emission of a beta particle and a gamma ray, coincidences between the two counters would be observed. A mica foil was placed between the two counters in order to avoid a high coincidence rate between the two counters due either to an interaction between the discharges or a triggering by the same beta-particle; the counter was placed in a magnetic field, though this was not essential.

The tritium gave a counting rate of 880 counts per minute (average) in each tube; the coincidence rate with tritium in the counter (2.25 ± 0.10 counts per minute) was actually less than the coincidence rate when no tritium was present (2.50 ± 0.16 counts per minute), so that the maximum possible increase of coincidences was 0.01 per minute. Since the counters had been calculated to have a minimum coincidence detection efficiency of one percent, including solid angle considerations for the traversal of each counter by radiation emitted in the other, the maximum possible number of complex transitions is certainly under one percent.

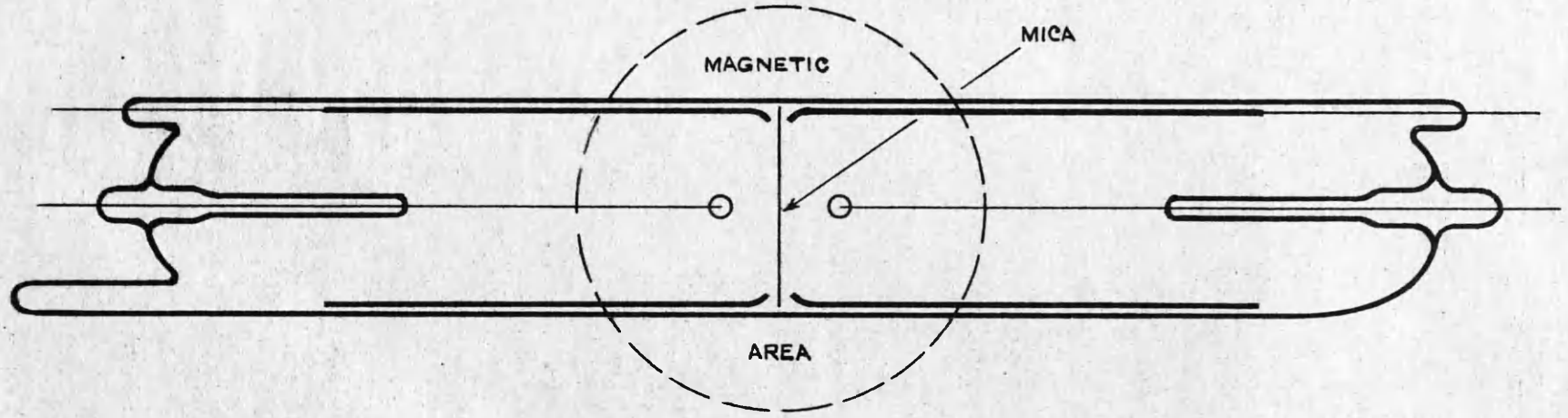


FIG. 23

DIAGRAM OF COUNTER
FOR COINCIDENCE MEASUREMENTS ON TRITIUM

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In view of the reasonable agreement of the experimental results with the predictions of the Fermi theory and the great sensitivity of the theoretical curves to the mass of the neutrino, the detailed end-point curve may be compared with Fermi curves calculated for various neutrino masses. Before considering this question, however, it is necessary to discuss the effect on the shape of the spectrum of the limited number of ion pairs produced by the beta particles. Since the number of ion pairs produced at the maximum energy is 600, the resolving power of the instrument (or the spread in the number of ion pairs) will be significantly large. In practice, the finite resolving power of the instrument will not affect the main part of the curve since the variations in the curve are small. Near the end-point, however, the effect may be relatively more pronounced. If a Fermi distribution with end-point at 18 kev is drawn, and the resolving power of the instrument is taken to be of the form shown in Appendix 2 (full width of $n^{\frac{1}{2}}$ at half height, in agreement with the results of Section 5), the experimental curves may be calculated. These curves are shown as the full and broken curves of Figure 36; it will be seen that the effect is small and that no appreciable distortion occurs except within the last one kev from the end-point. A full analysis of the correction is given in Appendix 2.

Fermi curves have been calculated for various

neutrino rest masses between 0 kev and 4 kev, and the resolving power correction has been included in them. The formula from which these curves were calculated is:-

$$F(W) = pWG\,(W_o - W + \mu)\sqrt{(W_o - W)(W_o - W + 2\mu)}$$

where μ is the neutrino mass. A comparison of these curves with the experimental points (Figure 24) referring to grouped values from Figure 19 indicates good agreement with the curves for a neutrino mass of 0 or 1 kev. The experimental errors of these points do not rule out a neutrino mass of zero and additional accuracy would be necessary. Assuming that the neutrino theory is valid the results may be stated as:-

$$\mu = m/500 \pm m/500$$

The end-point value of 17.9 kev is in much better agreement with the half-life value than the results of Watts and Williams and lessen the theoretical discrepancy without necessitating the assumption of a relatively large neutrino mass.

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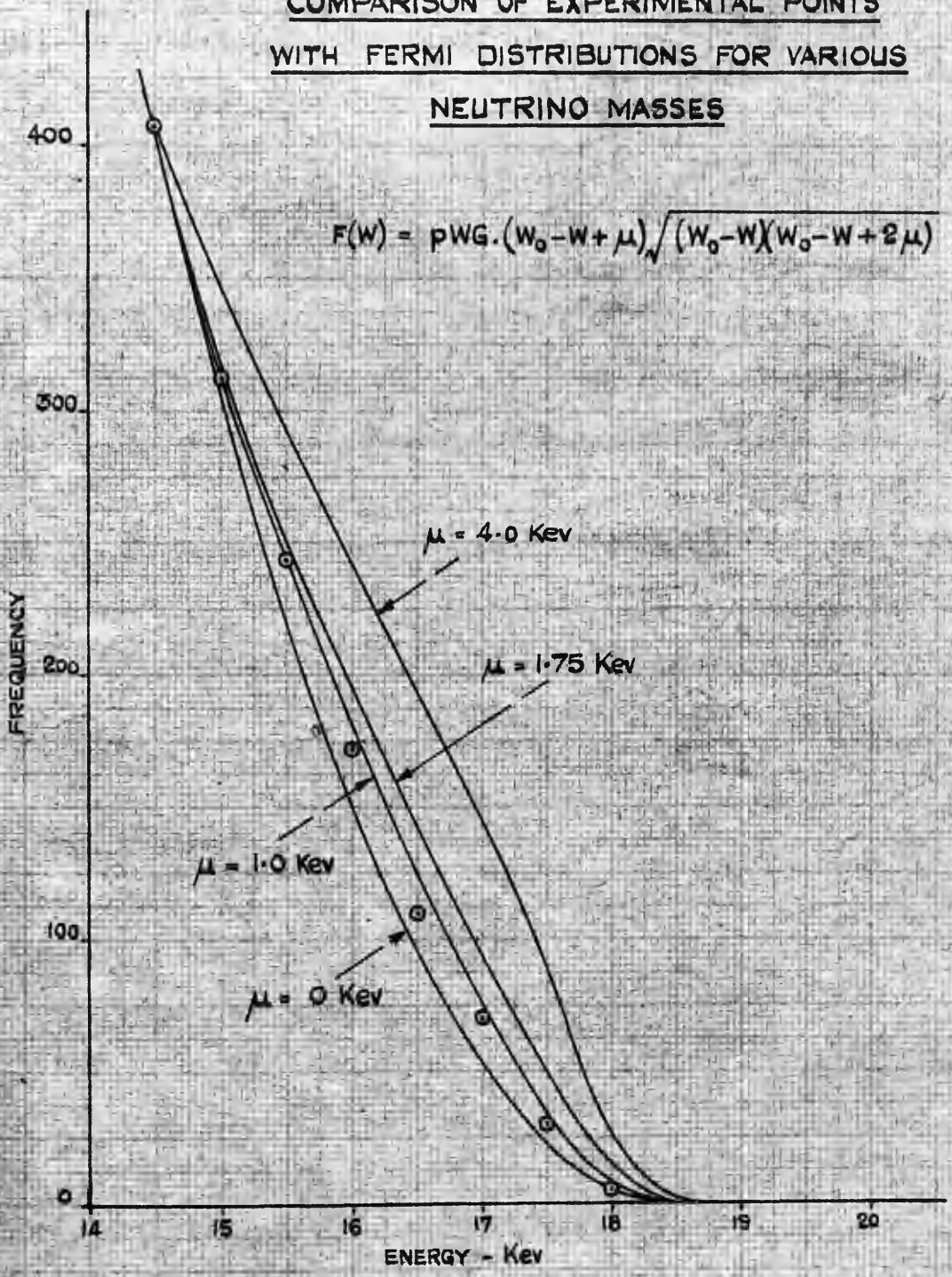
FIG. 24

COMPARISON OF EXPERIMENTAL POINTS WITH FERMI DISTRIBUTIONS FOR VARIOUS NEUTRINO MASSES



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SECTION 8

THE BETA SPECTRUM OF CARBON 14.

Introduction.

Carbon 14 was the second radioactive source to be investigated in this series. As in the case of tritium, it was possible to introduce the source into the gas mixture of the counter; the source was in the form of carbon dioxide, and the amount of this gas which was introduced into the mixture was so small that it could produce no alteration in the performance and characteristics of the gas mixture.

The upper energy limit of the spectrum of this source has been reported as about 155 kev. Since this is very much larger than the maximum energy of tritium it was necessary to use a counter of much larger dimensions, in order to ensure that the complete energy of such an electron would be detected. The energy corresponds to a range of 28 mg/cm^2 of aluminium, i.e., approximately 14 cms. of argon at atmospheric pressure; the counter was pressurised in order to reduce the track length corresponding to the maximum energy to a small fraction of the diameter of the counter.

In this investigation a first application of the beaded counter method of correction for the end-effect was made; the central wire was divided into two unequal sections by a glass bead, thus dividing the counter into two volumes.

The distributions of pulses from the sources were recorded in each section in turn, and, since both sections have identical end assemblies and should therefore have identical end-effects, the difference between the two distributions represents the distribution which would have been obtained from a counter free from end-effect.

Measurements were made of the pulse distributions at two gain levels, before and after the radioactive gas had been introduced. One of these records gave the general pulse distribution over the whole energy spectrum, while the other covered the region below the peak intensity of the distribution. A record was also made from which a more accurate determination of the upper energy limit could be made. In each case records were taken at both ends of the counter; while this increased the amount of analysis necessary, it was considered to afford the simplest and most accurate method of correcting for the distortion at the ends of the counter.

The value of the half-life of carbon has been determined as $6 - 7 \times 10^3$ years [34]; this value, together with the upper energy limit, indicates that the transition is at least first, and probably second order "forbidden"; such a transition would be expected to differ considerably in shape in the low-energy region from the spectrum of an allowed transition, as, for example, that of tritium. In a "forbidden" transition, for example, the spectrum might be expected to fall to zero

intensity at zero energy, and the results of this investigation showed that the spectrum differed considerably in shape from that of tritium.

During the course of this work a distribution covering the major part of the spectrum was reported by Cook, Langer and Price[31]. Their results, which were obtained with a high resolution magnetic spectrograph of large radius, were to a certain extent subject to uncertainties associated with source thickness and mounting; these uncertainties were more pronounced at low energies. There is a reasonable agreement between the results obtained by these authors and the distributions described here, and both indicate that certain discrepancies exist between the experimental curve and the theoretical predictions.

Previous Work.

Reuben and Kamen[34, 35] showed that carbon 14 emitted a continuous beta spectrum, unaccompanied by gamma radiation; from absorption measurements they evaluated the maximum energy of the beta-particles as around 145 kev.

Levy[36] examined this source and evaluated the maximum energy as 154 ± 3 kev; a Kurie plot of the spectrum in the range 110 to 154 kev gave a straight line relation.

Lewis and Paul[37] reported a maximum energy value of 152 ± 5 kev, also from magnetic spectrograph analysis, although no spectrum was given.

Solomon et al.[38], using aluminium absorption measurements and the Feather plot, obtained a value of 154 ± 4 kev for the end-point. This method does not give any indication of the shape of the spectrum.

Cook, Langer and Price[31] gave more detailed results of spectrometer studies of the spectrum covering an energy range from about 5 kev to the upper energy limit, the value of which they determined as 156.3 ± 1.0 kev. The spectrum rises from almost zero intensity at a few kev to a maximum at 30 kev and thereafter falls gradually to the end-point. When the results are plotted on a Fermi plot the experimental curve falls rapidly below a straight line relation for energies less than 50 - 60 kev, but above this value the curve is reasonably linear, although there is a slight alteration in the slope of the line at high energies.

The spin of carbon 14 is 0[39], and the corresponding value of the product nucleus, N^{14} is 1[40], so that the transition involves a change of quantum number of 1. In the paper of Konopinski to which reference has already been made the Fermi selection rules and those of Gamow and Teller are set out for such a transition. These show certain differences and will be referred to in the discussion of the results.

Experimental Method.

The counter used in these experiments, which is shown schematically in the inset Figure 25, was made from a

copper cylinder 14 cm. diameter and 75 cm. long, with a wall thickness of 3/8 in. (0.96 cm.). Thick copper end-plates were soldered to the tube and clamped in position by four copper tie-rods running the full length of the counter. The wire was supported by the normal insulated mountings shown in Figure 3, although the ebonite screwed sleeve was lengthened so that over one inch of thread could be engaged in the end-plate bushing. This was found to prevent the ebonite from being forced out by the pressure of the gas inside the counter.

The central wire was of tungsten, 0.004 in. in diameter; the wire consisted of two sections joined together by a glass rod 1.5 cm. in length and about 1 mm. in diameter. The two sections of the wire, which were insulated from each other by this bead, could be joined in turn to the head amplifier and the pulse distribution of either of the two counter sections measured. The lengths of the two wire sections exposed inside the counter were 47 cm. and 16 cm.

Two windows, 0.25 in. diameter and 0.03 in. thick, were provided, through which fluorescence X-rays of various metals could be introduced for calibration of the low energy distributions of each end.

The gas was prepared from a sample of barium carbonate containing a few microcuries of radioactive carbon. A small part of the carbonate was heated on a platinum filament inside a vessel containing argon at a pressure of 10 cm.

Hg; during the heating process carbon dioxide was released, the argon providing a diluent and carrier. An activity of 10^7 counts per minute was obtained, indicating that the process had been reasonably efficient. This method of preparation ensured the minimum introduction of impurities into the gas mixture, as, for example, oxygen or water vapour, either of which would have been extremely troublesome at the higher pressures used here.

A small amount of the gas mixture, with an activity of 50,000 counts per minute, was removed from the main sample and introduced into the counter after the latter had been evacuated; nitrogen was added to a pressure of 30 cm. Hg. and the counter was then pressurised to a total pressure of $5\frac{1}{2}$ atmospheres with argon. Nitrogen was used as the quenching gas, since the methane used in the lower pressure work appeared to have a small water vapour content.

The counter was operated at a voltage around 6,000 volts, and the ebonite insulators gave satisfactory behaviour, since the number of spurious pulses was very small. At this voltage satisfactory uniformity of calibration X-ray pulses was obtained.

It is interesting to note that, in a preliminary experiment in which the central wire was of 0.0015 in. diameter, homogeneous gas multiplication could not be obtained; the wire was used in order to reduce the voltage necessary to

operate the counter, but, while the voltage was almost one-half of that eventually used, the resulting small value of the field strength near the circumference of the counter appeared to be insufficient to overcome the recombination effect. The inhomogeneity of pulse size may have been due also to the fact that methane was used in these early experiments.

As in the case of tritium, the radioactive gas did not interact in any way with the walls of the counter, since all activity could be removed very easily by evacuating the vessel.

Experimental Results.

For the first run the overall gain was set to cover an energy range from 20 kev to 200 kev. At full amplifier gain this would have involved a gas gain of between two and three; the amplifier gain was therefore cut by a factor of 10, and the gas multiplication was raised to between 20 and 30. This was considered to be well below the maximum value for the quenching gas, and at the same time to be sufficiently large to avoid fluctuations due to high statistical variations of gas gain.

Calibration of this run was supplied by means of the characteristic X-rays of tungsten. The bombarding voltage of the X-ray machine was set so that these X-rays would be excited in the target, and the output beam was fired directly

through the wall of the counter. In passing through the copper wall the K_α X-rays were absorbed much more strongly than the K_β X-rays due to their lower energy, the ratio of the absorption coefficients being about 40 : 1. The pulse distribution of photoelectrons therefore resulted almost entirely from the higher energy group. In each case the X-ray spectrum showed a single peak, and the position of this peak was identified with the energy of the K_β X-ray line of tungsten, 66.7 kev. The energy scales of the distributions were set up on this basis. An example of the distribution obtained is shown in Figure 28 (a).

The single exception to this method of energy estimation was that of the distribution shown in Figure 25; in the calibration of this film a rather high intensity of X-rays was used, and the pulse distribution was unsatisfactory due to a partial biassing-off in the first valve of the amplifier. In this case it was necessary to use the end-point position determined from the distribution, and to correlate it with the energy value obtained in the more detailed results of Figure 28, taken under identical conditions before the gas mixture was released.

The background counting rates at the long and short ends were respectively 3,000 and 1,500 per minute; corresponding counting rates after the carbon had been introduced were 20,000 and 13,000 per minute. In each of these analyses

pulses were recorded at each end of the counter for $2\frac{1}{2}$ minutes; background pulses distributions were recorded for periods between $3\frac{1}{2}$ and 5 minutes.

The histogram for the pulse distribution obtained in the longer section is shown in Figure 25, and that obtained in the shorter section in Figure 26. The smoothed curves were obtained by drawing a mean curve through averages of pairs of groups. These curves were corrected to the same energy scale by reducing the abscissa of the lower curve by a factor of 71.5 / 63.5, and increasing its ordinate by the same factor. The difference between the two curves, which gives the pulse distribution free of end-effect, is plotted in Figure 27 (b). The background curve was also obtained in the same manner, and is plotted in this figure. The intensities of the individual sections are so small that plots for these are not given. The close agreement between the intensity values beyond the end-point energy for these two curves ($\sim \frac{1}{2}\%$ over ten groups) indicates that no radiations of greater energy are present in this source. This confirms earlier work.

The end-point distribution of the spectrum was obtained from a more detailed investigation designed to cover the region from 120 kev to 200 kev. In this experiment the film was exposed at a much slower speed in order to allow a greater number of pulses to be obtained. The measurements, which were taken at the longer end of the counter only, are

FIG. 25

PULSE DISTRIBUTION OF CARBON 14
LONG SECTION

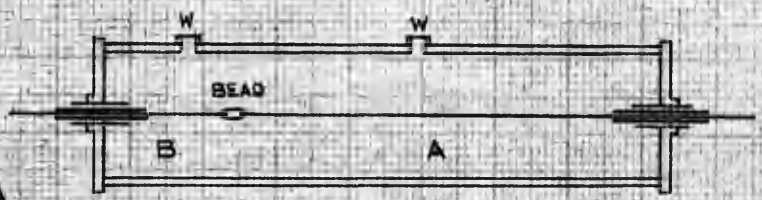
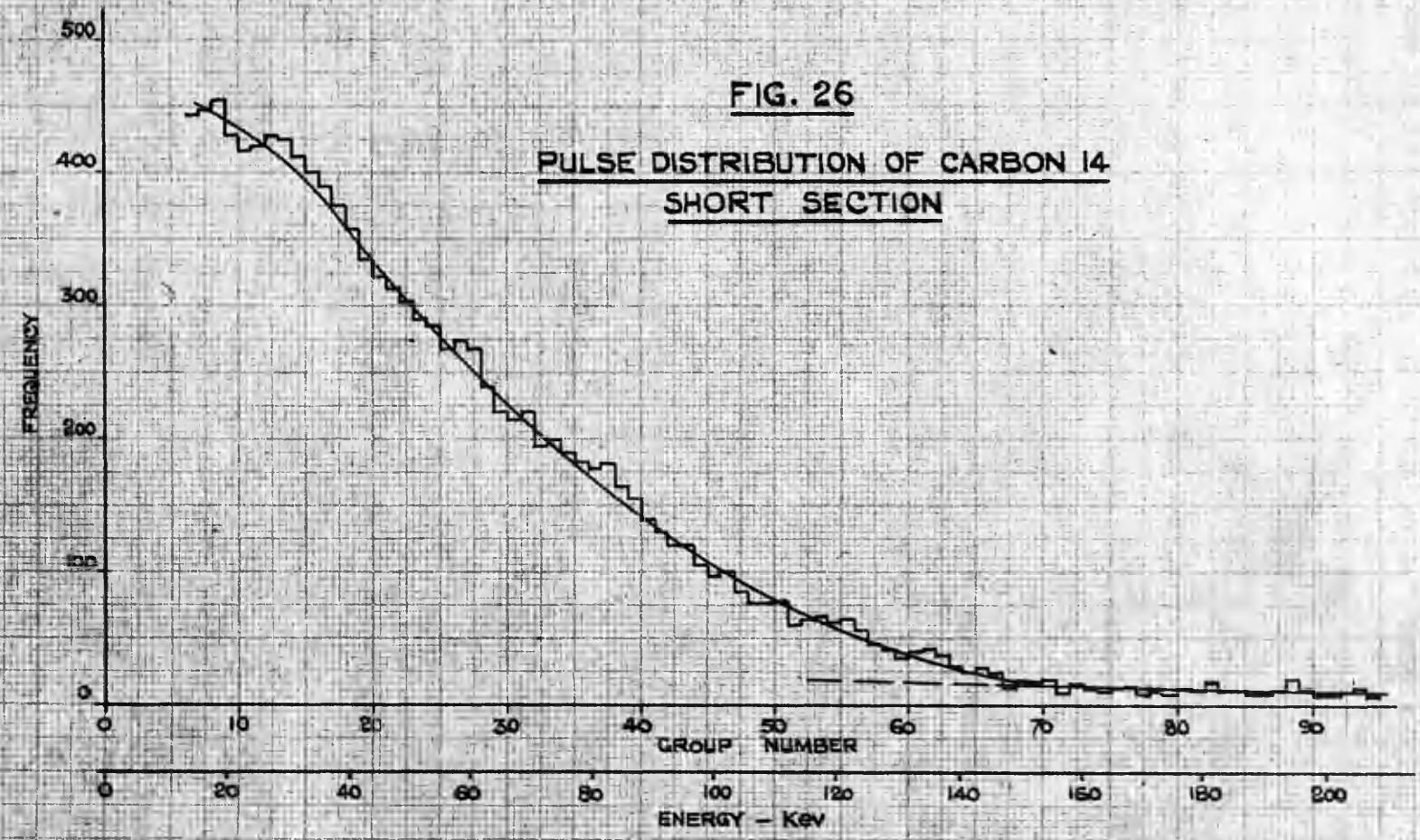


FIG. 26

PULSE DISTRIBUTION OF CARBON 14
SHORT SECTION



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shown in Figure 28 (b) together with the pulse distribution for the fluorescence X-rays of tungsten already discussed. The value of the end-point (group number 65.5) together with the statistical mean of the X-ray distribution (group number 27.5) gives an energy of 157.5 kev. The error of each group number reading has been estimated as about one division and this gives an error in the energy determination of 5 kev.

The final curve for the spectrum is plotted in Figure 27 (a) together with the Fermi distribution calculated for an upper energy limit of 157 kev and a charge of 7 for the product nucleus. This curve was obtained from the relation:-

$$N(W) = p\,W(W_0 - W)^2 .\ G.D.$$

where p is the momentum in units of mc

W is the total energy in units of mc^2

G is the Coulomb factor $\xi\, e^{\xi} / (e^{\xi} - 1)$

$$\xi = 2\pi Z/137.\ (W/p)$$

and D is the Dirac factor. For these energies the last factor is constant to $\frac{1}{2}\%$ over the whole spectrum and has been neglected.

The intensity values of the theoretical curve have been scaled so that the two distributions have almost equal areas. A comparison of the two curves shows that, whereas the theoretical curve has a pronounced maximum at about 24 kev, and the reduction in intensity below the maximum is only 18%,

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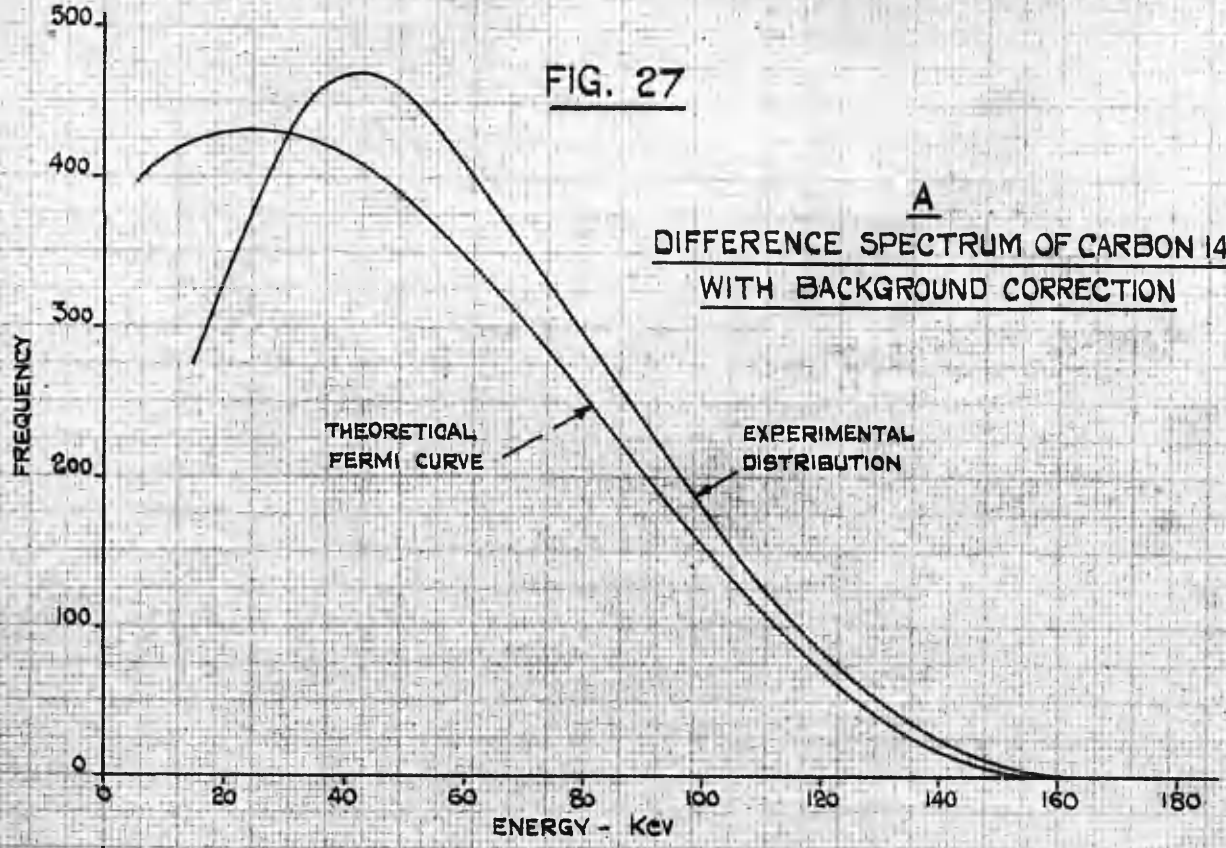
FIG. 27



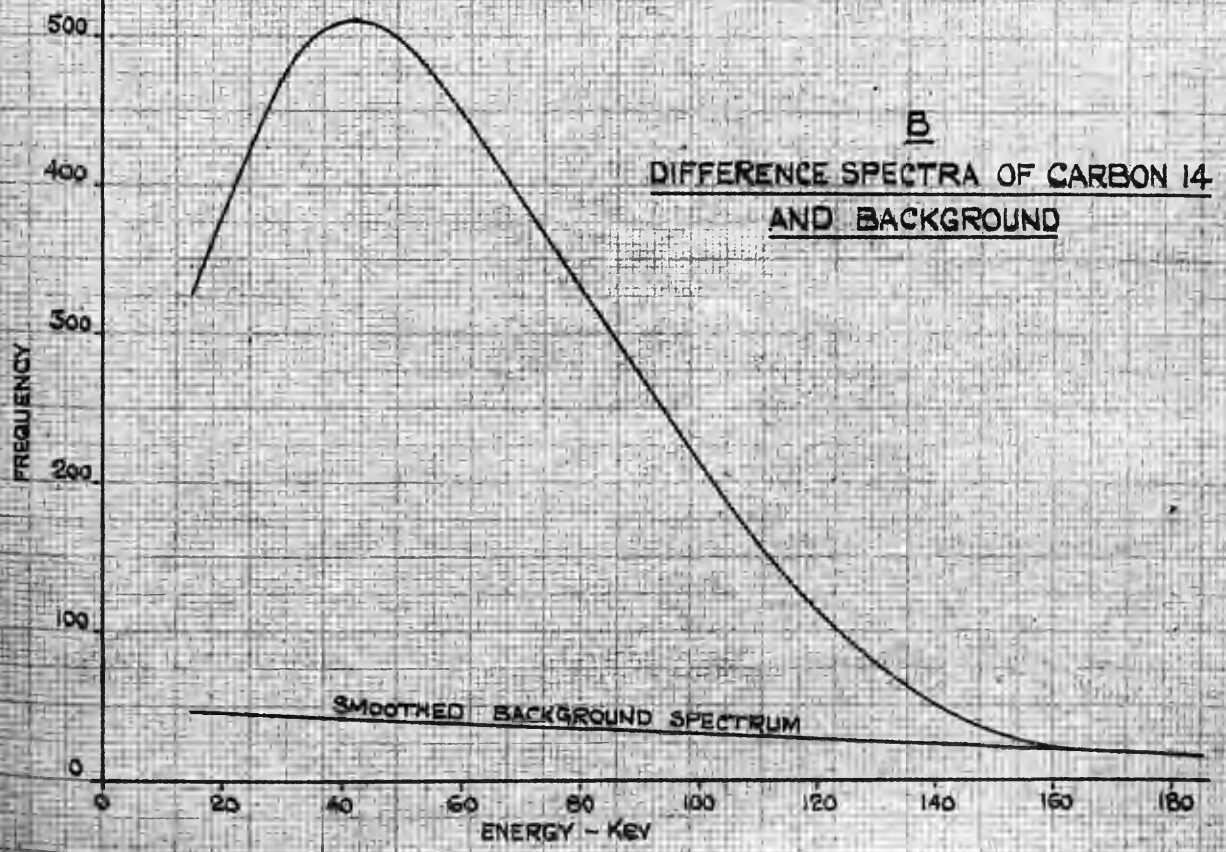
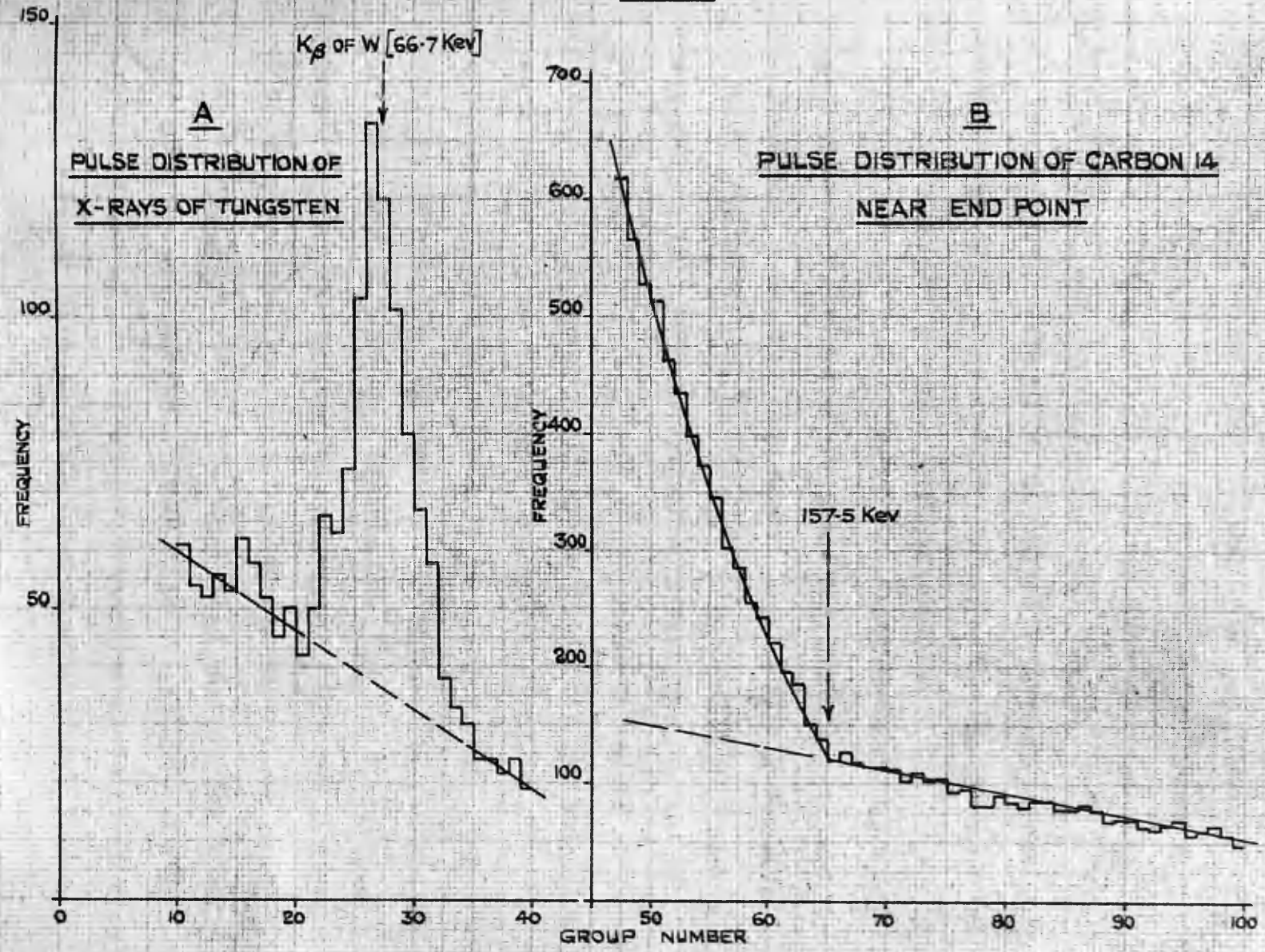


FIG 28



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the experimental curve shows a relatively sharp maximum at 40 kev and falls towards zero intensity at zero energy. The rate of fall of intensity for energies below the maximum has been somewhat exaggerated by the limitations in the method of analysis, since these small pulses were slightly more difficult to observe; the results of the more detailed investigation of the low energy spectrum confirm the position of the maximum and the fall towards zero intensity.

A second investigation, designed to cover an energy region between 5 kev and 40 kev, was carried out with the same counter and gas filling, and at approximately the same gas gain. The energy calibration for this experiment was obtained by introducing fluorescence X-rays through the windows and superimposing their photoelectrons on the particles from the source. The pulse distributions obtained at each end are given in Figure 29, together with the smoothed background difference between the two ends. The difference between the two source distributions is plotted in Figure 30 (curve A) with the smoothed background difference subtracted. The statistical fluctuations in the final curve must be considerable, due to the proximity of the two distributions below 10 kev, but, even below this energy, the trend of the curve is well marked. Since this is probably the most important region of the curve from a theoretical point of view the results of the investigation of Cook, Langer and Price

have been included in this figure; these results, shown in curve B, which were obtained from the figure in their paper and re-plotted on an energy scale, may be compared with the distribution obtained here. Apart from the difference in the position of the maximum, agreement is reasonably good, small discrepancies between the curves being explained by window effects in the magnetic spectrometer. The agreement between the results obtained by these two entirely different techniques leaves little doubt as to the behaviour of the intensity distribution at low energies.

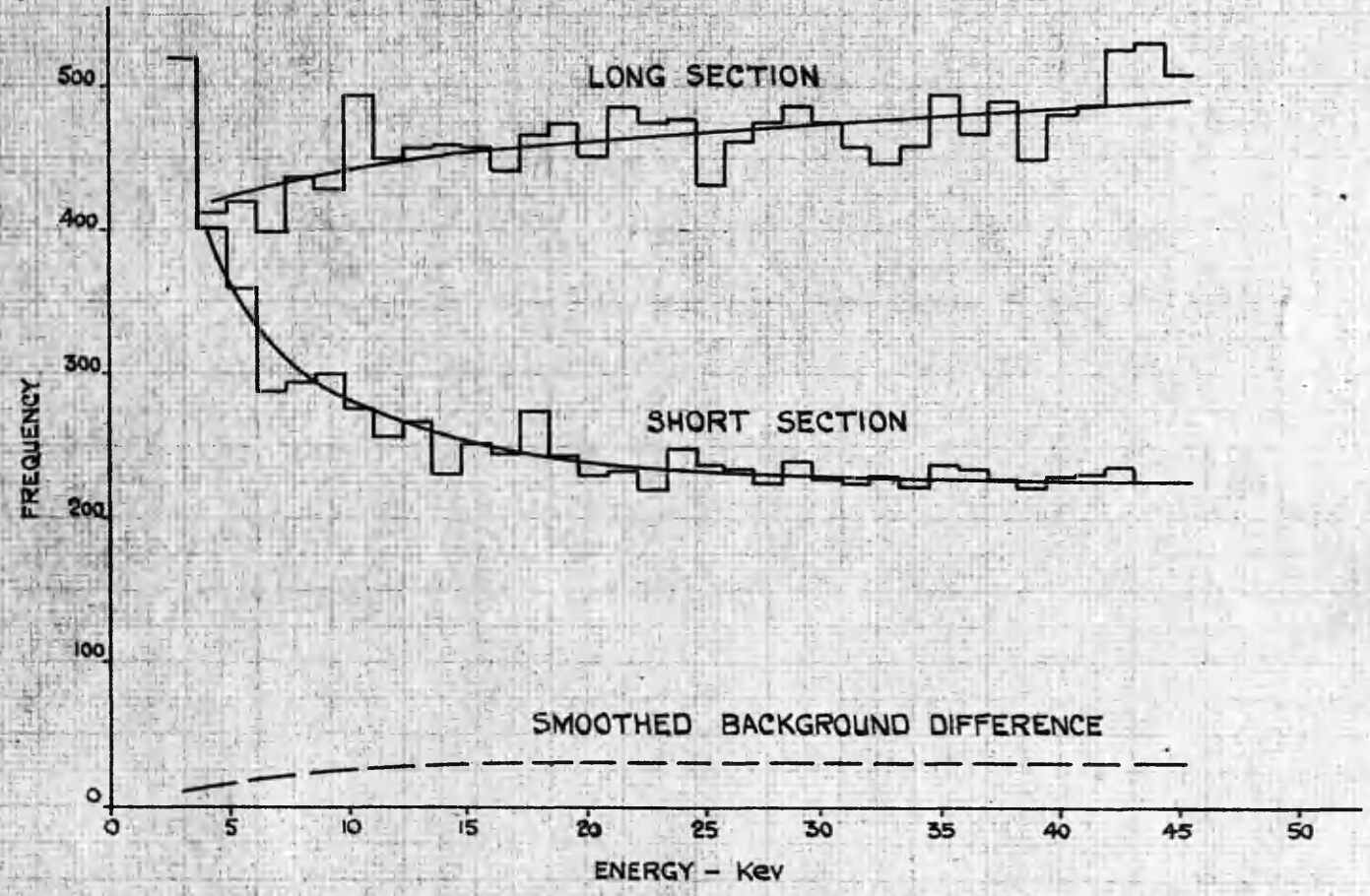
A Fermi plot of the spectrum is given in Figure 31; this plot, $\left(\frac{N}{p.W.G.D}\right)^{\frac{1}{2}}$ against E, shows that the experimental curve departs considerably from the linear relation expected at energies less than 50 kev. Above this energy the results are linear, in agreement with the finding of Cook et al.; near the end-point large fluctuations due to the small number of pulses make this plot unsatisfactory, and it is not therefore possible to confirm or disprove the change in slope observed by them. The straight line passes exactly through the energy limit 157 kev.

The considerable discrepancy which exists between the experimental and theoretical curves is of especial interest in view of the fair agreement obtained with tritium. It is not possible to draw any detailed conclusions regarding the transition due to the unsatisfactory position of the theory of

FIG. 29

PULSE DISTRIBUTIONS FOR EACH COUNTER SECTION
AT HIGH GAIN

CARBON 14



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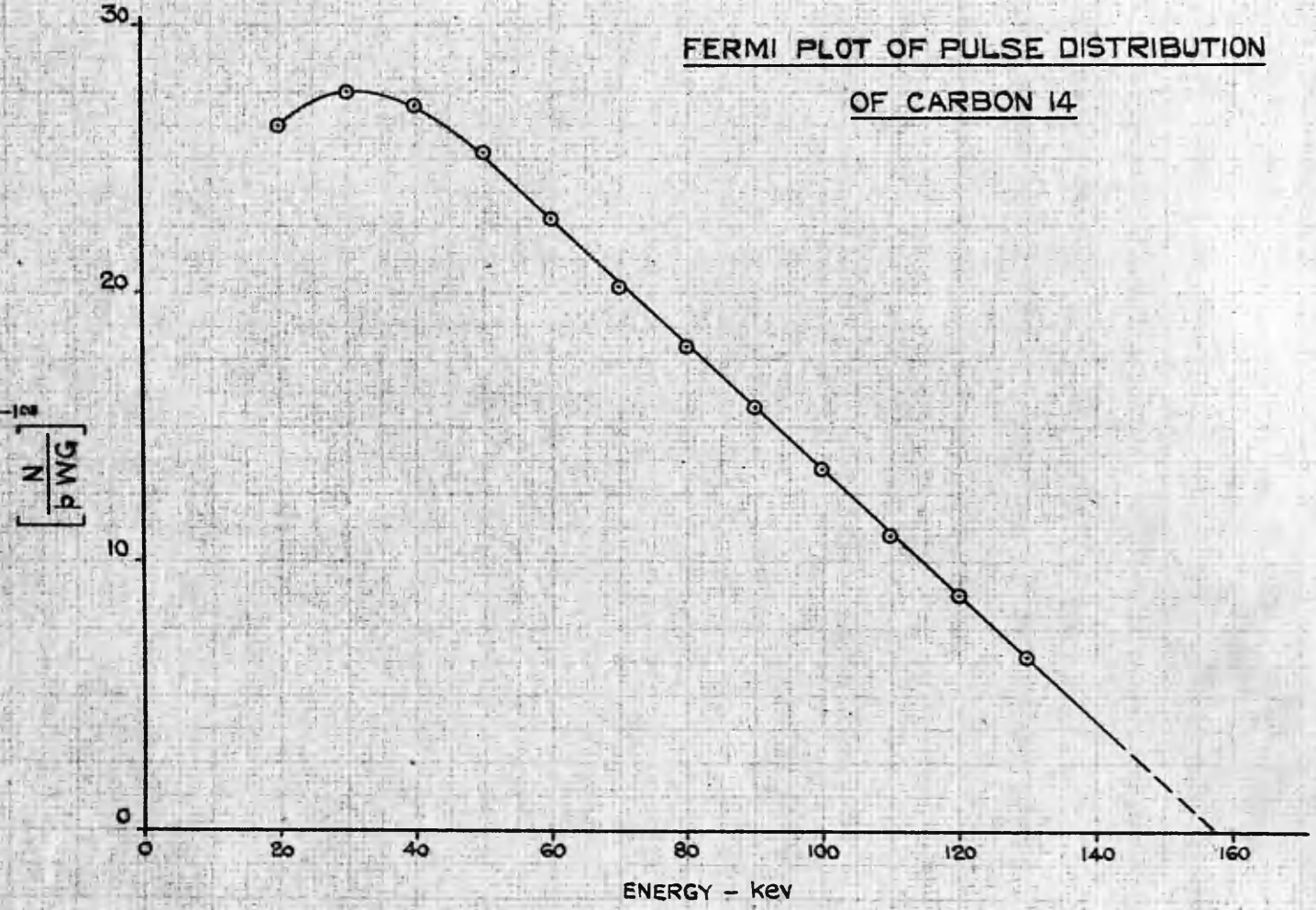
beta-decay, since a variety of energy distributions may be obtained for a transition which is forbidden, depending on the values of spin and parity; the considerable difference in shape between the spectra of tritium and carbon might, however, be related to the difference in the ft value. From the value of this quantity (7×10^8) Konopinski has shown that the transition should be classified as "second order" forbidden[30]; since carbon 14 is a fairly simple nucleus and since the energies of the particles are well below the relativistic region, the results of this investigation should afford an opportunity of testing future theoretical explanations of forbidden transitions in much the same manner as the results obtained with tritium provide considerable information regarding the simplest of the "allowed" transitions.

No conclusions have been drawn regarding the mass of the neutrino due to the much smaller sensitivity of the spectrum to neutrino mass; the results are not in disagreement with the conclusions of Section 7.

FIG. 31



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SECTION 9

THE GAMMA RADIATION OF RADIUM D.

Introduction.

Radium D was the first source for which a comprehensive study of the gamma radiation and X-rays was made with this technique. This source was chosen principally for two reasons:-

(1) A considerable number of studies of the radiations emitted in the disintegration of this element have been reported, and these results have indicated the following characteristics of the process. The disintegration appears to occur by means of the emission of a beta particle of low energy accompanied by the emission of a variety of gamma rays, also of low energy. These are accompanied by the characteristic L X-radiations of the product atom (Z = 83) with a considerable intensity, and there is evidence also for a much weaker intensity of K_α and K_β radiation from the same element. Results appear to indicate the presence of at least five gamma rays of various intensities with energies between 0 and 50 kev, and it must be concluded from the X-ray evidence that these are strongly internally converted in the extranuclear shells of the atom.

The results of previous investigations appear to indicate the existence of considerable discrepancies between

the numbers of secondary electrons and the resulting X-rays, and at the same time lead to abnormally high internal conversion coefficients. It was considered that a redetermination of the energy and intensity values of these gamma rays and X-rays could be carried out with an appreciable increase in accuracy over many of the previous determinations, and that such a study would provide more detailed evidence for the understanding of the decay process of this element.

(2) It was proposed that the technique should be applied to a study of the energy spectrum of the primary beta-particles emitted during this disintegration. The introduction of the source into the counter necessitated by this experiment would result in the detection of the electromagnetic radiations and any electrons produced by internal conversion. It was therefore necessary to obtain a clear picture of the gamma ray processes in order to separate the beta-spectrum from the electrons resulting therefrom. The results of this study have confirmed earlier work in regard to this question, while also indicating the energies of the electrons, which will be of both secondary and tertiary nature.

The results of this investigation have provided information regarding the three principal gamma rays of radium D. The energy and intensity values which have been obtained for the two radiations of lower energy are considered to be more accurate than those previously obtained. It is, however,

impossible with the present information to assign a definite decay scheme to the disintegration since the abnormal features of the process already noted remain unresolved. It would appear that these difficulties may be overcome only when correspondingly definite information is obtained regarding the distributions of both primary and secondary electrons.

Previous Work.

Ellis[41], Meitner[42], Danysz[43] and Black[44] severally investigated the beta line-spectrum of radium D by the spectrographic method and obtained a series of energy values which could be fitted to the atomic levels of elements 82 or 83. Curtiss[45] by the discovery of a further line with higher energy showed that the atomic charge must be taken to be 83, and that in this case the energy of the gamma ray could be calculated to be 46.7 kev. For some time following this result the 46.7 kev gamma ray was considered to be the only primary electromagnetic radiation emitted.

Stahel[46] studied the radiations by means of absorption measurements, using an ionization chamber as detector. He found that two components were present; the harder of the two agreed with the energy value of Curtiss, while the softer had an energy of about 11.5 kev, in agreement with the expected value for the L X-rays of radium E (bismuth). By comparing the absolute values of the currents in the

ionization chamber with the counting rate of the alpha-particles of polonium in equilibrium with the source, it was estimated that 3.8 gamma rays were emitted per 100 disintegrations, and values of 17.1, 6.5 and 4.8 were calculated for the internal conversion coefficients of this gamma ray in the L, M and N shells respectively. These were deduced from an observed value of 25.1 L X-rays per 100 disintegrations, the calculation involving a large correction for the Auger effect for the X-rays of bismuth.

It was pointed out by the same author that these conversion coefficients were considerably greater than would be expected theoretically for dipole or quadripole radiation, but no explanation could be given.

The results of more recent investigations have shown that several other gamma rays of smaller energy are emitted during the disintegration. Frilley[47], using a crystal spectrometer, observed gamma rays with energies 46.7 kev, 43 kev, 37 kev and 32 kev, to which he assigned relative intensity ratios of 100 : 5 : 5 : 10. San-Tsiang[48] observed the same gamma rays by absorption methods; by critical absorption in Pr and Nd he obtained a value of 43 kev for the second gamma ray observed by Frilley, and, by absorption in Al, obtained the following values for the intensities, including both groups of X-rays:-

GROUP	1	2	3	4
Energy - kev	14	31.5	44.5	88
Intensity (per 100 disns.)	27	0.7	3.0	1.2
Designation	L X-rays	$\gamma_{32}+\gamma_{37}$	$\gamma_{37}+\gamma_{43}+\gamma_{47}$	K X-rays

San-Tsiang and Marty[49] give the results of measurements of the track lengths of the photoelectrons produced by these gamma rays in a cloud chamber; the range distribution which they obtained agreed with the conclusions of the earlier work; out of 515 tracks measured the K-radiations gave 5 tracks and a further 100 tracks indicated a group with energy less than 25 kev. In a later publication by the same authors[50] the investigation is extended to lower energies, the cloud chamber being operated at reduced pressures. From an analysis of some 260 tracks, in which the L_α, L_β and L_γ X-rays are observed, a further group of photoelectrons with range 9.9 mm. is observed. The value of the gamma ray energy deduced from this range is 23.2 ± 0.6 kev; it is considered, however, that the uncertainty in this value is appreciably higher. A further analysis of 100 tracks revealed a group with range 1.3 mm, for which an energy of 7.3 ± 0.7 kev has been evaluated. The intensities of these two gamma rays have been estimated from these distributions to be 1 and 10 per 100 disintegrations respectively: an examination of the curves

indicates that these intensities may have been over-estimated, and the small number of events studied introduces a high uncertainty in the estimation. Some tracks of very short range have been assigned to M X-rays.

These results have been summarised in a paper by Frilley, Surugue and San-Tsiang[51]. It is pointed out that, while the number of conversion electrons from the 47 kev gamma ray is in agreement with the assumption that the gamma ray is quadripole (8.2 per 100 disintegrations) the conversion electrons of the other gamma rays are extremely weak in comparison with the expected values. Further, the number of L X-rays observed (25 - 30 per 100 disintegrations) is very much greater than would be expected from the number of conversion electrons observed. While it may be possible that a great proportion of the secondary electrons have not been detected, due to their small energies, and that these gamma rays are in fact strongly converted, the possibility remains that the primary beta-particles interact directly with the extra-nuclear electrons in such a manner as to excite the L X-rays without the agency of electromagnetic radiation. Such an interaction is more strongly demanded if account is taken of the high Auger effect. Since this is of the order of 60%, the appearance of 25 L X-rays per 100 disintegrations must be taken to imply the removal of an electron from the L-shell in $25/(1 - 0.6)$, i.e. 60 disintegrations per 100.

While it is not proposed here to discuss the results of previous investigations of the beta-spectrum of radium D, it may be pointed out that magnetic spectrometer studies failed to reveal any primary beta-spectrum. A similar result was obtained by Kikuchi[52], indicating that the maximum energy of the spectrum, if it existed, must be very small. Richardson and Leigh-Smith[53] observed pairs of tracks in a cloud chamber containing radium D in gaseous form (as lead tetra-methyl), and assigned these to the emission of a conversion electron and Auger electron. A few triple tracks were observed, assigned to the secondary and Auger electrons accompanied by a beta-particle; an analysis of the primary particles indicated that the number was small and that the maximum energy was less than 15 kev. This remains the only evidence for the presence of a primary beta-spectrum in radium D.

In view of the results described above, and particularly of the very small intensities of the radiations when expressed in terms of 100 disintegrations, it is extremely difficult to account for the majority of the transitions by which radium D decays. The only radiations which exhibit reasonably large intensities are the L X-rays and the 7-kev gamma ray; with these alone it is impossible to frame a decay scheme, since the maximum energy of the beta-spectrum is some 30 kev less than the energy of the 47-kev gamma ray, and there

is no indication of a direct beta-transition to the ground state of the product nucleus.

Sources.

Several sources of radium D were used in this work. The first consisted of about 50 capillary tubes about one inch in length containing radium D, E and F produced in the decay of the original content of the tubes, radon gas. These tubes were glued into a shallow cavity in a metal-backed polythene holder, and were covered with a sufficient thickness of polythene (0.3 cm., 500 mg/cm^2) to absorb the high-energy beta-particles emitted in the decay of radium E. The absorber also removed the alpha-particles of radium F, while transmitting a measurable number of gamma rays and X-rays from the radium D. The strength of this source was of the order of several microcuries, and, from the method of preparation, contained only the activities given above.

The use of such a source introduced large correction factors in the calculation of the relative intensity values of the various radiations; for this reason two other sources were prepared for use without this polythene covering. Both of these were prepared from a solution of radium D, E and F in nitric acid; for one of the sources a few drops of the solution were evaporated on a small brass disc 1 cm. in diameter to give a source with a strength of the order of several microcuries. This source was used in a number of the later

experiments with thin window counters, beta-particles being removed by collimating the radiations and applying a magnetic field of 5,000 gauss to prevent their entry into the counter. The other source was prepared by removing radium D chemically from the radium E and F prior to its evaporation on the disc; the principle of the separation, and the manner in which it was carried out, is given in Appendix 3. It was found that the source was almost completely free from radium E and F, but, since the intensity was rather weak, it was used only to check the identification of several of the gamma ray lines observed.

Identification of the X-Radiation.

In the initial experiments, which were carried out with a proportional counter of the type shown in Figure 1, the polythene source was used. When this source was placed near the window of the counter a very intense group of pulses was observed; these pulses were of fairly uniform height and had energies close to those of the L X-rays of lead or bismuth. A record was made of these radiations, together with the fluorescence (L) X-rays reflected from lead obtained under the same conditions. An analysis of the distributions is given in Figure 32. It will be seen that the pulse distribution for the fluorescence X-rays gives two groups of approximately equal intensities at 30.5 and 36.0 units. When these are related to the energies of the L_α and L_β lines of lead, 10.5 and 12.55 kev, an energy scale may be set up. The pulse distribution of

FIG. 32

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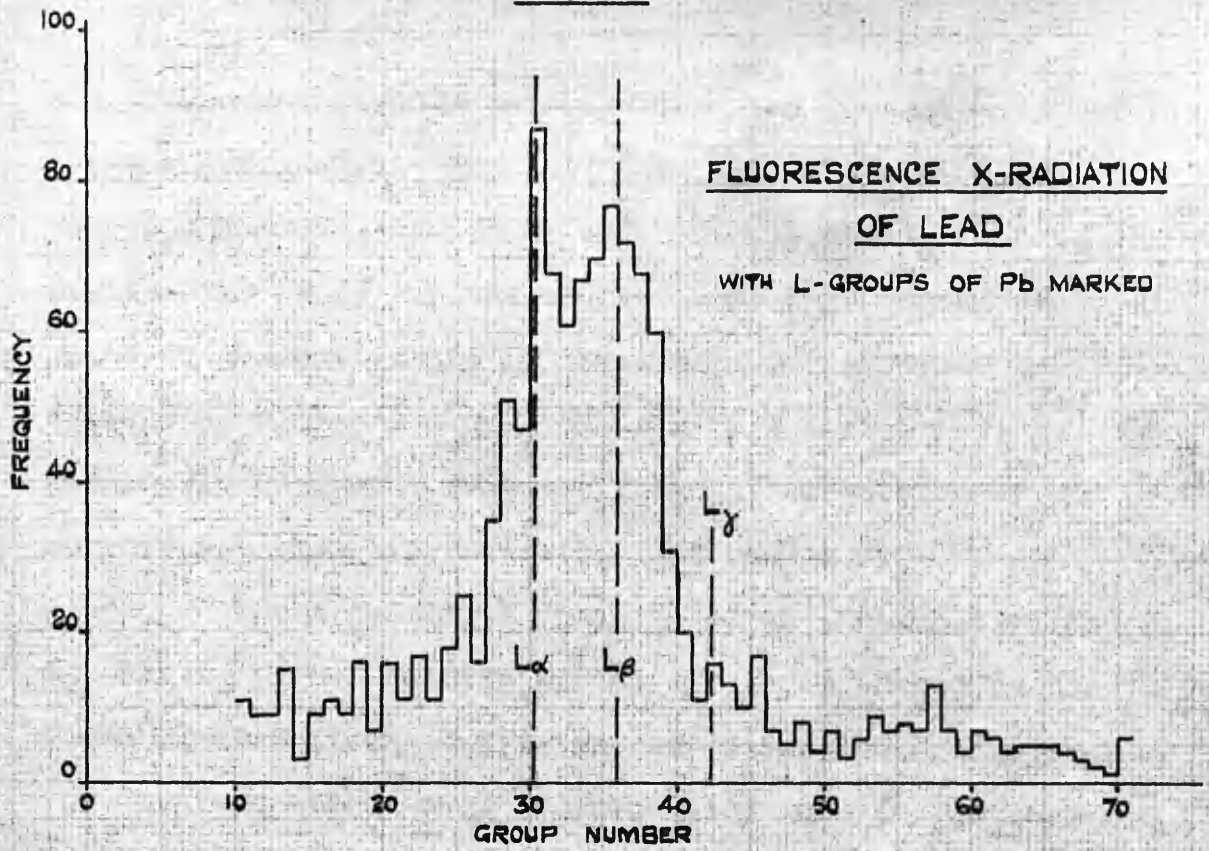
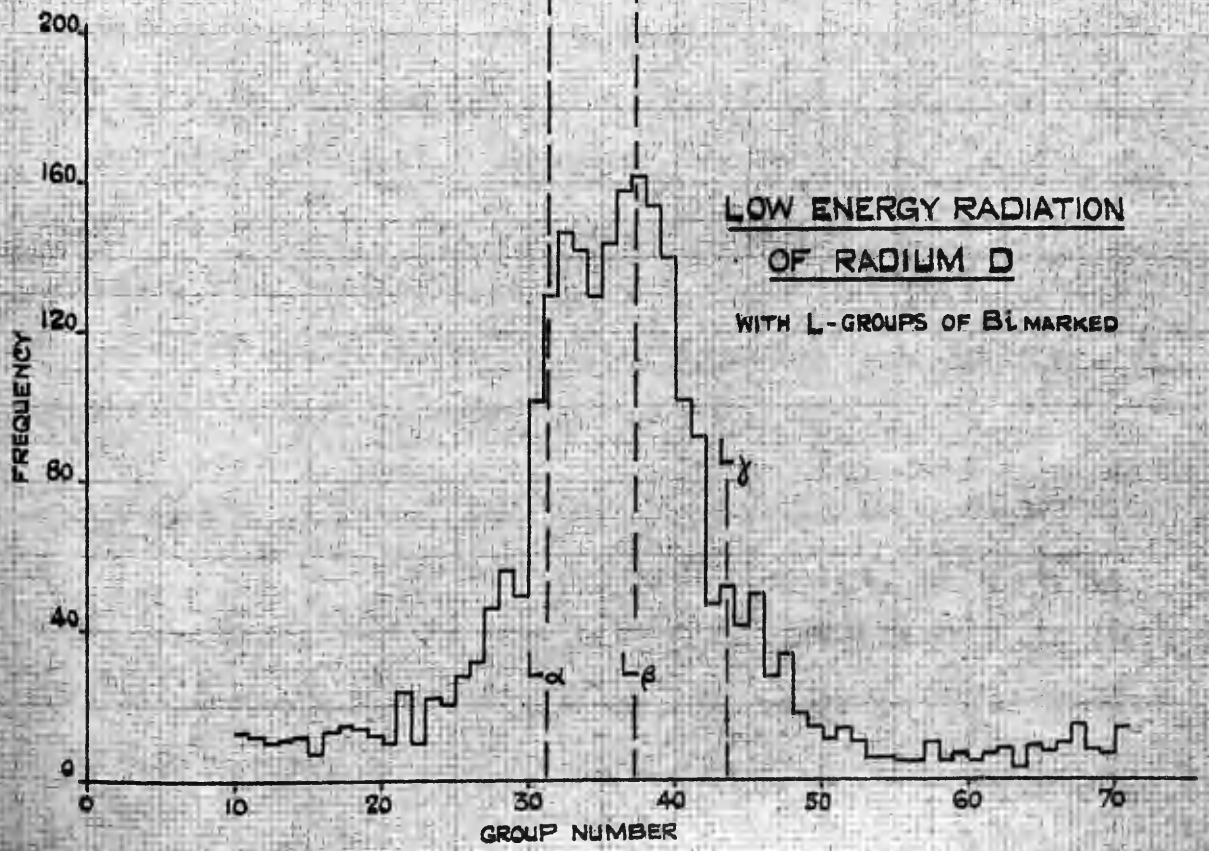




the source may also be resolved into two groups, again of approximately equal size, and the positions of the maxima correspond closely to those expected for the L_α and L_β lines of bismuth. They may therefore be assigned to radium E, and, since no evidence could be found of their presence in sources containing radium E alone, these X-rays must arise, as previously surmised, from the internal conversion of one or more of the gamma rays emitted from radium D.

The approximate equality of the intensities of the L_α and L_β lines emitted by the source is caused by the variation of absorption coefficient and detection coefficient of the counter, since it is observed also for the fluorescence X-rays, and there is no indication that the relative intensities of these X-rays are abnormal.

In at least one later experiment a group of pulses of very low energy were observed; these had energies in the region expected for the M X-rays of bismuth (2 - 4 kev), - no detailed measurements were carried out, - and indicate the presence of X-rays which would be expected from consideration of the secondary electron line spectrum.

Measurements of the 47 kev Gamma Ray.

The large number of X-rays observed, at least 30 times the background at peak intensity, made it appear possible that the gamma rays might also be measurable in spite of their much smaller intensity and the reduced efficiency of detection

of argon for gamma rays of this energy. The distribution in this energy region is shown in Figure 33 together with the X-ray distribution of the source, now unresolved due to the much narrower spacing of the groups. The group which appears at a pulse height of 45 units can be identified with the 46.7 kev radiation; the energy calculated from this figure is 46 kev, although the accuracy is not very high.

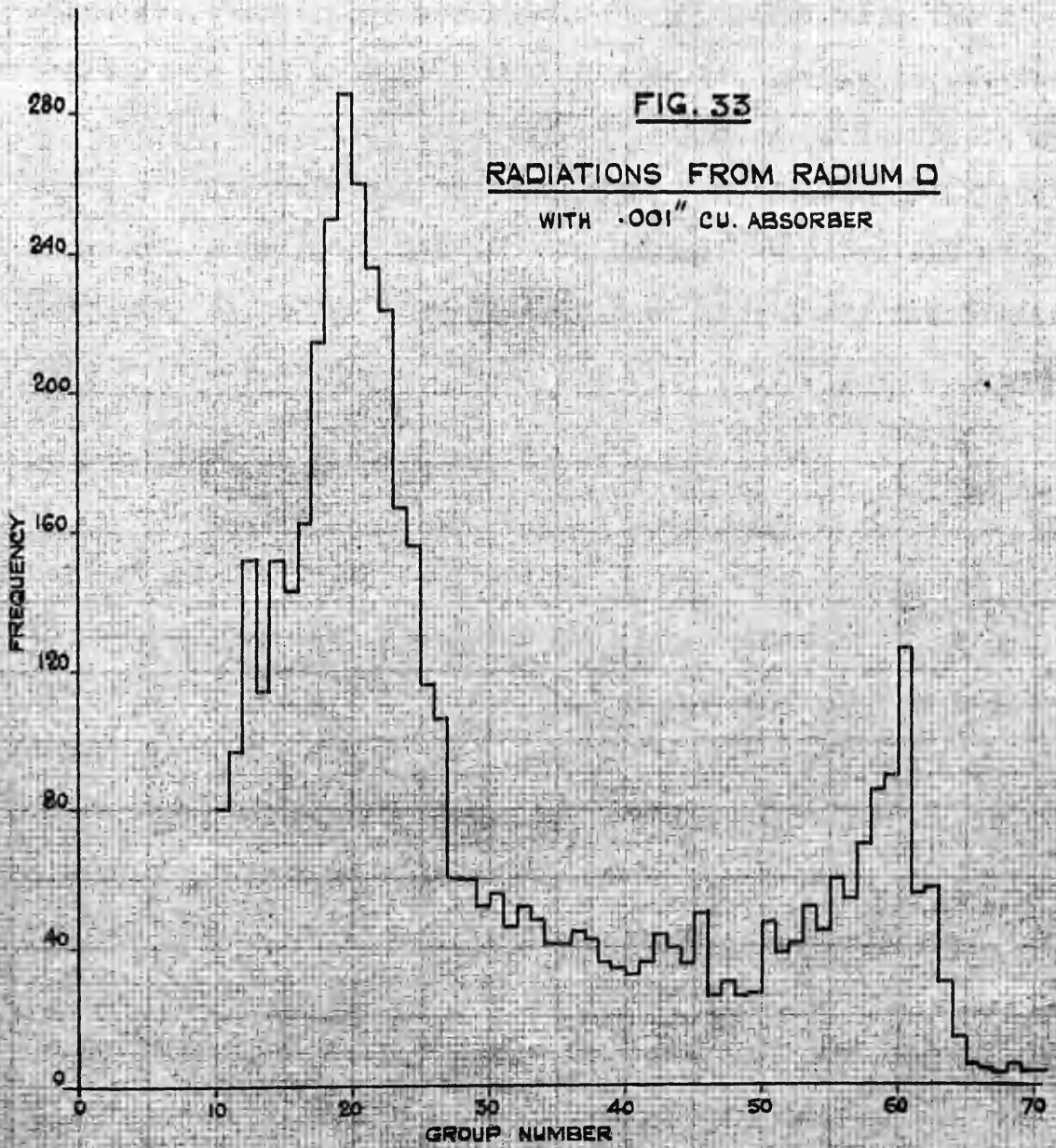
An interesting feature of the curve is the considerable assymmetry of the 47 kev group. The range of the photo-electrons released from the argon is $\sim$ 2 cms, and the radius of the counter used for these measurements was 3 cms.; it is therefore concluded that the assymmetry in favour of pulses of slightly smaller energies is caused by the limitations of the counter dimensions, and is not necessarily evidence for gamma rays between 35 and 45 kev.

The anomalies in previous investigations between the number of line electrons arising from this radiation and the number of X-rays made a further determination of the ratio of gamma rays to X-rays of some value. The number of pulses in the two groups was measured on two records, one in which the source was covered only by the polythene, and the second in which a further 0.001 in. copper absorber was introduced. The experimental ratio of the two intensities was calculated as 18 : 1 and 15.6 : 1 from the two measurements. When allowance is made for the ratio of the efficiencies of absorption in the

FIG. 33

RADIATIONS FROM RADIUM D

WITH .001" CU. ABSORBER



polythene and the window, and the efficiency of detection in the gas, the absolute values of the ratio become 12 : 1 and 9 : 1. A further experiment was carried out later in which the evaporated source was used in conjunction with the magnetic field and a counter with a mica window 0.0018 cm. thick. In this investigation only the efficiencies of detection of the two radiations and the absorption in air are involved, and this gave a ratio of 350 : 1. The final value of the absolute intensity ratio has been estimated as 11 : 1 and the limits have been set at 8 : 1 and 15 : 1.

Measurements on the 7.8 kev Gamma Ray.

In order to investigate the energy region between 0 kev and 20 kev in more detail the evaporated source was used in conjunction with a counter of 6 cm. diameter, which had a window of mica, 0.0018 cm. thick. The radiations from the source were collimated in a tube 1 cm. diameter and 6 cm. long, and a magnetic field was applied. The results of the investigation are shown in Figure 34. It is clear that, as well as the two groups at 48 units and 58 units, clearly identified as the L_α and L_β X-rays groups (at 10.8 kev and 13.0 kev), a further group of strong intensity appears. When the X-rays are used for calibration the value of the group number for this group (34.75 units) gives a value of 7.8 kev for the energy. The relative intensity of this group with respect to the two

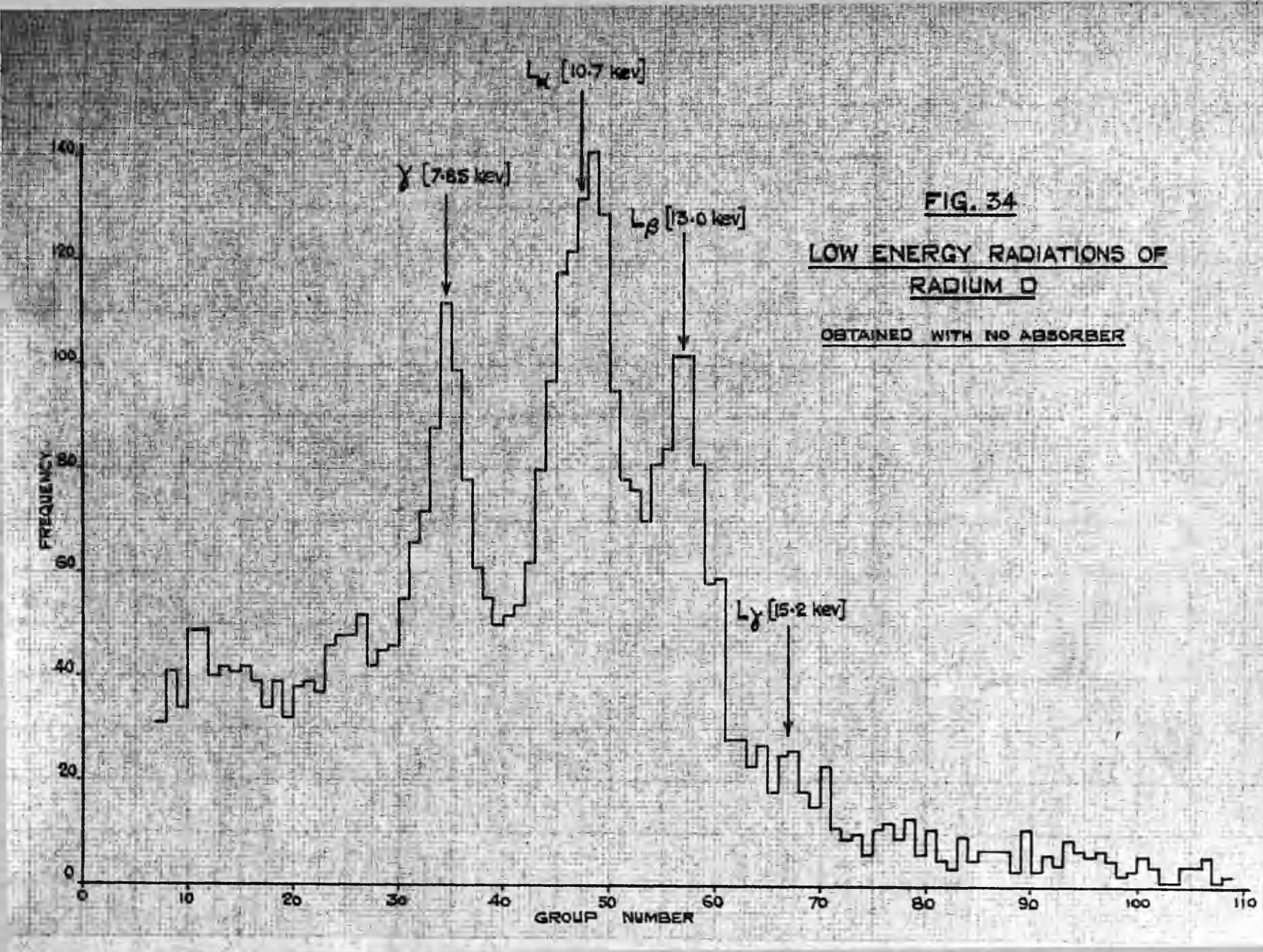


FIG. 34

LOW ENERGY RADIATIONS OF RADIUM D

OBTAINED WITH NO ABSORBER

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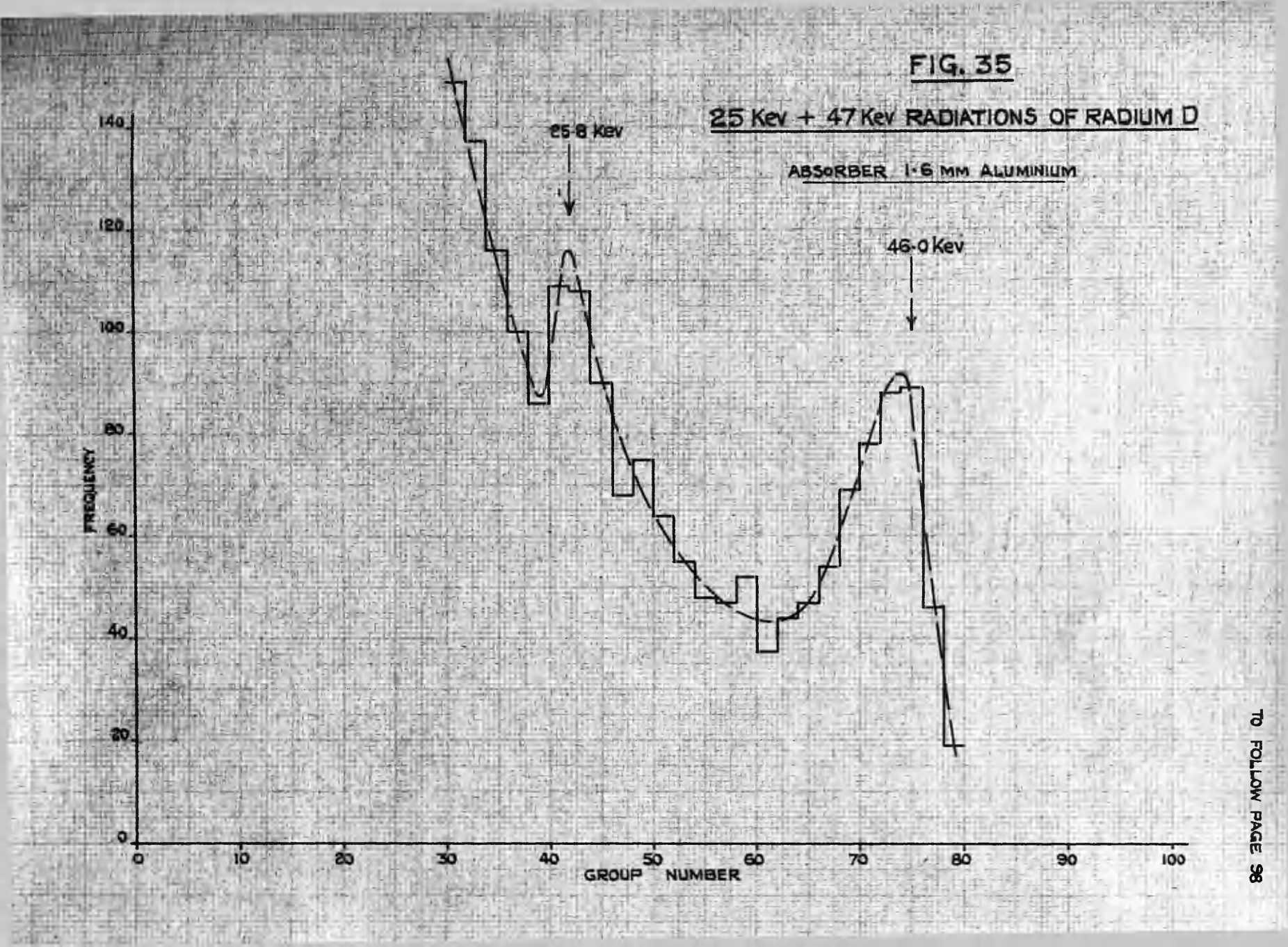
X-ray groups has been evaluated as 0.22 : 1, after correction has been made for the relative efficiencies of absorption of the radiations. In this distribution, it will be seen that the relative intensities of the L_α and L_β groups are in better agreement with the expected intensities, due to the small corrections which have to be applied. It seems possible that the group appearing at a height of 26 units may be caused by a further gamma ray of weak intensity, although the statistics are not sufficiently accurate to establish this; the group of particles between 10 and 15 units is undoubtedly caused by M X-rays, since their energies lie between 2.3 and 3.4 kev. Due to the complex nature of this group and the very small energy, no intensity value has been calculated, but this group appears to be relatively strong.

Measurements on the 25 kev Gamma Ray.

A further film was taken in which the evaporated source was covered with an aluminium absorber 1.6 mm. thick, in order to cut down the intensity of the X-rays and allow an investigation of the radiations between 20 kev and 50 kev. The results of this investigation are given in Figure 35; it will be seen that a group of rather small intensity appears at a height of 44 units, while the normal group caused by the 46.7 kev gamma ray occurs at a height of 72.5 kev. The energy of the weaker gamma ray has been estimated as 25.8 kev; while it is difficult to determine the intensity ratio from this curve,



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it is estimated as 1 : 8 with respect to the 47 kev ray, the limits placed on this value being 1 : 5 and 1 : 10. This gamma ray may be identified with the 23.2 kev gamma ray previously observed[47]; the intensity ratio does not agree with the ratio previously determined as 1 : 3. For the reasons already given, however, the new value is considered to be more accurate.

Conclusions.

The results may be summarised in the following table, in which the generally accepted value of 3 quanta per 100 disintegrations has been assumed for the 47 kev gamma ray[47]:-

Energy - kev	7.8	10.0 - 15.0 (L X-rays)	25.8	46.7
Intensity - no. per 100 disint^{ns}.	7.3	33	0.4	3.0

These figures are considered to be more accurate than those previously determined; they indicate, however, that the problem of interpretation remains, and a full understanding of the decay scheme cannot be given until a detailed energy distribution of both primary and secondary electrons has been obtained. It is clear that, over and above the conversion electrons established for the 46.7 kev gamma ray, conversion electrons of 11 to 16 kev will be present, together with Auger

electrons of 6 to 10 kev and a possible group of electrons arising from internal conversion of the 7.8 kev ray in the M-shell.

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SECTION 10

DISCUSSION OF RESULTS.

Since the results obtained for the various sources have already been discussed in the previous sections it is proposed here only to draw some general conclusions regarding the performance of the instrument and point out some of the future applications of this technique.

Firstly, the results obtained for the energy distributions of tritium and carbon indicate that the instrument may successfully be applied to studies of low energy beta-emitters - provided that the radioactive materials can be introduced into the counter as a component of a gas which will be sufficiently inactive to allow successful operation in the proportional region. If such a gas may be prepared the energy distribution obtained will be more accurate than that obtained by use of any alternative method at present available. Among such low energy emitters are sulphur 35, nickel 63 and radium D, each of which possesses some characteristic property which renders it of special interest.

Secondly, the results obtained in the study of the gamma rays of radium D show that corresponding accuracy may be obtained in the study of gamma rays of low energy. Where the gamma rays are complex energy and intensity measurements and determinations of the internal conversion coefficient may be

carried out in a small number of concurrent experiments. In a great number of cases the energy determinations have been made by means of secondary electron spectra and the proportional counter technique affords a direct determination of the gamma ray energy: this is of great value where there are several gamma rays of approximately equal energies and the line spectrum of the conversion electrons is indeterminate. The possibility of the excitation of nuclei by a synchrotron or Van der Graaf accelerator to give low-energy gamma rays from meta-stable states enhances the value of such an instrument, since direct energy and lifetime measurements may be made concurrently.

Thirdly, the investigation of the properties of this instrument shows a high degree of resolution for mono-energetic radiations, even for very weak intensity sources. While future developments of the technique may result in higher resolution, the method may clearly be used to give accurate data regarding radiations from weak sources; the rapidity with which an examination of such a source for low energy gamma rays may be effected, in comparison with the crystal spectrometer method, for example, should make the proportional counter an important instrument in the search for gamma rays emitted in radioactive transitions and those provoked by nuclear bombardment.

APPENDIX I.

STATISTICAL CONSIDERATIONS OF FLUCTUATIONS IN PULSE SIZE.

An analysis of the relative importance of the variations in gas amplification and the variations in the number of ion pairs formed by electrons of mono-kinetic energy is difficult unless certain initial assumptions are made. The following treatment has been carried out using certain alternative assumptions; it is not possible to determine the true nature of the variations arising in the gas amplification process, but the treatment is given in order to point out the assumptions which are necessary for a mathemetical formulation of the problem and the results which may be derived from such an analysis by statistical methods.

1. From the considerations put forward by Korff we have the expression:-

$$A = e^{\int \alpha \, dx}$$

If it is assumed that α is sufficiently constant to be written as $1/\bar{\ell}$, then $\bar{\ell}$ may be defined as the mean free path length in the radial direction between two multiplications. Also, in a region of high field strength it is assumed that every process is rigidly determined by this value, i.e., that once an electron has attained the required energy the probability of producing a second electron is unity. The main uncertainty in the gas amplification A

arises from an uncertainty of one mean free path length, since it is impossible to define the number of path lengths in a distance X to a higher degree of accuracy.

Since $\log A = \int \alpha x$ such an assumption leads to:- $\Delta \log A = \pm 1$

writing:- $$\sigma^2_{\log A} = \sum_{r \to \infty} (\Delta \log A_r)^2 / r$$

$$\sigma^2_{\log A} = 1$$

since:- $$\sigma^2_{\log A} = \sum_{r \to \infty} \left[\log(\bar{A} + \varepsilon_r) - \log \bar{A}\right]^2 / r$$

where $\bar{A}$ is the mean value of A,

$$\sigma^2_{\log A} = \sum_{r \to \infty} \log \left(1 + \frac{\varepsilon_r}{\bar{A}}\right)^2 / r$$

$$\approx \sum_{r \to \infty} \left(\frac{\varepsilon_r}{\bar{A}}\right)^2 / r$$

for small values of $\frac{\varepsilon_r}{\bar{A}}$

$$\sigma^2_{\log A} \approx \frac{\sigma^2_A}{\bar{A}^2}$$

where:- $$\sigma^2_A = \sum_{r \to \infty} \frac{\varepsilon_r^2}{r}$$

Hence, for the assumptions noted above,

$$\sigma^2_A = \bar{A}^2$$

2. If, however, we assume that the actual distances between collisions are ℓ_1, ℓ_2, ... etc., with a mean value of $\bar{\ell}$, and that:-

$$\sigma_\ell^2 = \Sigma\left(\ell_r - \bar{\ell}\right)_r = \left(K\bar{\ell}\right)^2,$$

then, for a number of mean free paths n, the variation in n may be written as:-

$$\sigma_n^2 \approx K\sqrt{n}$$

Now, since $A = e^n$, $\log A = n$, and:-

$$\sigma^2_{\log A} = K\sqrt{n} = K(\log A)^{1/2}$$

This value may be considerably greater or less than the value obtained from the first treatment, according to the value of K. While the expression shows a dependence on the value of A, this dependence is sufficiently small with respect to the other variations to allow it to be assumed constant. To a first order we may therefore write:-

$$\sigma^2_{\log A} = K$$

and hence:-

$$\sigma_A^2 = K\bar{A}^2$$

3. Having obtained these tentative relations for the mean square deviation in the gas amplification it is now possible to calculate the variations introduced by fluctuations in the numbers of ion pairs formed.

Suppose that n_r electrons are produced in an ionization event, and that the average number of electrons is $\bar{n}$, then:-

$$\sigma_n^2 = \sum \left(n_r - \bar{n}\right)^2 / r$$

The final number of electrons observed from an initial number n_r by gas multiplication may be written as:-

$$P_r = A_1 + A_2 + \ldots + A_j + \ldots + A_{n_r},$$

where the average value of P_r, $(\bar{P})$ is equal to $\bar{A}\bar{n}$ and the average value of A_j is $\bar{A}$.

$$\text{If} \quad \sigma_P^2 = \sum_{r \to \infty} \left(P_r - \bar{P}\right)^2 / r$$

$$\left(P_r - \bar{P}\right)^2 = \left(A_1 + A_2 \cdots + A_{n_r} - \bar{n}\bar{A}\right)^2$$

If $A_j = \bar{A} + \varepsilon_j$ this expression becomes:-

$$\left(n_r \bar{A} + \sum_1^{n_r} \varepsilon_j - \bar{n}\bar{A}\right)^2$$

$$= \left[(n_r - \bar{n})\bar{A} + \sum_1^{n_r} \varepsilon_j \right]^2$$

$$= (n_r - \bar{n})^2 \bar{A}^2 + 2(n_r - \bar{n})\bar{A} \sum_1^{n_r} \varepsilon_j + \left(\sum_1^{n_r} \varepsilon_j \right)^2$$

Therefore:- $$\sigma_P^2 = \sigma_n^2 \bar{A}^2 + \sum_{r \to \infty} \left(\sum_1^{n_r} \varepsilon_j \right)^2 / r$$

$$= \sigma_n^2 \bar{A}^2 + \bar{n} \sigma_A^2$$

since:- $$\left(\sum_1^{n_r} \varepsilon_j \right)^2 = n_r \Sigma \varepsilon_j^2$$

and $$\sum_{r \to \infty} \varepsilon_j = 0$$

If $\sigma_n^2 = \bar{n}$, and $\sigma_A^2 = \bar{A}^2$

$$\sigma_P^2 = 2\bar{n}\bar{A}^2$$

and $$\sigma_{P/\bar{P}} = \sqrt{2/\bar{n}}$$

If $\sigma_n^2 = \bar{n}$, and $\sigma_A^2 = K\bar{A}^2$

and $$\sigma_P^2 = \bar{n}(1+K)\bar{A}^2$$

and $$\sigma_{P/\bar{P}} = \sqrt{(1+K)/\bar{n}}$$.

APPENDIX 2.

EFFECT OF RESOLVING POWER ON THE END-POINT DISTRIBUTION OF TRITIUM.

The effect of the finite resolution of the counters caused by the finite number of ion pairs produced has been calculated on the following basis:-

(1) The number of ion pairs produced in the region 16 to 18 kev may be taken to be on the average $17 \times 10^3/_{30}$, i.e. 570.

(2) The X-ray distributions shown in Section 5 indicate that the full-width at half amplitude is approximately equal to $\sqrt{n}$; this corresponds to an energy of 700 volts.

(3) The distribution given in the inset of Figure 36 has been approximated by the grouping shown, and this grouping has been used in the calculations given below.

The Fermi distribution for neutrino mass zero gives the following values for the energy values quoted:-

E - energy (kev)	18.0	17.65	17.3	16.95	16.6	16.25	15.9
S (E) arbitrary	0	30	120	270	480	750	1,080

Each intensity value has been divided with its two immediate neighbours on both sides in the manner indicated in

the inset figure. The calculations, which are given in the table overleaf, give the distribution corrected for the finite number of ion pairs; these values are plotted in Figure 36, together with the true distribution. It will be seen that the shape of the spectrum remains substantially unaltered, but that the apparent end-point has been moved about 300 volts beyond the true value.

While a more accurate value would be obtained by the expression:-

$$S^1(E) = K.\int_{-\infty}^{\infty} S(E_r)_e^{-(E - E_r)^2/2A^2}\, dE_r,$$

where $A \simeq 1/3\sqrt{n}.\ dE/dn$, it is considered that the approximations used here give a very accurate estimation of the effect.

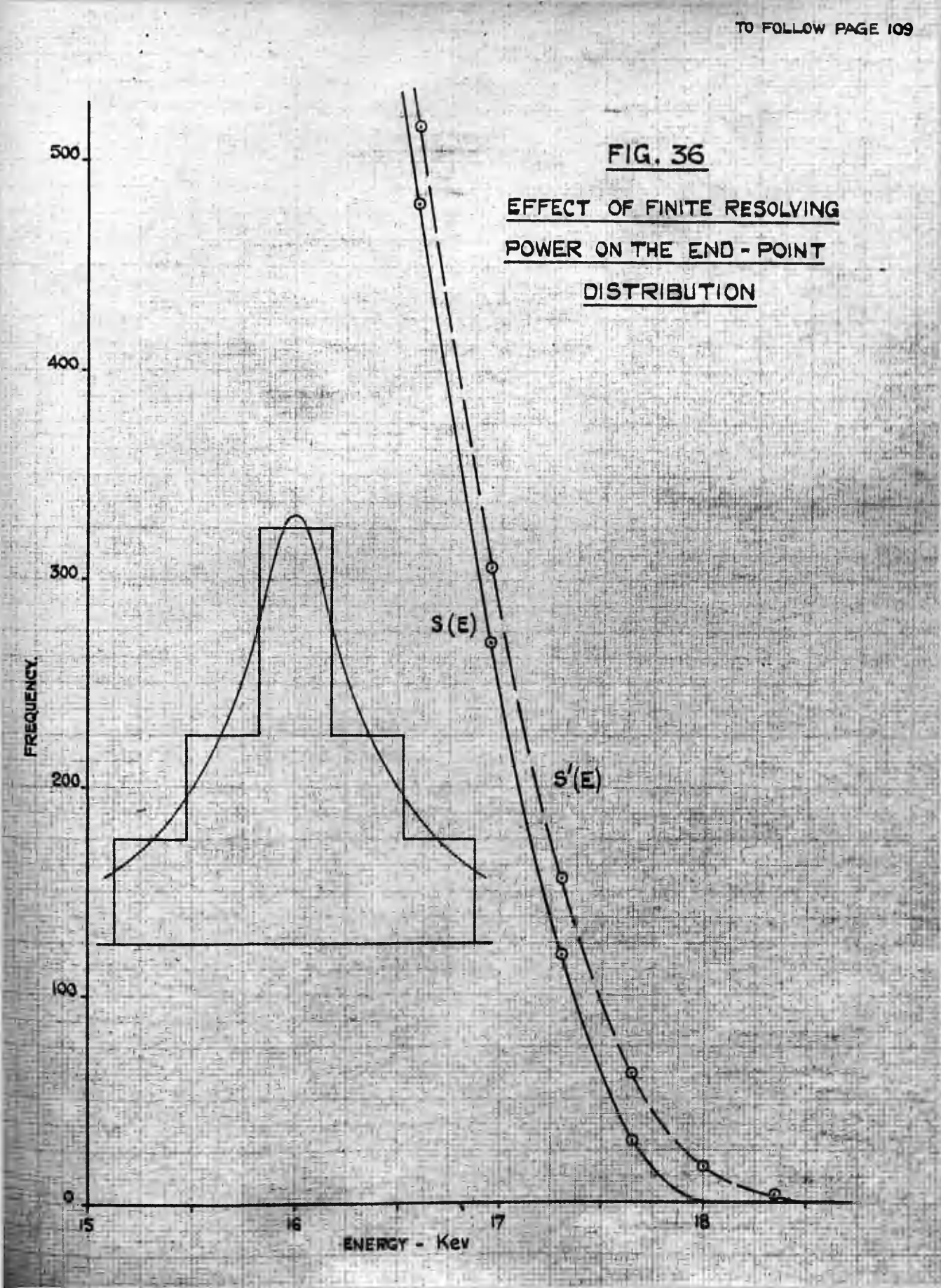

FIG. 36

EFFECT OF FINITE RESOLVING POWER ON THE END - POINT DISTRIBUTION

E								$S^1(E)$
15.2	108							-
15.55	216	75						-
15.9	432	150	48					-
16.25	216	300	96	27				-
16.6	108	150	192	54	12			516
16.95		75	96	108	24	3		306
17.3			48	54	48	6	0	156
17.65				27	24	12	0	63
18.0					12	6	0	18
18.35						3	0	3
18.7							0	0
S(E)	1,080	750	480	270	120	30	0	

Similar calculations were carried out on the curves shown in Figure 24.

APPENDIX 3

CHEMICAL SEPARATION OF RADIUM D.

The solution containing several microcuries of radium D, E and F in equilibrium was evaporated to dryness to remove any excess nitric acid. 2 cc. of concentrated hydrochloric acid was added and the solution evaporated to dryness; this process was repeated twice, and the residue was then dissolved in 0.1 N hydrochloric acid. Radium E and F were then partially removed by rotating in this solution a small strip of nickel of about 4 sq. cm. area.

20 cc. of a solution of lead chloride and bismuth chloride containing 0.2 gm. per litre were added to act as carriers of radium D and radium E respectively. This solution was evaporated to dryness with excess nitric acid to re-convert the salts to nitrates. Further evaporations with distilled water result in the formation of lead oxy-nitrate and bismuth oxy-nitrate. The residue from these evaporations was digested with 5 cc. of a solution of ammonium nitrate (2 gm. per 100 cc.) and this was allowed to stand for 15 hours. This treatment results in the solution of the lead content as lead nitrate, the bismuth oxy-nitrate remaining undissolved. Filtration of the solution results in the removal of the radium E, the filtrate containing radium D and the lead carrier. Evaporation of this filtrate on a source mounting gave an active deposit, the activity consisting almost entirely of the radiations of

radium D. At the very most, only one or two percent of radium E was present since the high energy beta-particles of this element were not detected in the experiments carried out with the proportional counter.