



<https://theses.gla.ac.uk/>

Theses Digitisation:

<https://www.gla.ac.uk/myglasgow/research/enlighten/theses/digitisation/>

This is a digitised version of the original print thesis.

Copyright and moral rights for this work are retained by the author

A copy can be downloaded for personal non-commercial research or study,
without prior permission or charge

This work cannot be reproduced or quoted extensively from without first
obtaining permission in writing from the author

The content must not be changed in any way or sold commercially in any
format or medium without the formal permission of the author

When referring to this work, full bibliographic details including the author,
title, awarding institution and date of the thesis must be given

Enlighten: Theses

<https://theses.gla.ac.uk/>
research-enlighten@glasgow.ac.uk

T H E S T U D Y O F B E T A - G A M M A

D E C A Y S C H E M E S

U S I N G P R O P O R T I O N A L C O U N T E R S .

BY

J. G. BALFOUR.

----oOo----

Presented as a thesis for the degree of
Ph.D. in the University of Glasgow.

September 1955.

ProQuest Number: 10656259

All rights reserved

INFORMATION TO ALL USERS

The quality of this reproduction is dependent upon the quality of the copy submitted.

In the unlikely event that the author did not send a complete manuscript and there are missing pages, these will be noted. Also, if material had to be removed, a note will indicate the deletion.



ProQuest 10656259

Published by ProQuest LLC (2017). Copyright of the Dissertation is held by the Author.

All rights reserved.

This work is protected against unauthorized copying under Title 17, United States Code
Microform Edition © ProQuest LLC.

ProQuest LLC.
789 East Eisenhower Parkway
P.O. Box 1346
Ann Arbor, MI 48106 – 1346

P R E F A C E.

This thesis contains an account of research carried out by the author at the University of Glasgow during the period October 1950 to October 1953.

After a short introductory chapter, a critical summary of previous work on β - and γ -spectroscopy is contained in chapter 2, and the need for coincidence methods in dealing with more complex decay schemes is discussed. The contents of these chapters are drawn mainly from original papers although use has been made of previous reviews.

In chapter 3 an account is given of a search for possible low - energy quanta emitted in coincidence with β -particles in the decay of tritium. This work formed the final part of a more extensive examination of the decay of tritium by Dr. G. M. Inch. The author's part in these coincidence experiments consisted in assisting Dr. Inch with the experimental work and with analysis

of the results.

Chapter 4 describes how the primary β - spectrum of RaD was isolated for the first time. The design of this experiment and preparation of the source were due to Dr. Inch; the experiment itself was conducted solely by the author; the analysis of the results was shared equally between Dr. Inch and the author.

Further elucidation of the decay scheme of RaD was practicable only by the aid of coincidence techniques. Chapter 5 gives an account of how a single - channel pulse analyser, built by the author during his early research programme, was developed to provide a satisfactory method of recording coincident events. The electronic circuits described here are due to Mr. R. Giles although several modifications were made by the author.

Application of the resulting equipment to the further study of the RaD decay scheme is described in chapter 6. The work described in this and in the succeeding chapters was conducted independently by the author although the method of approach was influenced by frequent discussions with Dr. S. C. Curran.

The use of solid radioactive sources, as in the experiments with RaD, leads to some uncertainty regarding the effect of back - scattering of electrons from the source support, and data on back - scattering generally available were inconsistent. The author therefore initiated independent experiments upon this general problem. The results of these experiments, described in chapter 7, are in fairly good agreement with those obtained by earlier workers using a source of β -particles placed outside a Geiger counter, but show marked disagreement with those which made use of a source inside a Geiger counter. Chapter 8 describes a series of experiments aimed at explaining this disagreement between the two types of Geiger counter measurement.

This thesis is not presented in strict chronological order. Due to the duration of the experiments on back - scattering, these were to some extent conducted in parallel with the coincidence examination of the RaD decay scheme.

The author wishes to express his gratitude to Professor P. I. Dee and Dr. S. C. Curran for the benefits

derived from their advice and criticism. He is also indebted to D.S.I.R. for a maintenance grant which made possible the three years of research.

P U B L I C A T I O N S .

- (1) The Beta - Spectrum of RaD.
Insch, G. M., Balfour, J. G., and Curran, S. C.
1952
Phys. Rev. 85,805.
- (2) The effect of back - scattering of electrons on
measurements in β -spectroscopy and absolute
counting.
Balfour, J. G. 1954 J. Sci. Instrum. 31,395

C O N T E N T S.

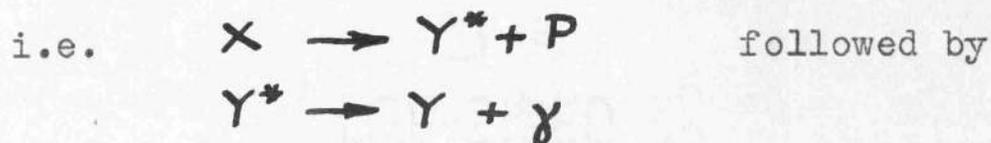
| | Page. |
|--|-------|
| <u>PREFACE</u> | (i) |
| <u>CHAPTER 1.</u> Introduction. | 1. |
| <u>CHAPTER 2.</u> Historical Survey. | 10. |
| <u>CHAPTER 3.</u> The β - decay of tritium. | 28. |
| <u>CHAPTER 4.</u> The decay of RaD. | 38. |
| <u>CHAPTER 5.</u> Development of coincidence technique. | 58. |
| <u>CHAPTER 6.</u> Application of coincidence techniques to the further examination of the RaD decay scheme. | 73. |
| <u>CHAPTER 7.</u> The effect of back - scattering of electrons in β - spectroscopy. | 89. |
| <u>CHAPTER 8.</u> Examination of possible secondary electron emission accompanying back - scattering of electrons. | 120. |
| <u>REFERENCES.</u> | 149. |

CHAPTER 1.INTRODUCTION.

Nuclear spectroscopy is a branch of nuclear physics which, at present, expresses mainly the results of experiments not completely related in the framework of a comprehensive theory. It is the analogue of ^{optical γ} X-ray spectroscopy in atomic physics. During the last century a vast amount of experimental results on ^{optical γ} X-ray spectroscopy was collected and it was not until the start of this century that theoreticians like Bohr related the various experimental results to an atomic theory. In nuclear spectroscopy, although there is as yet no comprehensive theory of the nucleus, experimental results are already numerous.

A nucleus, as shown by the phenomenon of radioactivity, can exist in different discrete energy states. The disintegration of a radioactive nucleus X to a nucleus Y, by the emission of a particle P, can leave the residual nucleus in an unstable excited state.

This nucleus can liberate its excess energy by the emission of one or more quanta, so returning to the ground state. The energies of these quanta determine energy levels of the particular nucleus Y.



In this thesis attention is confined to the case where particle P is a β -particle.

The aims of beta - gamma spectroscopy are threefold; the cataloguing of the radiations and decay schemes of all β -emitting nuclei; the investigation of properties such as spin and parity of particular nuclei to establish regularities of nuclear structure; and the investigation of selected transitions to study the interactions involved, especially in the case of β -transitions.

This thesis is concerned mainly with the methods of establishing β - γ decay schemes and particularly with the two techniques of most general importance:-

- (1) The measurement of energy spectra of β -particles and γ -rays.
- (2) The correlation of these spectra into a decay scheme.

β -SPECTRA.

β - γ decay schemes can be classified roughly according to whether the β -spectra are simple or complex.

SIMPLE β -SPECTRA: In this type, emission of β -particles occurs from the ground state of the parent nucleus to only one of the daughter nucleus. If the latter is in an excited state, de-excitation to the ground state occurs by the emission of one or several quanta per disintegration (Fig. 1(a)). P. 4.

COMPLEX β -SPECTRA: In this type, β -transitions occur between the ground state of the parent nucleus and more than one state of the daughter nucleus. (Fig 1(b))

The numbers of transitions leading to the different states are always in a certain ratio which is a measure of the relative probabilities of the two processes.

SPECTRUM SHAPE. The β -particles emitted by a radioactive source have a continuous distribution of energies with a definite maximum value. On the assumption that a neutrino is emitted simultaneously with each β -particle, both the maximum energy and shape of the energy spectrum can be accounted for by a theory of β -emission. The difficult part of such a theory lies in

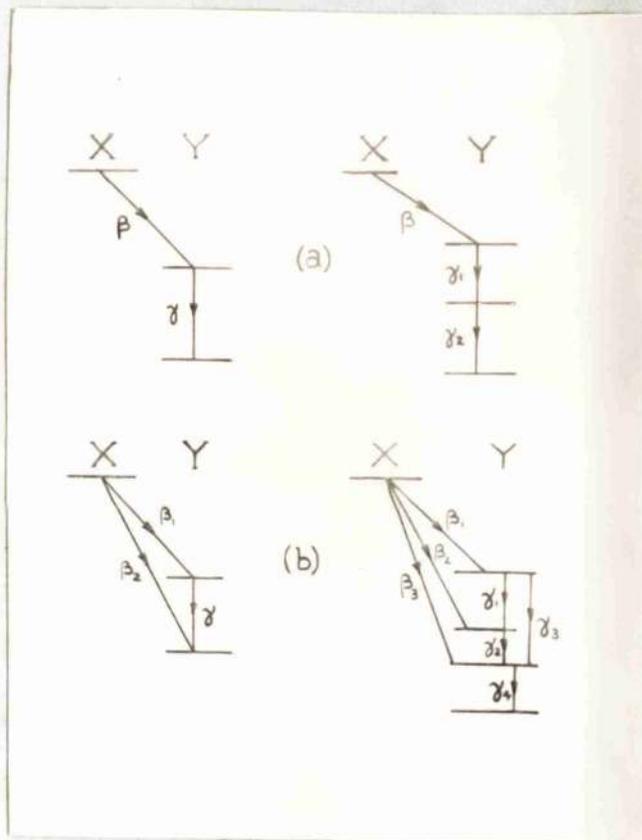


FIG. 1. (a) Simple β -spectra, and
 (b) Complex β -spectra,
 with one or more quanta per disintegration.

the choice of the type of interaction between the electron - neutrino field and the nucleons. Of the five possible interactions, the most commonly used are the polar - vector type originally assumed by Fermi and the tensor type favoured by Gamow.

Fermi theory predicts that the probability $P(W)$ of a β - particle of energy between W and $W + dW$ being emitted by a nucleus in time dt is

$$P(W)dWdt = G|M|^2 F(Z,W,R)W(W^2 - 1)^{1/2}(W_0 - W)^2 dWdt$$

where G is a constant expressing the strength of the interaction between the nucleons and the electron - neutrino field; $|M|$ is a matrix element, independent of W and dependent only on wave functions; and $F(Z,W,R)$ is a slowly varying function depending on the nuclear radius R and nuclear charge Z .

The matrix element can be expanded as a series of successively decreasing terms. For certain relations between the spin and parity of the initial and final nuclei the first term is non - vanishing, in which case the transition is termed "allowed" and the resulting spectrum an "allowed spectrum". Similarly ^{when the first} ~~non -~~ vanishing ^{term is the} 2nd, 3rd, ... ^{we have a} ~~term leads to a~~ first, second, ...

forbidden spectrum. The probability of decay decreases as the order of forbiddenness increases.

Because it is a measure of the energy difference between the two energy levels concerned in a β -transition, it is desirable to know the exact end - point W_0 of the β -spectrum as accurately as possible. In view of the shape of a spectrum in the region of the end - point this is a measurement open to some error. The so - called Kurie plot is of assistance towards the more accurate determination of end - point energy. If $N(W)$ is the number of β -particles observed in the energy range between W and $W + dW$ then it is seen from the above equation that if $\left[\frac{N(W)}{(W^2-1)^{1/2} \cdot W \cdot F(Z,W,R)} \right]^{1/2}$ is plotted against W the resulting graph should be a straight line cutting the axis at $W = W_0$ for the case of an allowed transition.

Good agreement is found between experiment and theory for the shape of an allowed spectrum but such agreement gives no information regarding the uniqueness of the five possible forms of interaction. In order to obtain the theoretical shape of a forbidden spectrum it is necessary to multiply the expression for an allowed

transition by a "correction factor" which does depend on the type of interaction assumed. Thus, if the Kurie plot of an observed spectrum is not straight, the various possible correction factors are applied with a view to deciding which of the interactions is the correct one to be assumed.

To summarise, the experimental examination of a β -spectrum should be conducted with a view to yielding:-

- (a) The exact end - point W_0 in order to establish the energy levels of the daughter nucleus.
- (b) The accurate shape of the spectrum in order to decide, if possible, which of the theoretical forms of interaction is correct. This in turn, through selection rules, can yield information on the spins and parities of the two energy states.

γ - RAYS ACCOMPANYING β - DECAY.

β - decay is almost invariably accompanied by emission of γ - rays and it is as important, in constructing decay schemes, to measure the γ -ray energies as to measure the end - points of the β - spectra. If

de - excitation takes place by internal conversion as an alternative to emission of quanta, measurement of the energies of the internal conversion electrons enables calculation of the γ -ray energies. This is important since the detection and energy measurement of these electrons is often more reliable and accurate than that of the alternative γ -rays.

The quantum theory of γ -ray emission uses the classical concept of a radiating source as an oscillating electric or magnetic moment, together with the classical method of analysing complicated spatial distributions into spherical harmonics of order 1, 2, l

The corresponding electric and magnetic multipoles are termed dipole, quadrupole, 2^l pole, these names also being applied loosely to the type of radiation emitted. As in β -decay theory this procedure gives an expression which can be expanded as a decreasing series, the first, second, ..., l terms giving the probability for emission of dipole, quadrupole,, 2^l pole radiation. Also in a way analogous to β -decay theory, certain relations between spin and parity give a set of selection rules.

The decay constant for the transition depends

strongly on l . In theory, measurement of half life of the transition, together with the energy change, could give a measure of the spin change involved, but for small spin change ($l = 1$ or 2) the half life is immeasurably short ($< 10^{-13}$ sec) and only in the special case of isomeric states ($l > 3$) is the half life measurable.

However, in the case of internal conversion, coupling between the nucleus and the orbital electrons is strong, and independent checks on multipole order can be made either by measurement of absolute values of K - conversion coefficients, α_K (Rose et al, 1951), or from the ratio of K- to L- conversion. Both methods are particularly applicable to low energy transitions (< 300 keV). The K/L ratio is dependent not only on multipole order but on whether the transition is magnetic or electric, particularly for higher order transitions.

In brief therefore, in the study of $\beta - \gamma$ decay schemes, examination of γ - rays is made with a view to:-

- (a) Fixing the energy levels, from determination of the γ - ray energy.
- (b) Obtaining information regarding spin and parity of the initial and final states, particularly if the γ - ray is internally converted.

C H A P T E R 2.HISTORICAL SURVEY. β -RAY SPECTROSCOPY.

During the development of β -spectroscopy various techniques have been used, each with its own merits and shortcomings.

The absorption method consists in plotting the absorption curve obtained by interposing increasing thicknesses of aluminium foil between the source and a Geiger counter. Although much early information was obtained by the absorption method, it is of limited usefulness and accuracy. Erroneous conclusions may be made unless great care is exercised, both in obtaining the absorption curve, and in interpreting it. Due to scattering in the absorbers the electrons quickly lose homogeneity of direction and energy, with resulting poor resolution. In fact the nature of the spectrum may be such that it is almost impossible to determine the end - point.

The magnetic spectrometer method overcomes many of the difficulties due to scattering of the electrons, since measurements are made in a vacuum chamber. There are many variations of the technique, mainly in the method by which electrons of the same energy are brought magnetically to the same focus, but it can be said, in general, that the order of performance is about the same for all. Usually the transverse type of focussing is preferred for high resolution whereas the longitudinal or lens type has the advantage of higher transmission in dealing with weaker sources. Excellence of focus is only one part of the problem, and good results depend more on the careful preparation of the source and detector, than on superior focussing.

With magnetic spectrometers various factors combine to cause a false suppression of an observed β - spectrum towards the lower energy regions. For this reason the magnetic spectrometer is not capable of the most accurate results at low energies ($< 20\text{keV}$ say). However the resolution of 1 per cent or better has given the most accurate results in β -spectroscopy, particularly in the energy region 100keV to 5MeV . If the spectrum consists of two component spectra, the individual end - point

energies can be obtained fairly accurately. If there are more than two spectra present the possible accuracy is reduced.

The results obtainable at low energies are somewhat better when focussing is effected electrostatically (Purcell, 1938; and Backus, 1945). Generally, however, electrostatic focussing of this type is not popular, mainly because the very high fields required render the technique hardly practicable.

The difficulties of detection and energy measurement of low - energy β - particles was largely overcome through the full exploitation of the properties of the proportional counter by Curran, Angus and Cockroft (1948, 1949(a), 1949(b)). Some of the difficulties encountered at low energies with the magnetic spectrometer, namely, absorption of energy in the window of the counter, in the source itself, and scattering in and near the source, can be overcome by introducing the sources into the counter in gaseous form, provided that the gases or vapours introduced as sources do not exhibit electron affinity. In this way an efficiency of detection of 100 per cent is afforded down to energies of a few hundred electron volts. A difficulty encountered in the form of reduction

of gas multiplication near the ends of the counter could be allowed for (Angus, Cockroft and Curran, 1949). Later Cockroft and Curran (1951) modified the electrode structure and the resulting counters provided uniform gas gain along their whole active length.

Solid sources may also be introduced into the counter. The source, mounted on the thinnest possible nylon film, or on solid metal, is supported on a probe, whose potential is adjusted to suit its position in the electric field. Depending on the type of source support, the acceptance solid angle is either 4π or 2π respectively.

An extension of this technique, namely the introduction of a source deposited on the wall of the counter, to combine a solid angle of 2π and a large surface area, has been used in the examination of sources of very low activity (Curran, Dixon, and Wilson, 1952).

Although primarily used at low energies, the range of usefulness of the proportional counter can be extended by increasing the pressure of the gas filling, pressures up to 7 atmospheres having been used without much difficulty (Cockroft, unpublished). An additional aid in

confining the range of the particles within the counter was provided by Curran, Cockroft and Insch (1950), who established the value of placing the counter in a magnetic field.

These various advantages of the proportional counter provided β - spectroscopy with a powerful additional tool. To a large extent the instrument is most suitable in the low energy regions in which the magnetic spectrometer proves least accurate. In general however the magnetic spectrometer has the advantage of better resolution.

In the past few years crystals have been made which enable the detection and energy - measurement of β - particles by scintillation spectrometers. At present the method offers no major advance on previous techniques of β - spectroscopy, particularly at low energies where the resolution is very low (about 65 per cent at 10keV for a crystal of sodium iodide).

γ - RAY SPECTROSCOPY.

The diffraction method is the most fundamental in yielding precise results, because the wavelength of the γ - rays is measured in terms of the lattice spacing of a crystal. The method is limited by the necessity of

using strong sources and by the fact that the angles decrease with increasing energy, thus causing an upper limit to energy measurement of $\sim 1.3\text{MeV}$. Using the focussing effect of a bent crystal the diffraction method is a really accurate device applicable in the energy range 0.015 to 1.3MeV .

As in β - spectroscopy much of the early work of γ - ray measurement was done by absorption methods. The absorption of the γ - rays in a material is measured and the results compared with those obtained with γ - rays of known energy. Care has to be exercised because of the ambiguity caused by the fact that the absorption coefficients have a minimum value. Because of the slow variation of absorption coefficient with energy above $\sim 1.5\text{MeV}$ the reliability of the method is doubtful above this value. The method becomes of little value if more than one γ - ray is present.

Use can be made of the fact that, on passing over a K - absorption edge, absorption coefficients vary by a factor ~ 5 , the variation being rapid on either side. A γ - ray can thus be isolated between the K - absorption edges of materials of neighbouring Z value. The energy is then fixed within limits having the ratio $\frac{Z}{Z+2}$.

Accurate measurements are possible by this method but the useful range ($\sim 1\text{keV}$ to $\sim 80\text{keV}$) is rather limited.

Absorption methods are of little value if more than one energy of γ -ray is involved. This is no great obstacle for the magnetic spectrometer applied to γ -ray spectroscopy. Use is made of its high resolution for electron groups together with the aid of two effects, namely, production of Compton electrons by γ -rays in materials of low Z, and production of photoelectrons by γ -rays in materials of high Z. By employing both effects, assignment of the various photoelectron peaks to K, L, or even M levels, is simplified by reference to the Compton spectrum. Within the energy range 50keV to 20MeV the accuracy of energy measurement is 1 per cent or better, and lines separated by 10 per cent of their energy can be resolved. Even better resolution is possible if the γ -rays are internally converted.

Together with the bent crystal spectrometer, the magnetic spectrometer provides the present really accurate instrument of γ -spectroscopy. Both, however, require fairly strong sources for accurate results. The other two instruments used, namely the proportional counter and the scintillation counter, are more efficient and can be

used ~~in conjunction~~ with good geometry (2π or 4π). As regards resolution however both the proportional and scintillation counters are comparatively approximate devices.

Before the work of Curran, Angus, and Cockroft (1949) with proportional counters, the investigation of soft γ -rays was rather difficult, particularly if it was necessary to use weak sources. Because of the large coefficient of absorption at energies below 150keV, high efficiency of detection of γ -rays is impossible by the proportional counter. For energies of ~ 3.5 to 10keV, argon as a filling gas is suitable, the efficiency of detection being ~ 55 per cent. At higher energies (~ 100 keV) larger counters, higher gas pressures, or heavier gas (eg. xenon) are employed. The proportional counter is especially applicable when the γ -ray is internally converted, again particularly in the lower energy range.

Generally the most efficient detector of γ -rays at present is the scintillation counter. In addition, the measurement of energy is capable of considerable accuracy (Hofstater and McIntyre, 1950). Below ~ 20 keV the proportional counter has the better resolution but

the resolution of the scintillation counter improves with increasing energy, reaching about 8 per cent at 1MeV.

The importance of high resolution in γ -ray spectroscopy has been pointed out by Deutsch (1952) with regard to using the combination principle ~~to~~^{for} the location of levels. In ¹³¹I six γ -rays were known. If these are detected with an uncertainty of 1.5 per cent there are 7 combinations which might be interpreted as alternative transitions between the same initial and final states. With an accuracy of 0.5 per cent, 4 of these remain.

THE USE OF COINCIDENCE TECHNIQUES IN DETERMINING β - γ DECAY SCHEMES.

Once the maximum β -particle energies and γ -ray energies of a particular disintegration have been determined by means of the above methods, it is necessary to consider them with reference to a possible decay scheme. Sometimes in early work it was possible to group the γ -rays into a self - consistent decay scheme by means of their energies alone. Almost invariably however there is a certain arbitrariness in decay schemes deduced from the measured γ -ray energies. In the case of ¹³¹I, even with a resolution of 0.5 per cent, application of the

combination principle leaves four possible decay schemes. Fortunately the development of coincidence techniques has provided a means of further examination of this decay scheme and it has been shown by coincidence methods which of the four possible decay schemes is the correct one.

Sometimes it is necessary to examine a source where a series of short lived radioactive elements are in equilibrium with a parent of longer decay period. In such cases, chemical separation may be difficult and of no value owing to the very short lifetimes involved. In such cases, coincidence methods can be of assistance in examining only the particular decay of interest.

The study of β - spectra does not afford a very accurate determination of energy levels when two or more superimposed β - spectra have to be analysed into components. Often it is possible, by examination of coincidences between β - particles and associated γ - rays, to separate out the component spectra and make possible a more complete solution.

These various factors have led to considerable development of coincidence techniques as an aid to more exact determinations of β - γ decay schemes.

Two or more ionising events, such as the emission of a β -particle or γ -ray, are said to be in coincidence if they occur simultaneously, the simultaneity being defined only by the precision of the apparatus. For example the time taken for de - excitation by γ -ray emission is $\sim 10^{-13}$ sec (except for isomeric states), and so a γ -transition occurring in relation to a β -transition, or to another γ -transition, will be observed as a single coincidence count by a coincidence circuit of resolving time greater than 10^{-13} sec. This gives a means of determining whether a particular γ -ray is related to a β -spectrum. Similarly if two γ -rays are observed, it becomes possible to establish whether they are in cascade or are competitive modes of de - excitation. If they are in competition they will be recorded as coincidence merely by chance, and because of the finite resolving time of the coincidence circuit.

In this connection it is to be noted that if two counters, having separate counting rates of N_1 and N_2 , are connected to a coincidence circuit of resolving time τ , then a number of chance coincidences, N_c , will be recorded, where

$$N_c = 2\tau N_1 N_2$$

In addition a relatively small number of chance

coincidences will be recorded due to energetic cosmic rays being detected by both counters. In practice it is necessary to correct coincidence observations for both of these false types of coincidence. The latter may be reduced by placing lead shielding around the counters and by suitable adjustment of the geometry of the counters. The former effect can be minimised by using as low a value of resolving time, τ , as is practicable. The resolving time, however, is dependent on the time taken for sufficient of the output pulse from the counter to form, in order that it may operate the coincidence unit. This in turn is a function of the type of counter and the manner in which it is used. A resolving time of about 10^{-6} sec is often practicable with Geiger or proportional counters whereas a figure of $\sim 10^{-9}$ sec is possible with a scintillation counter.

It is natural to expect that the number of chance coincidences should be kept less than the number of true coincidences. Analysis of this point by Dunworth (1940) showed that no advantage is gained by using a source of strength greater than $N = \frac{1}{2\tau}$. With $\tau \sim 10^{-6}$ sec this gives a maximum source strength of $\sim 100 \mu$ curies.

DEVELOPMENT OF COINCIDENCE TECHNIQUES.

The method of coincidences was first used by Bothe and Geiger (1925) to show the simultaneous ejection of an electron and the emission of a scattered quantum in the Compton effect of X-rays. A coincidence method for the determination of the absorption of secondary electrons ejected by a γ -ray was ~~deduced~~ ^{introduced} by ~~Bothe and Kolhorster~~ ^{Becher (1932)} (1928). Worthy of particular mention in the early development of coincidence methods is the work of Rossi (1930) who perfected the necessary electronic apparatus and extended it for use with multiple coincidences.

From around 1930 coincidence counting was used in cosmic ray work, in determining quantum energies, and in investigation of Compton effect. In all of these experiments a single particle operated both counters, either directly or indirectly. A new application of coincidence techniques was made by Bothe and von Baeyer (1935). Two different particles from the same nucleus were examined, namely the coincident protons and photons in the reaction



Bothe and his co-workers went on to study the coincidences between the β -rays of a spectrum and the γ -rays

emitted simultaneously (Maier Leibnitz, 1936).

Later the use of the method increased, became of general use, and was much improved from the point of view of technique. Around 1940 there appeared several articles by Dunworth (1940), Norling (1938), Maier Leibnitz (1936), Mitchell, Langer and McDaniel (1940). In the new applications of the method, mention should be made of the study of angular correlation between two γ -rays emitted in cascade, and the measurement of internal conversion coefficients with the aid of β - e^- coincidences. The latter application, first used by Norling, was developed by Weidenbeck and Chu (1947). The position up to 1948 regarding β - γ coincidence techniques has been well reviewed by Mitchell (1948).

The early examination of β - γ decay schemes was done by absorption methods, two Geiger counters being placed near the source. An aluminium absorber eliminates β -particles from the γ -ray counter and the γ -ray energy is obtained by measurement of range of the Compton electrons ejected from the aluminium foil. Energy of the β -particles is measured by interposing various thicknesses of absorber between source and counter. The procedures for dealing with different forms of β - γ decay scheme has been well detailed by Mitchell (1948).

It is possible to measure the maximum energies and relative probabilities of the two component spectra if the β - spectrum is complex with only two components. If, however, there are more than two component spectra, analysis of the results becomes a problem of considerable difficulty. It is possible to determine whether the highest energy β - transition goes to the ground state or to an excited state, but beyond this little can be deduced.

The method has the advantage that it is possible to use sources of lower activity than can be employed with the higher resolving-power, and more accurate, magnetic spectrometer. It does, however, suffer from the shortcomings of all absorption methods, the main weakness being in the relatively low accuracy of energy measurement. Information obtained by the absorption method, taken together with the results from a magnetic spectrometer, still provides one of the best methods of determining decay schemes.

A magnetic spectrometer in coincidence work was used by Bothe and Maier Leibnitz (1936(a) and (b)) to resolve a complex β - spectrum. Measurements were made

of the coincidences between γ - rays, detected by a Geiger counter placed near the source, and successive energy intervals of the β - spectrum (detected by the spectrometer). A weakness of the method lay in the very low coincidence counting rate obtained. The reasons for this were twofold, namely, low transmission of the spectrometer and low efficiency of detection of the γ - rays. Two magnetic spectrometers were used by Feather, Kyles and Pringle (1948). Coincidences were examined between internal conversion electrons, recorded in one spectrometer, and various parts of the β - spectrum recorded in the other. Such application of the method is capable of great accuracy although the coincidence counting rates were low. The spectrometers used were of low transmission. Some improvement ~~could be~~ ^{has been} made by using the greater transmission of magnetic lens spectrometers.

While the use of spectrometers with coincidence techniques gives more accurate and detailed information than is possible by the absorption method, it is not much more powerful in analysing complicated decay schemes.

Exploitation of the scintillation counter has led to great advances in coincidence techniques in recent

years. The associated photomultiplier is superior in many ways to the conventional type of amplifying system, due to the very short pulse duration. Consequently resolving times $\sim 10^{-9}$ sec are possible. On account of its very high efficiency of detection of γ -rays, the scintillation counter is particularly applicable to the study of γ - γ coincidences. In view of the poor resolution with low energy β -particles, coincidence methods are best applied to high energy β -transitions. However low energy decay schemes have been examined by this means (Bannerman and Curran, 1952).

It is seen that application of coincidence techniques makes possible more thorough study of β - γ decay schemes than is possible by separate investigation of the several radiations. As regards the type of detecting instrument to be used, there is no one method of universal applicability. Ideally coincidence measurements should be made with two detectors each sufficiently selective to detect one radiation alone with a minimum of interference from the others. Each decay scheme has to be considered on its own merits in choosing which instrument is to be employed.

In this thesis attention is confined to β - γ

disintegrations of low total energy. The power of the proportional counter in dealing with both β - particles and γ - rays of low energy has been noted. Application of the proportional counter, together with coincidence methods, to examination of low energy β - γ decay schemes appears to be a matter worthy of attention. At the start of the work described in this thesis, such application had received little attention. The problems and results of this development form the pattern of the following chapters.

CHAPTER 3.THE β - DECAY OF TRITIUM.1. INTRODUCTION.

The β - decay of tritium is an allowed transition. An examination of the β - spectrum by Curran, Angus, and Cockroft (1949, a) showed that between $\sim 1\text{keV}$ and the upper energy limit, the spectrum is in good agreement with Fermi theory. These authors, however, made a tentative suggestion of some deficiency in the number of β - particles in the lower energy region.

An examination of the various published spectra to date led Cooper and Rogers (1950) to conclude that agreement with Fermi theory was excellent between 2.5keV and 18keV .

A re - design of the proportional counter electrodes by Cockroft and Curran (1951) made possible an examination of spectrum shape to lower energy limits than previously, without appreciable uncertainties arising from spuriously

small pulses. This was done by Insch and Curran (1951) who obtained reliable results down to energies of $\sim 400\text{eV}$. Below $\sim 1\text{keV}$ the intensity of the spectrum appeared to be considerably less than expected on the theoretical grounds. In view of the unexpected results they repeated the experiment with a view to establishing whether the deficiency of low energy particles could be due to limitation of the amplifier, the recording technique, or uncertainty of "end effect". The results obtained by the different methods were in good agreement (Fig 2). Theoretically, at zero energy the intensity of the β - spectrum should be 86 per cent of its maximum value. After taking into consideration the experimental accuracy (10 per cent, even at 400eV) Insch and Curran concluded that the intensity of the spectrum below 1keV is definitely less than predicted theoretically.

The assumption that the counter was, in fact, acting proportionally down to the lowest energy values was confirmed in the light of existing data. Explanation of the spectrum shape was therefore sought on the hypothesis that, in coincidence with the β - particles, quanta or particles of low energy are

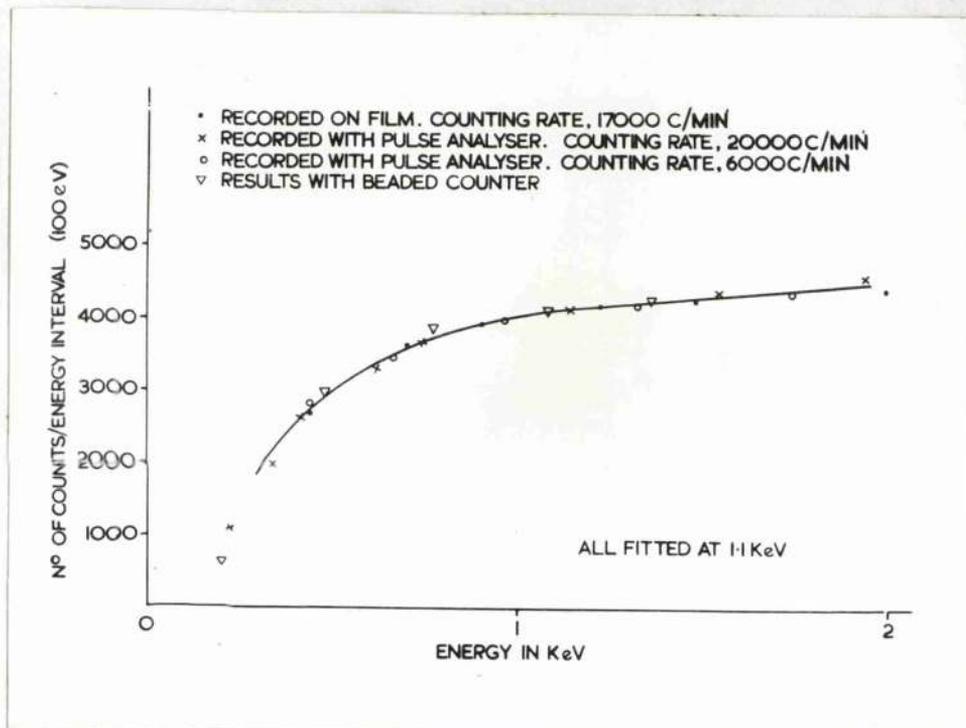


Fig 2. The low energy end of the β - spectrum of tritium as observed by Insch and Curran (1951).

emitted. If the quantum radiation were of energy $\sim 400\text{eV}$ it would give rise to a peak, on the observed pulse distribution, centred on this energy. If these quanta were emitted in coincidence with the β -particles then, due to the property of the proportional counter (of integrating two ionising events occurring within $\sim 10\mu\text{sec}$ of each other to give a resultant output pulse proportional to the sum of the two energies), the resultant spectrum would have a shape similar to that observed.

At this stage the author joined in the experimental confirmation, or otherwise, of this coincident - quantum hypothesis.

2. APPARATUS.

The proportional counter used (Fig 3(a)) consists of a cylindrical cathode 5 ins in diameter and 16 ins in effective operating length. The counting tube is lined with aluminium sheet to minimise secondary effects at the cathode. For calibration purposes a thin aluminium window is fitted at the centre of the counter.

A glass bead $\sim \frac{1}{2}\text{mm}$ in diameter and 1.5cm long divides the wire into two equal sections insulated

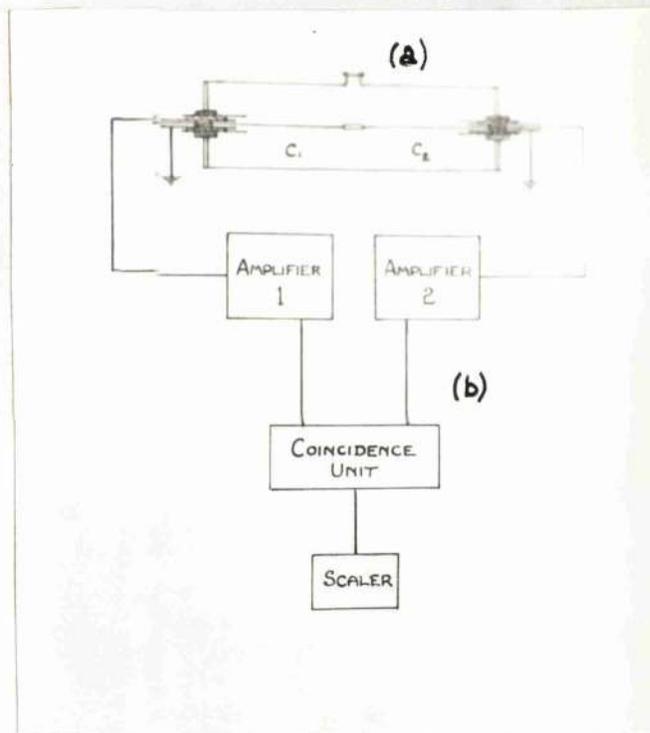


Fig 3. (a) Proportional counter used in coincidence examination of the decay of tritium.
(b) Schematic diagram of circuit used.

electrically from each other. The counter is thus divided into two similar, but independently operating, counting volumes. The pulses from each counter, after suitable amplification, are fed into a coincidence unit, the output pulses from which operate a scaler directly (Fig 3(b)).

3. EXPERIMENTAL PROCEDURE.

Tritium, together with a trace of helium to act as a carrier, was introduced into the counter, and methane added to give a total pressure of 1cm mercury.

Due to the extremely high absorption of quanta of energy $\leq 400\text{eV}$ it is extremely difficult to establish their presence by coincidence methods. Values for the mass absorption coefficient of methane for such low energy quanta were not available, but it seemed likely that, if quanta of energy $\sim 400\text{eV}$ (but not greatly less than this) were present, coincidences would be obtained. In order to ensure that no $\beta-\beta$ coincidences were obtained due to a β -particle travelling from one counting volume to the other, the counter was placed with its axis perpendicular to a magnetic field of strength sufficient to prevent this.

The counting rate was $\sim 10,000$ counts per minute in

each counting volume, and the counter operated satisfactorily at the low pressure with about 1.5 kilovolts on the case. At this working voltage the gas gain was sufficient to detect radiations of energy as low as 40eV. Calibration was achieved by reducing the amplifier gain and firing K_{α} fluorescence X-rays of copper through the central window.

4. RESULTS.

A measure of coincidence counting rate was made as a function of voltage applied to the case, i.e. as a function of the minimum energy recorded. As this voltage was increased, however, the number of coincidences increased markedly. The curve obtained (Fig 4) by plotting the ratio, R , of coincidence counting rate to total counting rate in terms of voltage, indicates that, on increasing the voltage, the counters lose their independence and that the coincidences observed are due to "mutual firing" of the two counting volumes.

Confirmation of this interpretation was obtained by firing K_{α} fluorescence X-rays of copper through the central window, the counting rate due to the X-rays in each counter being approximately the same as that due to the tritium. The K_{α} X-rays were chosen since they give

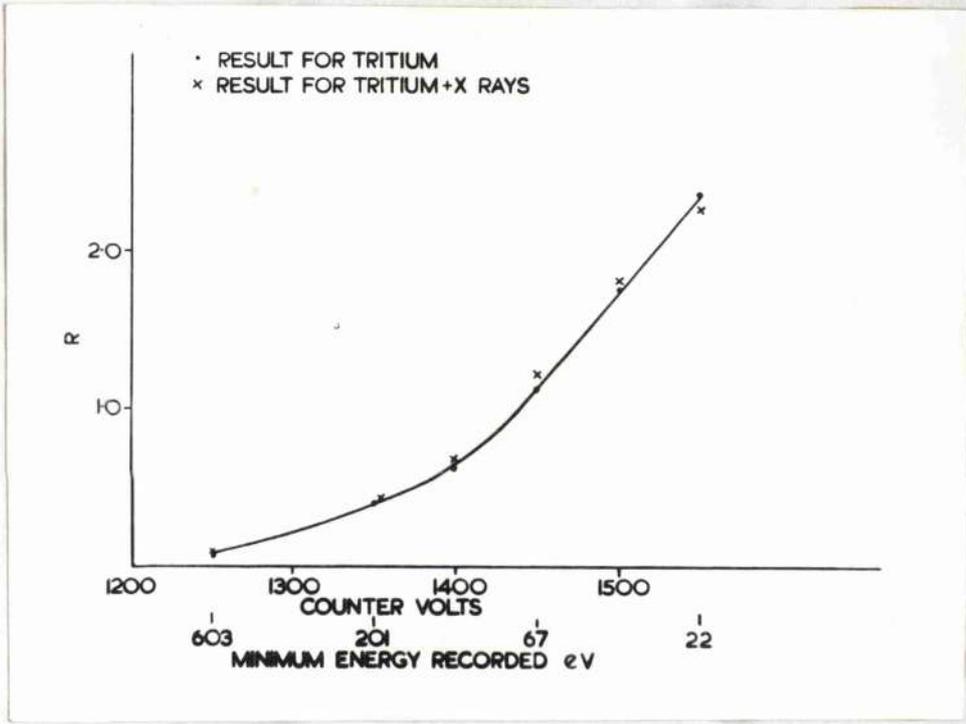


Fig. 4. Ratio, R, of coincidence counting rate to total count rate, as a function of counter voltage.

the same average pulse size as the β - particles of tritium. The points on the curve obtained in this way coincided with those found previously, within the accuracy of the experiment. The very close agreement between the two sets of points in Fig 4 shows that very few, if any, of the coincidences observed with tritium can be ascribed to the supposed β - quantum decay process.

5. CONCLUSION.

The technique employed has failed to establish the reason for the anomalous shape of the β - spectrum of tritium at low energies.

Even if the truth of the hypothesis were established experimentally it would still be difficult to explain the origin of such radiation on a theoretical basis. It is known that, following β - emission from ^3H , the resulting readjustment of the system of atomic electrons to the changed nuclear charge of the ^3He ion will result in a de - excitation. The resulting quanta, however, will have a total energy of only 60eV, and therefore even assuming that all of the available readjustment energy goes to the β -particle, or in coincidence with it, the gap in the spectrum would be very much smaller than that observed.

At present, therefore, the anomalous shape of the spectrum of tritium remains unexplained.

CHAPTER 4.THE DECAY OF RaD.1. INTRODUCTION.

Since the discovery of radioactivity there has probably been no more widely studied source than RaD. Until 1939 the mode of decay was thought to be well understood. It was believed that decay was by a single β -transition, of maximum energy approximately 15 to 30keV, to an excited state of RaE of energy 46.7keV. De - excitation was thought to occur by means of a highly internally converted γ -ray of this energy. The presence of the internal conversion electrons had been well established through analysis by magnetic spectrometers (Danysz, 1911; Ellis, 1922; Meitner, 1922; Black, 1925).

The somewhat complex situation existing a decade later has been well summarised by Feather (1949). It has been shown by Lee and Libby (1939), Kinsey (1948), and Cranberg (1950), that the sum of conversion electrons

and unconverted γ - rays accounts for only 75 per cent of the disintegrations. In addition, extensive examination of the γ - rays, by means of the cloud chamber (Tsien, 1946), curved crystal spectrometer (Frilley et al, 1951), proportional counter (Curran, Angus, and Cockroft, 1949(b)), and scintillation counter (Bannerman and Curran, 1952; and Bannerman, Lewis, and Curran, 1951) has led to an increase of the number of low intensity γ - rays to a total of seven (energies 7.3, 16.1, 23.2, 31.3, 37, 42.6, 46.7keV). Further complexity is caused by the fact that these γ - rays do not readily suggest a unique system of levels.

In addition, isolation of the primary β - spectrum has not been achieved. Although most observers agree that the β - radiations are soft, (Richardson and Leigh-Smith, 1937; "Nuclear Data", 1950), even the limiting energy has not been well defined, estimates from ~ 15 to ~ 40 keV.

A systematic examination of the decay of RaD thus seems to be desirable. The particular applicability of the proportional counter to low energy β - radiations and highly converted low - energy γ - rays, as emitted in the decay of RaD, has been pointed out in Chapter 2. This

has been done by Curran, Angus, and Cockroft (1949,b) although detailed examination of RaD was not then the object of their work. The main object of the author's work contained in this chapter is examination of the β - spectrum.

2. APPARATUS.

The proportional counter, 1.4 ins in diameter and 8 ins in fully operating length (Cockroft and Curran, 1951), was enclosed in a brass cylindrical vessel C (insert Fig 5) into which the source could be slipped. (The apparent unnecessary complexity of the additional outside cylinder is due to the fact that the counter was designed with a view to its possible use in later coincidence work). The vessel was filled with a mixture of ethylene (80 per cent) plus argon (20 per cent) to a total pressure of one atmosphere. This filling was chosen so that the efficiency of detection of the L X-rays of RaE from the source was < 5 per cent. The counter was thus rendered responsive chiefly to conversion electrons and β - rays, but it was sensitive to the soft M X-radiations.

Pulses from the counter, after suitable amplification, caused deflection of the spot of a cathode

ray tube in a horizontal direction. The pulses were recorded on 35mm film by moving the film vertically at uniform speed. The camera lens used had an aperture $f/1.8$ and, as a rule, a film speed of 2 ins per sec was used for counting rates of $\sim 10,000$ per min. In this way a continuous range of pulse amplitudes was recorded permanently in a single run of very short duration, thus reducing errors due to any instability of the apparatus. Analysis of the film was made by projecting it on a screen, subdivided into a suitable number of equal amplitude intervals. Pulses falling in each interval were recorded and the results shown in a histogram.

Calibration was achieved by increasing the amplifier gain by a factor 4 and firing the fluorescence X-rays of copper, K_{α} , of energy 8.04keV, through an aluminium side window in the counter, the aluminium lining of the counter being perforated in this position.

3. SOURCE.

In early work, difficulty was encountered due to large disturbing pulses, arising from the presence of RaE and RaF which were difficult to eliminate in the preparation of the source. Considerable improvement was

achieved by using the method of separation described by Cranberg (1950). In this way, sources of about 97 per cent purity were obtained. The thinnest possible source was obtained by the method of Langer, Motz, and Price (1950), namely, the use of a 5 per cent solution of insulin in water to enable wetting of a relatively large area of the source backing. A small drop of RaD, in the form of chloride solution, was dropped on to this, and the source evaporated to dryness, the sample being rocked to prevent the formation of large crystals. In this way a thin source ($\sim 20 \mu\text{gm}$ of chloride), covering an area of approximately 1 cm^2 , was obtained. Counting rates were generally $\sim 20,000$ per minute.

4. RESULTS.

The pulse distribution was examined in two overlapping parts, from ~ 4 to $\sim 40\text{keV}$ (Fig 5), and from 33keV to beyond 80keV (Fig 6). The end point of the distribution was found to occur at an energy of $62 \pm 2.5\text{keV}$, this value being confirmed by a close examination of spectrum shape between $\sim 50\text{keV}$ and 70keV using improved statistical data. Despite its remarkable shape, the pulse distribution was confirmed with different arrangements of counter and source, the source being deposited either on nylon film ($\sim 50 \mu\text{gm}/\text{cm}^2$) or on

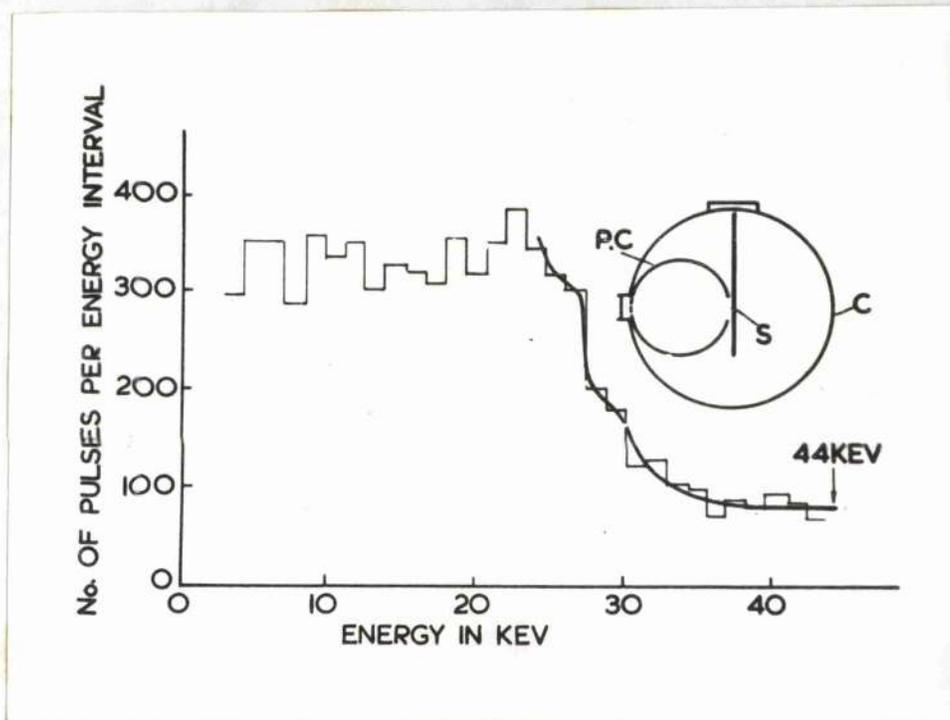


Fig 5. Pulse spectrum shape between ~ 4 and 40keV using a source mounted on aluminium, $1.6\text{mg}/\text{cm}^2$. The counter is shown in the inset. The source S is at an aperture in the proportional counter PC.

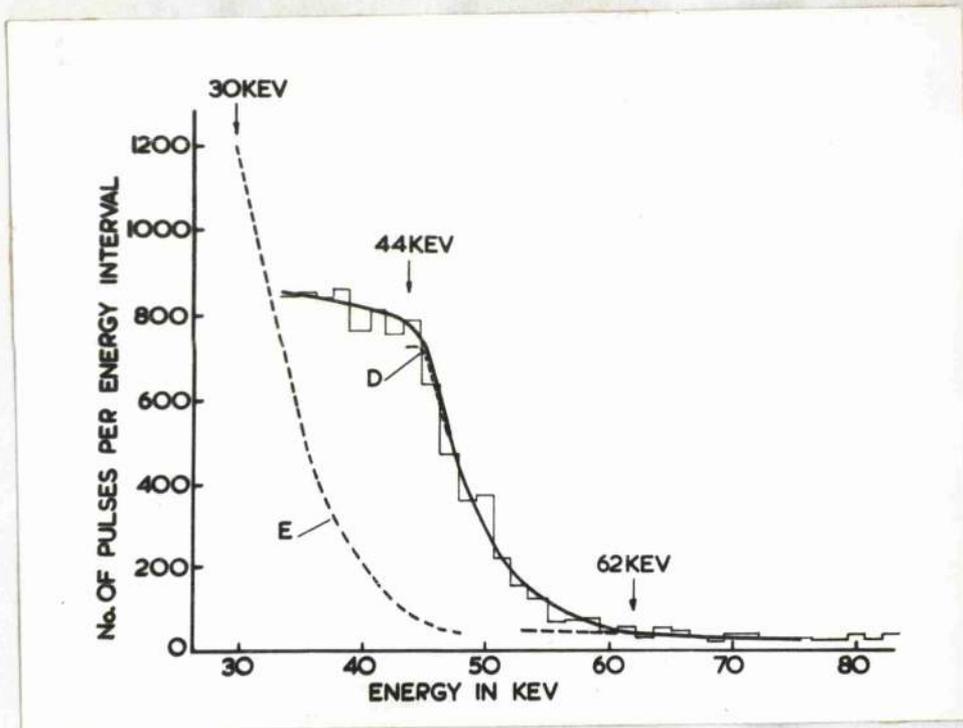


Fig 6. Pulse spectrum shape between
~ 33keV and 80keV.

aluminium ($\sim 1.6\text{mg/cm}^2$).

Between the energies of ~ 4 and 23keV the distribution is complex and is probably explained by the large number of homogeneous electrons of various energies produced by the L- and M- conversion of the soft γ - rays of RaE. (The available energy is insufficient for K - conversion to occur). Apart from the fact that these are probably often emitted in cascade, they can occur associated with β - particles and thus add to the complexity. The presence of the associated β - particles is, no doubt, largely responsible for the almost complete failure to resolve the many groups. Further analysis of this part of the curve was considered to be impracticable by the present technique.

Four combinations of β - particle and photoelectron, e^- , are possible in the arrangement used:-

- (a) β and e^- both away from counter - neither detected
- (b) β into counter, e^- away from counter - β alone detected
- (c) e^- into counter, β away from counter - e^- alone detected
- (d) e^- and β both into counter - both detected as a single ionising event by virtue of the integrating

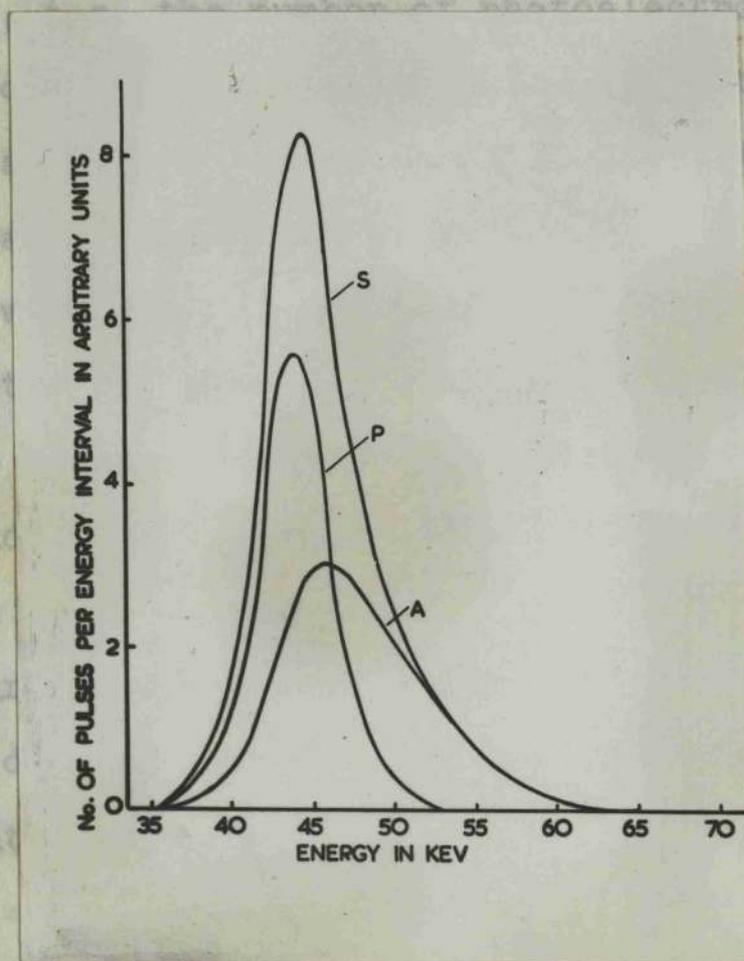
property of the proportional counter.

Case (b) gives rise to a β -spectrum which merges into the complex distribution at low energies. Case (c) gives the tendency to peak at energies of 30 and 44keV. Case (d) would be responsible for the "tails" at energies above these peaks. If the β -particles were of negligible energy, then no pulse of energy $> 44\text{keV}$ should be observed. Therefore the pulse distribution beyond 44keV is due to process (d), the conversion electron being of energy 44keV.

The end-point of 62keV therefore corresponds to an end-point for the actual β -spectrum of $(62 - 44) = 18\text{keV}$.

5. ANALYSIS OF THE β -SPECTRUM.

The following considerations enabled a derivation of the actual distribution of energy of this β -spectrum from the observed shape of the curve between 44 and 62keV. A theoretical Fermi distribution, F , for an allowed transition, a Z value of 83, and end point E_0 of 18keV, was evaluated. The curves of Fig 7 show the method of using the calculated distribution F . The curve, P , centred at 44keV, is based on experimental observations with the 46.7keV γ -rays of RaE, and represents the form that a homogeneous group of electrons takes when



Before such final comparison, the experimental curve has to be modified slightly to take into account the fact that, associated with the 30keV photoelectron peak, there is also a β -spectrum which thus extends to 48keV. Since the heights of the 30keV and 44keV peaks are as 3 : 2, the curve B (Fig 6) is drawn to represent the tail due to this spectrum, the height of this curve being $\frac{2}{3}$ times the height of the tail above 44keV. Thus, below 48keV, the area below curve B was subtracted from the main distribution to give the curve D which was photoelectrons + β -rays, and S the composite curve,

analysed by the counter. It is drawn so that the areas under curve P and under calculated curve F (not shown) are equal, i.e. the number of photoelectrons and β - particles of the continuum are the same. From the curves F and P, on taking their shapes into account, curve A was drawn. It represents the distribution obtained when any β - particle of group F, and any photoelectron of group P, are detected simultaneously. Since the probabilities of a photoelectron being detected alone, and being detected with a β - particle, are equal, the areas under curves A and P are equal. The curves P and A are then combined to give the curve S, which is then the derived curve to be compared with the experimental results.

Before such final comparison, the experimental curve has to be modified slightly to take into account the fact that, associated with the 30keV photoelectron peak, there is also a β - spectrum which thus extends to 48keV. Since the heights of the 30keV and 44keV peaks are as 3 : 2, the curve E (Fig 6) is drawn to represent the tail due to this spectrum, the height of this curve being $\frac{3}{2}$ times the height of the tail above 44keV. Thus, below 48keV, the area below curve E was subtracted from the main distribution to give the curve D which was

then compared with the derived curve S.

The final check between theory and experiment is shown in Fig 8. The very satisfactory agreement obtained appears to confirm that the region of the observed spectrum above 44keV corresponds to the combination of β - rays of limiting energy 18keV with photoelectrons. The peak at 44keV is not very accurately defined because sometimes M X-rays are associated, but, allowing for this, the end point of the β - spectrum is given, with fairly conservative limits by,

$$E_0 = 18 \pm 2.5 \text{ keV.}$$

6. DISCUSSION OF RESULTS.

It appears that the total disintegration energy of RaD is 64.5 ± 2.5 keV and that the main mode of decay consists of the emission of β - rays of maximum energy $E_0 = 18$ keV, leaving the RaE nucleus excited to 46.7keV.

The values of $E_0 = 27$ keV and half life $\tau = 22$ yrs assumed by Feingold (1951), give a value of $\log f\tau = 6.02$, thus classifying the β - transition as first forbidden. However the newly found value of $E_0 = 18$ keV gives $\log f\tau = 5.5$, (using values of $\log f$ given by Feenberg and Trigg, 1950), and definitely classifies the transition as allowed unfavoured.

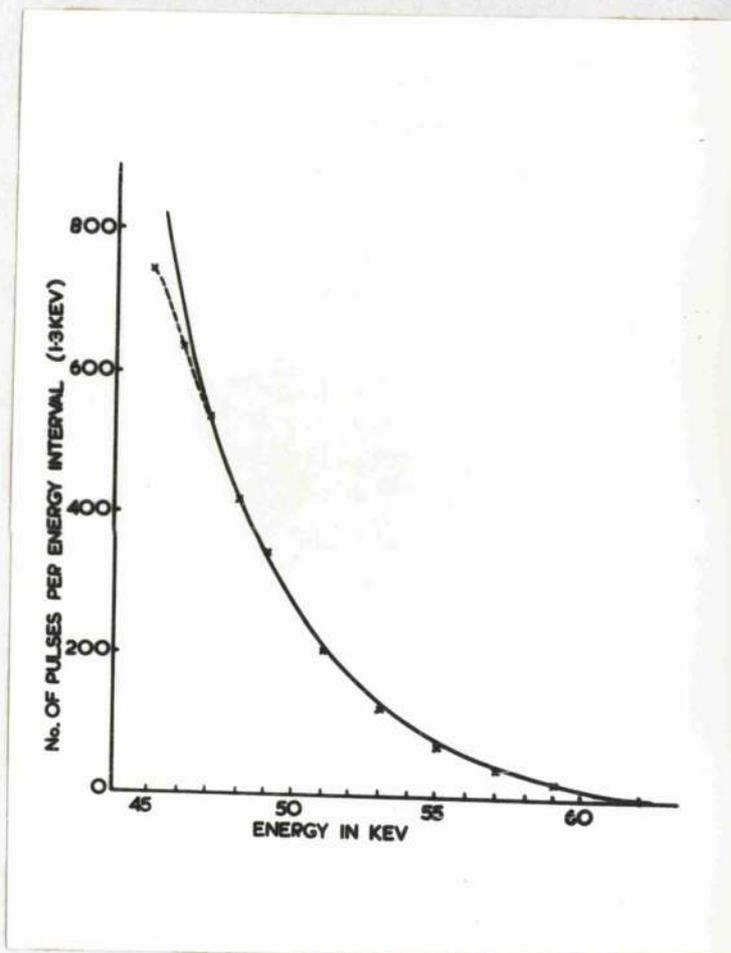


Fig 8. Comparison of the derived curve S with the experimental results. The experimental points are shown by crosses.

Ivanenko and Lebedev (1950), in order to account qualitatively for the results obtained with MsTh1 and MsTh2, have proposed a new type of β -decay which, for low energy spectra of heavy atoms, would reduce considerably the escape probability of the β -particles. In view of the rather close agreement obtained with Fermi theory applied to the decay of RaD, there seems to be no justification for these radical modifications, at least in this particular case.

It was first pointed out by Schwartz and Edwards (1951) that the experimental study of the β -spectrum of RaD should yield conclusive evidence on the effect of the readjustment of the atomic electrons to the changed nuclear charge of RaE. If E_z, E_{z+1} are the binding energies of the RaD and RaE atoms, then the readjustment energy, $D = E_{z+1} - E_z$, has a value of 15keV (Schwartz, 1952). Hebb (1938) suggested that, to a first order, all of this energy is carried away by the β -particles. Practically, this would give a displacement of the spectrum of RaD by 15keV, i.e. starting at 15keV and finishing at (15 + 18)keV. On the other hand, Schwartz (1952) concluded, from theoretical considerations, that the displacement energy is shared between the neutrino and β -particle, and

showed that, to a first order, this process would give no change in spectrum shape. The success of the analysis of section 5 leaves no doubt that the results obtained here definitely favour the theory that the readjustment energy is shared between neutrino and β -particle. Schwartz had already reached this conclusion with reference to the cloud chamber examination of the RaD decay by Richardson and Leigh-Smith (1937). Since their results depended on the examination of only 107 disintegrations and relied upon accurate measurement of track lengths of 1mm or less, the present results afford a more rigorous confirmation of Schwartz's theory.

7. RECENT CONFIRMATION.

Since the completion of the above isolation of the primary β -spectrum of RaD, the author's results have been confirmed by other workers. Bannerman and Curran (1952), have determined an end - point energy of $18 \pm 2\text{keV}$, while Kobayashi (1953) has estimated a value of between 16 and 18keV. In neither case, however, did the results permit an accurate examination of the spectrum shape.

By the same type of analysis as used by the author,

Jaffe and Cohen (1953), using a proportional counter with a gaseous source of RaD, obtained an end point of 15.2 ± 1 keV and found good agreement with Fermi theory down to 3 keV.

8. FURTHER STUDY OF DECAY SCHEME.

Although the primary β - spectrum has been successfully isolated there is still considerable doubt regarding the rest of the decay scheme. In view of the complexity of the pulse distribution observed (Fig 5), further investigations, of the type performed, promise little towards further elucidation of the decay scheme.

With regard to the β - spectrum, it has still not been established whether this is simple or complex. The analysis of section 5 assumes the absence of a transition between the ground states of RaD and RaE. (i.e. a spectrum of maximum energy 64.5 keV). In view of the success of the analysis, it appears that such a transition, if not entirely negligible, would be of very low intensity. The complex distribution of Fig 5 indicates that it is impracticable to make an accurate assessment of a ground to ground state β -transition.

There remains the possibility of β - transitions to

energy levels of RaE other than that at 46.5keV. With reference to the possible decay scheme shown in Fig 9, a β -transition to an intermediate level, and in coincidence with a γ -ray transition of 7.3keV, is of particular interest for examination by proportional counters, in view of their high sensitivity of detection of such low energy γ -rays. Clearly, investigation of this possibility is impracticable by the method used in isolating the primary β -spectrum. However, it should be possible, by measurements of the coincidences obtained between 7.3keV γ -rays in one proportional counter, and the β -particles, occurring in coincidence with them, in another proportional counter, to establish the existence or non-existence of this mode of decay.

Further, by examining coincidences between two γ -rays (γ - γ), between their conversion electrons (e^- - e^-), or between γ -rays and conversion electrons (γ - e^-), it should be possible to study in greater detail the various competitive modes of de - excitation of the RaE nucleus.

9. PROBLEMS RELATED TO FUTURE WORK.

At this stage two further problems require to be

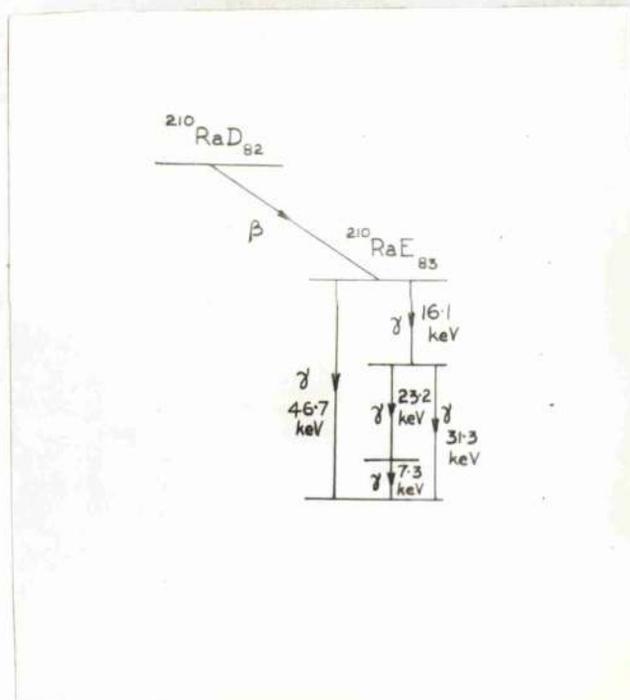


Fig 9. Possible decay scheme of RaD.

examined:-

- (a) It is necessary to develop a technique for recording and measuring the energies of the ionising radiations in coincidence.
- (b) In order to eliminate uncertainties arising from scattering of particles from the source support, or from self absorption of the particles in the source itself, it would be desirable to use the RaD source in gaseous form, after the manner employed with ^3H (Chapter 3). A gaseous source in the form of RaD tetramethyl would be suitable for this purpose. However, at this time a really satisfactory sample of RaD tetramethyl had not been produced, most of them suffering to some extent from contamination by Polonium (RaF). Since it decays by emission of α -particles, the presence of polonium in the source is objectionable, in view of the large disturbing pulses observed with the proportional counter.

It is therefore necessary to use a thin solid source for the ensuing experiments. This procedure, however, introduces uncertainties regarding the effect of scattering of the particles from the source itself. The somewhat alarming and confusing state of knowledge existing at this time, regarding the adverse effects of scattering,

made it desirable to seek fresh data by experiments with the proportional counter and with the type of geometrical arrangement to be used in the coincidence work.

CHAPTER 5.

DEVELOPMENT OF COINCIDENCE TECHNIQUE.

The method eventually used to measure the two separate energies of two coincident ionising events was developed from a single - channel pulse - analyser, constructed by the author during the early part of his research programme. The pulse analyser, now described, was originally intended to be a general purpose instrument.

1. SINGLE CHANNEL PULSE ANALYSER.

The working of the analyser, which consists of a pulse lengthening circuit and a modulation circuit, with an electronic gate between them (Fig 10), is as follows.

A positive pulse from the counter, after amplification, is applied to the grid of V_7 and causes the "flip - flop" arrangement comprised of V_7 , V_8 , and V_9 to apply a sharp negative cut - off voltage to the grid of V_3 . The same pulse is fed in at A, and V_3 is cut off

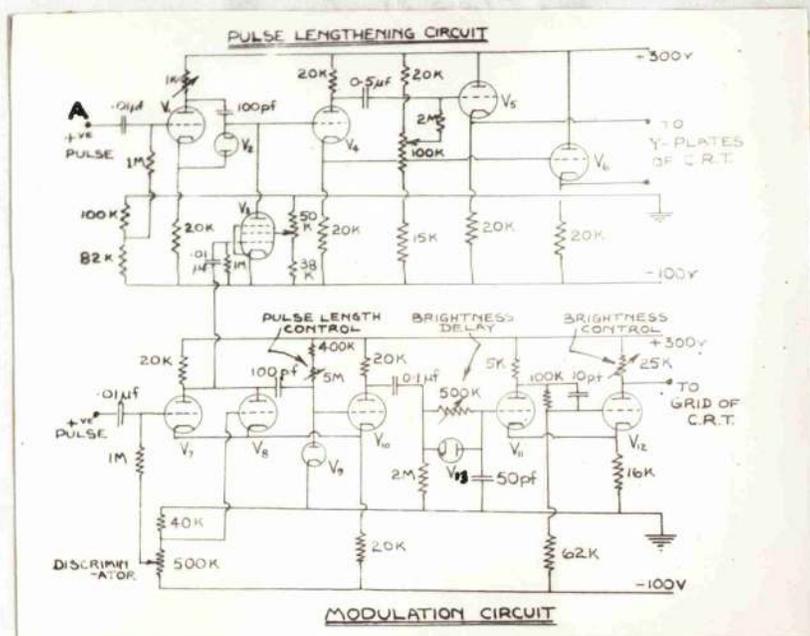


Fig 10. Circuit for the single - channel pulse - analyser.

before the maximum height of this pulse is reached. Consequently, a positive voltage, proportional to the maximum height of the pulse entering at A, is allowed to build up on the grid of V_4 , and be maintained at this steady maximum value for a time depending on the length of the cut - off pulse applied to the grid of V_3 . This time can be controlled by means of the pulse length control shown in the modulation circuit. Thus a positive and a negative pulse are obtained from the anode and cathode, respectively, of V_4 and fed in "push - pull" on to the Y plates of a cathode ray tube, causing the spot to be deflected by a distance proportional to the peak voltage of the pulse from the counter, i.e. proportional to the energy of the ionising event detected by the counter. The spot is held in this deflected position for a time determined by the pulse length control (usually $\sim 15 - 20 \mu\text{sec}$).

During the time the spot is held in the deflected position, a positive pulse is applied from the modulation unit to the grid of the cathode ray tube, thus causing the spot to be brightened. This pulse is such that its leading edge is delayed, and back edge advanced, with respect to the squared pulse applied to the Y plates.

This ensures that brightening of the spot will occur only when it is held in the position of maximum deflection. The delay is adjusted by means of the delay control (Fig 10), and advancing of the back edge is effected by means of V_{13} . The brilliance of the brightened spot can be varied by means of the brilliance control shown.

By reducing the brilliance control of the C.R.T. the "stems" of the pulses are made to disappear entirely, leaving a resultant distribution of bright spots along a vertical line.

To examine the pulse distribution, a light - tight box was built to contain a photomultiplier (type 931A), together with its base connections. A narrow slit in this box was fixed horizontally against the face of the C.R.T., so that, whenever a brightened spot appeared in front of the slit, it was detected by the photomultiplier, the output of which was found to be sufficient to operate a scaler connected directly to it.

The whole distribution of spots is then moved across the slit in steps, by varying the voltage applied to the Y - plates. This voltage, measured with an accurate voltmeter, gives a direct reading of the height

of the pulses being counted at a particular setting. To ensure accuracy of this setting, both a coarse and a fine Y shift control were fitted to the C.R.T. (Fig 11). The 300 volts line from the analyser circuit was used, via a dropping resistor, to give a voltage of about 100 across the double $50K\Omega$ potentiometer. The coarse adjustment is made by R_2 and R_3 while R_1 provides the fine adjustment. Defocussing of the electron beam was not caused by these adjustments but, in order to avoid such an effect, the X plates, (not used in this particular arrangement) were both connected to the astigmatism control.

The channel width of the analyser is determined by the width of the slit, which in turn must be greater than the diameter of the spots on the screen. It was found that a slit corresponding to about 1 volt of input pulse size (width of slit $\sim 1\text{mm}$) was satisfactory. Since the circuit deals satisfactorily with input pulses up to about 50 volts, pulse distributions can be examined in about 50 equal steps.

2. PERFORMANCE OF PULSE ANALYSER.

By means of a pulse generator the analyser was tested for proportionality of pulse input and output.

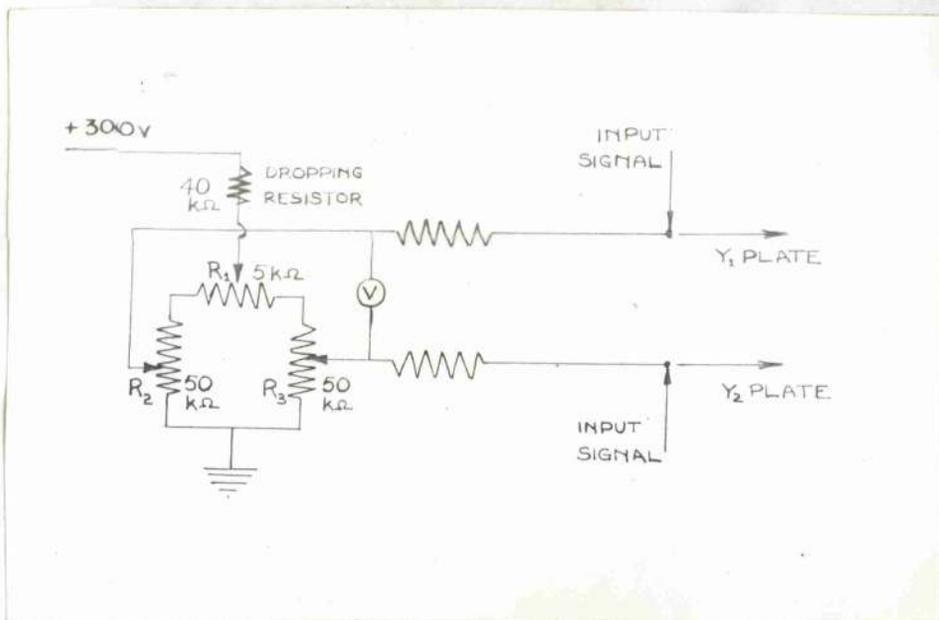


Fig 11. Connections to cathode ray tube used with single - channel pulse - analyser.

The results are shown in Fig 12. Using the push - pull output, the output voltage was found to be approximately double the input voltage. If the pulses are not lengthened, a linear relationship is obtained for input voltages from 0 to 60 volts. When they are lengthened, however, as is the usual practice, the relationship becomes non linear for low input voltages (< 15 volts). This non - linearity is due to the sudden cut - off of V_3 (Fig 10) by the negative signal from the modulation unit. This causes a slight positive increase in the grid voltage of V_4 , provided that it has not already been driven positive by a large signal pulse. Consequently, for inputs > 15 volts, a linear relationship is obtained, while for smaller inputs the output becomes progressively greater than expected, until an output pulse of 7 volts is obtained with virtually zero signal input.

A further test with the pulse analyser was made using a source of ^{125m}Te inside a proportional counter. (This source is discussed in Chapter 7). The pulse distribution (Fig 13) shows the peaks due to conversion electrons, of energies 105keV and 78keV, and compares quite favourably with the distribution obtained by the moving film technique of recording. For straightforward

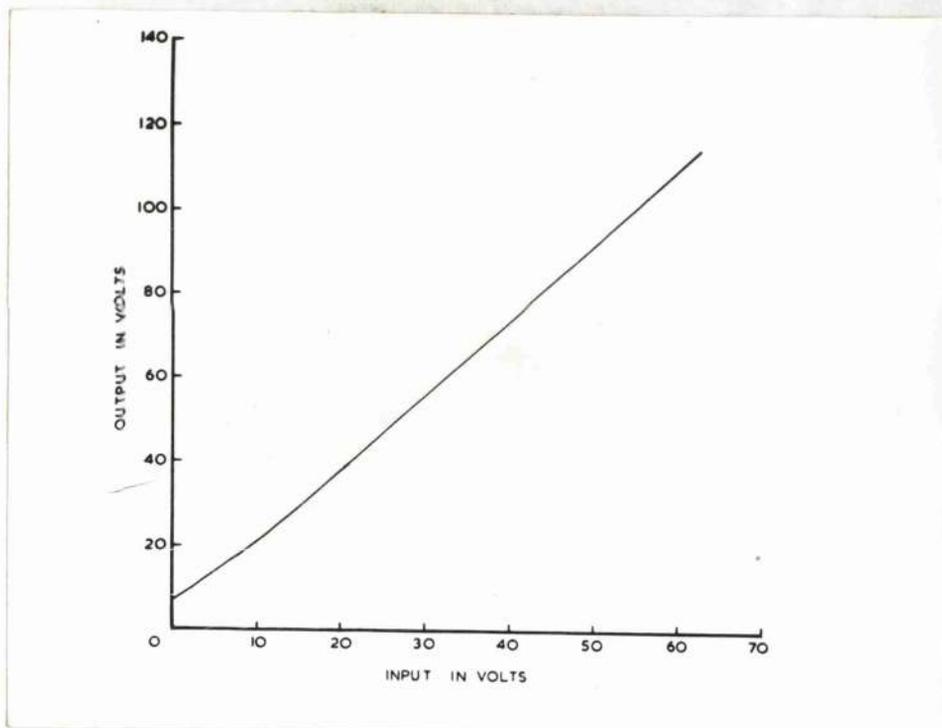


Fig 12. Response curve of pulse analyser.

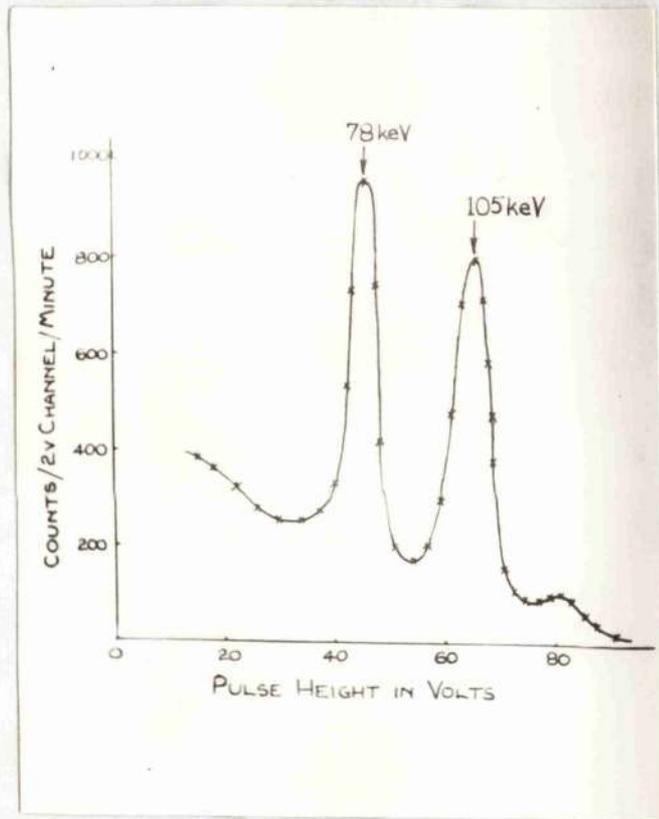


Fig 13. Electron spectrum of ^{125m}Te examined by proportional counter and single - channel pulse - analyser.

examination of pulse distributions the latter method was retained, however, since it permits a complete and permanent record of observations to be taken in a few minutes.

3. PULSE SHAPE OBTAINED WITH PROPORTIONAL COUNTER.

The pulse produced in a proportional counter by electron collection is relatively small. The major part of the pulse is produced by the motion of the positive ions away from the counter wire. Because of this fact, the duration of pulse formation is $\sim 10^3$ times greater than the time of electron collection. This rise - time of the pulse could be reduced considerably if the counter dimensions and the gas filling could be chosen freely. These factors are determined, however, by the nature and energy of the radiations being examined. In practice therefore a compromise has to be made, between counter design and electronic circuitry, to reduce to a minimum the deleterious effect of the rise time of pulses in the coincidence arrangements.

In effect it is necessary to employ some type of saturated amplifier between the main amplifier and the coincidence unit in order that the latter operates as

early as possible on the leading edge of the pulse. Clearly, since proportionality of pulse height with energy of radiation is no longer maintained, it is not possible to use two discriminations to select, say, a particular γ - ray energy. The following method was therefore adopted:-

5. X - Y PLOTTING METHOD.

A second pulse lengthening circuit was constructed, identical with the one described in section 1. and incorporated in the earlier pulse analyser as shown in Fig 14.

By this method the pulses from both counters are plotted. If counters 1 and 2 (Fig 14) detect coincident events the spot of the C.R.T. is deflected simultaneously in the X and Y directions, by amounts proportional to the energies of the ionising events detected in counters 1 and 2 respectively. The spot is held in the deflected position for $\sim 15 - 20 \mu\text{sec}$ and, since a coincidence is detected, during this time the spot is brightened. Hence all bright spots appearing on the screen denote a coincidence (whether true or accidental) and the X- and Y- coordinates of the spot give separate measurements of the energies of the two coincident radiations.

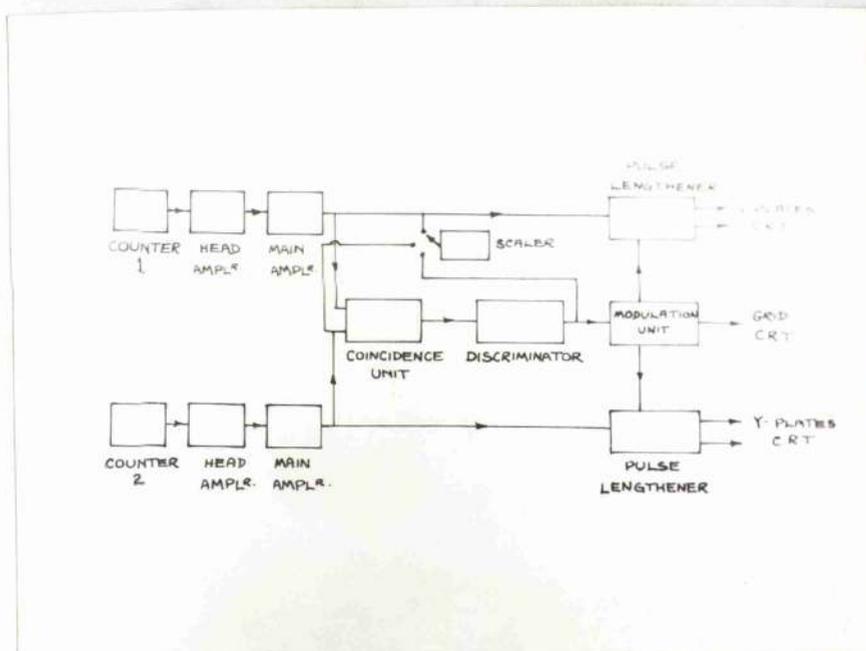


Fig 14. Block diagram of circuits used in recording coincidences.

Recording of the distribution of spots on the screen is effected photographically, employing a suitable time of exposure to ensure that not too many spots are recorded on the same frame of film.

In order to reduce the delay possibly caused by the slow leading - edges of pulses from a proportional counter, the coincidence unit is caused to operate as early as possible on the leading edge. For this reason the special coincidence unit, due to Mr. R. Giles, and shown in Fig 15, was used.

Each of the two input channels consists of a Schmitt trigger circuit, a pulse - shaping circuit, and an A.C.- coupled "flip - flop" circuit. The output from each channel is a rectangular pulse whose length is adjustable by potentiometer R_2 . The length of this pulse determines the resolving time of the circuit. Coincident pulses give rise to an output pulse of amplitude double that due to a non - coincident pulse. The discriminator, following the coincidence unit, is therefore set to allow a pulse to pass through to operate the modulation unit only when a coincidence occurs.

The use of the X - Y plotting method in further

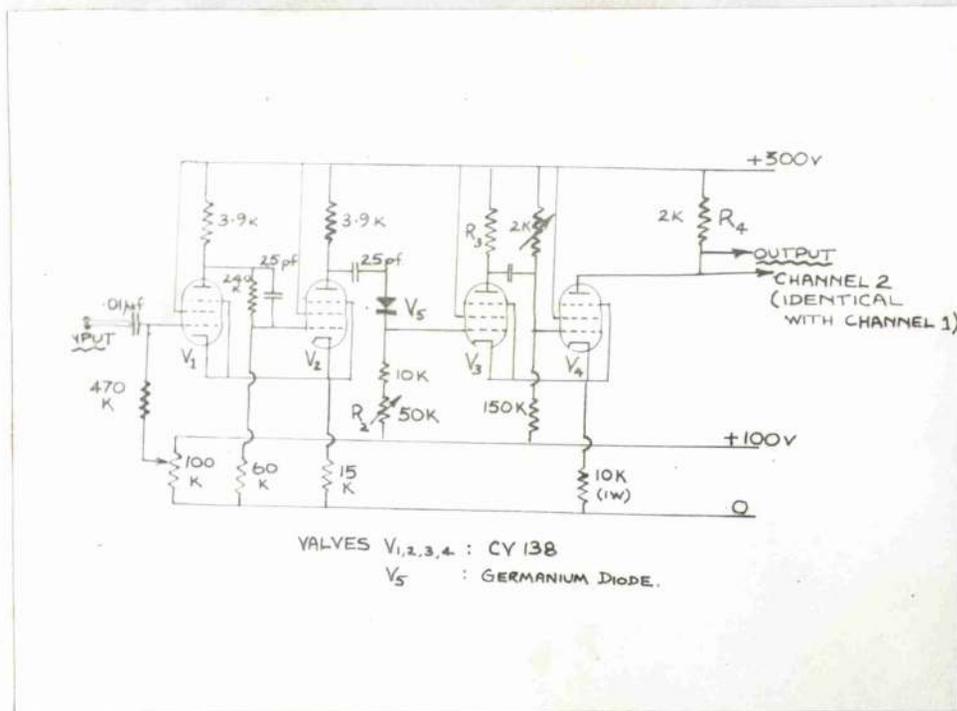


Fig 15. Circuit diagram of one channel of coincidence unit. R_4 is common to both channels.

examination of the decay of RaD is described in the next chapter.

C H A P T E R 6.APPLICATION OF COINCIDENCE TECHNIQUES
TO THE FURTHER
EXAMINATION OF THE RaD DECAY SCHEME.1. APPARATUS.

Two brass proportional counters C_1 and C_2 (Fig 16), each of diameter 1.4 ins and fully operating length 8 ins, were constructed to fit inside cylinder C , to which the cathode potential, common to both C_1 and C_2 , was applied. The source holder fits between the two counters in such a way that the source, evaporated on nylon film ($\sim 50 \mu\text{gm}/\text{cm}^2$), is located centrally at an aperture in each counter wall. The apparatus thus constitutes two spectrometers, each with an acceptance solid angle of 2π . The counters were filled with a mixture of methane (12 per cent) plus argon (88 per cent) to a common pressure of 76cm mercury. The counters were connected to the circuit as shown in Fig 14 and recording effected by means of the x - y plotting method.

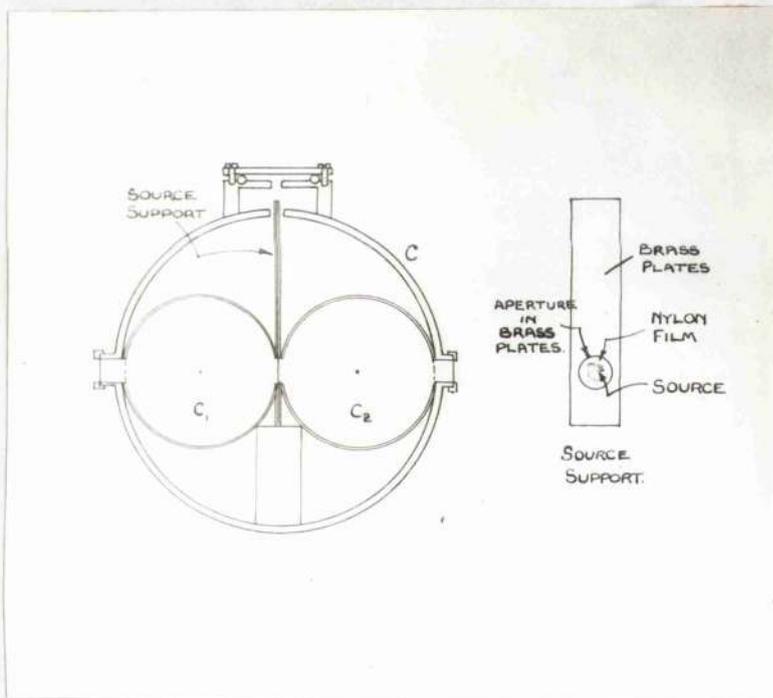


Fig 16. Double proportional counter used for coincidence study of RaD decay scheme.

2. PROCEDURE.

In parallel with the development of a suitable coincidence technique, the author was also investigating the effects caused by back - scattering of electrons from a source support (described in chapter 7). It became evident early in these experiments that possible errors introduced by the phenomenon of back - scattering could be minimised by using a source backing of low Z value. For this reason sufficient polythene (.004 in) was placed on the non - source side of the nylon film to absorb all the β - particles and conversion electrons of RaD, thus allowing only γ - rays and X- rays to pass into counter 2. In view of the small counter dimensions and relatively low pressure of gas filling (chosen to give as fast a leading edge of pulse as possible), counter 2 was efficient only for the detection of the softer γ - rays (i.e. 7.3keV). The other counter 1 detected only β - particles and electrons.

The β - counter caused deflection of the spot of the C.R.T. in the x - direction, and the γ - counter in the y - direction. Thus a β - spectrum in coincidence with a low energy γ - ray should appear on the screen as a distribution of spots along a line parallel to the x - axis, and at a distance from it proportional to the

energy of the coincident γ -ray.

The distribution of the spots was recorded photographically on 35mm film. To avoid data being lost through spots being superimposed, only 100 coincidences, counted by a scaler, were recorded on each frame of film. The x - and y - axes were marked as a distribution of bright spots on each frame. This was effected by allowing pulses from one counter at a time to enter the coincidence unit and reducing the discriminator bias to allow all pulses to pass through to the modulation unit (Fig 14). The same procedure was applied during calibration, which was made by the aid of the fluorescence K_{α} x - rays of copper (8.04keV) fired through the side window of each counter in turn. Too dense a distribution of bright spots around the position on the axis corresponding to 8.04keV was obtained in this way. Thus, to achieve greater accuracy of calibration, a 50 cycle time base was applied perpendicular to the particular axis.

Examination of the x - y plots was made by projecting each frame of film in turn on to a large sheet of paper and marking the position of each spot. Analysis of the resultant total distribution was then made by

dividing the pattern into equal component rectangles and counting the number of spots in each. Histograms were then drawn of the relevant parts.

A series of experiments was made to find the best value of resolving time (τ) for the coincidence unit, in order to compromise between too small a value for τ , which leads to loss of true coincidence counts, and too large a value of τ which gives a chance coincidence rate comparable with the true coincidence rate. To measure the chance coincidence counting rate a 4μ sec delay line was inserted in front of one channel of the coincidence unit. In this way a value of $\tau = 1.6\mu$ sec was found to be most suitable. The counting rates in the β - and γ -counters were $\sim 12,000$ and $\sim 3,000$ counts per min respectively. Of the observed coincidence counting rate of ~ 8 per min, ~ 2.5 of these were due to chance. Allowance for this high percentage of false data was made by repeating the x - y plotting experiment, for the same length of time, with the delay line in position, and subtracting the distribution obtained from that found with no delay line. All of the results described below have already been corrected in this way.

Occasionally, instead of a bright spot, a streak is

70.

obtained on the photograph. This is due to a pulse following within $20\mu\text{sec}$ of two coincident pulses. Although it is not itself recorded as a coincidence, it arrives at the pulse - lengthening unit during the lengthening period and causes further displacement of the already brightened spot. In view of the rarity of such an event with the counting rates used, it was sufficient to discard these streaks in the analysis.

3. RESULTS.

Examination was made of coincidences between β -particles or electrons up to an energy of $\sim 70\text{keV}$ and γ -rays up to an energy of $\sim 30\text{keV}$. The only relevant results are shown in Fig 17. The distribution of Fig 17(a) was obtained by analysis of a strip representing coincidences between β -particles and electromagnetic radiations of energy between 6.5keV and 8.5keV i.e. including the 7.3keV γ -ray originally reported by Curran, Cockroft and Angus (1949). In the course of the author's experiments Jaffe and Cohen (1952) detected this γ -ray and believed it to be in coincidence with a β -spectrum. The author's results (Fig 17(a)), on the other hand, give no indication of any such β -spectrum in coincidence with a γ -ray of this energy. This is in spite of the fact that, if the

7.3keV γ -ray is present with the intensity reported, (Tsien, 1946), and is in coincidence with a β -spectrum, calculation shows that definite evidence of this should have been found.

The curve of Fig 17(b) shows the electron spectrum obtained in coincidence with the soft M X - rays of RaE (energy \sim 1 to 4keV) resulting from internal conversion of the 46.7keV γ -ray. The double peak, resolved into two peaks centred on 33 and 44keV, gives evidence of coincidences between the M X - rays and conversion electrons (L and M respectively) of these energies. The results do not afford a better determination of L : M conversion ratio than already determined (Cranberg, 1950). However the ease with which the observed results can be regarded as consistent with existing knowledge of the decay scheme gives confidence in the technique used.

Further assurance is provided by the curve of Fig 17(c) which shows the electron spectrum in coincidence with the L X - rays of RaE, of energies between 10.8 and 13.3keV (Feingold, 1951). The peak at 30keV indicates coincidences between these X - rays and the associated L - conversion electrons of this energy. Further, the part AB of the curve could be explained by coincidences

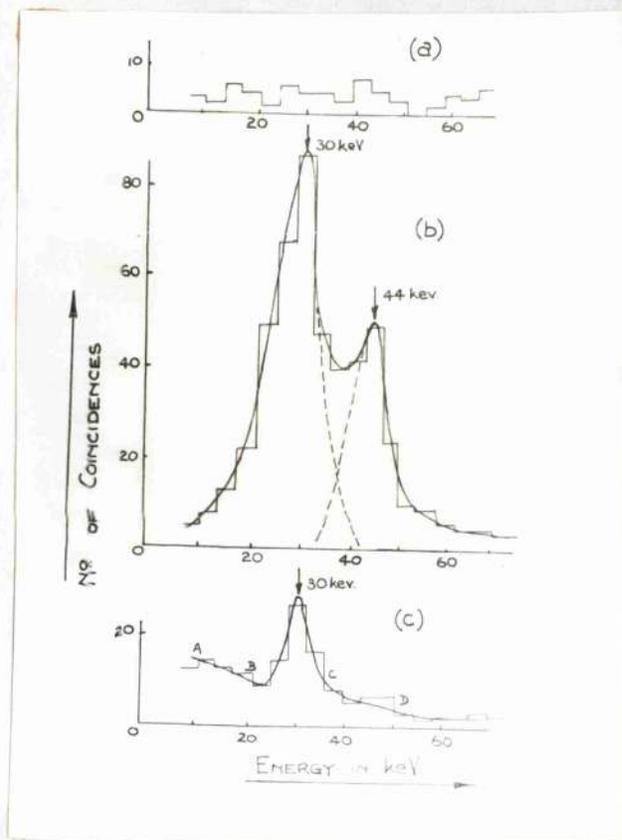


Fig 17. Spectrum of β - particles or electrons in coincidence with (a) γ - ray of energy 7.3keV
 (b) M X - rays (~ 1 to 4keV)
 (c) L X - rays (10.8 to 13.3keV)

with the primary β - spectrum. Likewise the higher energy tail CD would indicate integration of conversion electrons and the primary β - spectrum.

It is concluded therefore that, although the method used is capable of detecting a β - spectrum in coincidence with the 7.3keV γ -ray, the author's results give no evidence of such a transition.

In order to investigate further the γ - radiations, an examination of γ - γ coincidences was attempted. Polythene absorber was placed on each side of the source and the pressure of the counter filling (10 per cent methane, 90 per cent argon) increased to 150cm mercury, to improve the efficiency of detection. However, no significant distributions were obtained. Further experiments of this type were considered to be unable to compete with the γ - γ investigations being conducted at the same time by Bannerman and Curran (1952) using scintillation counters.

4. SUBSEQUENT CLARIFICATION.

It has been pointed out that the author's coincidence experiments were conducted, to some degree, in parallel with an investigation of back - scattering

of electrons. During the next few months, devoted almost entirely to the latter, several publications led to considerable simplification of the decay scheme of RaD (Ewan and Ross, 1952; Wu, private communication, later published, Wu, Boehm and Nagel, 1953). Re - examination had shown no evidence of a γ - ray of energy 7.3keV although Wu et al showed that a line of approximately this energy could be strongly excited by copper backing. The explanation of the author's results (section 3 above) is therefore obvious. Further, Wu et al showed that the γ - ray line at 23keV could be contributed from the piling effect of the detecting system. The upper limits put on the various reported γ - ray lines by Ewan and Ross are shown below.

TABLE 1.

| | | | | | | |
|-----------------------|------|------|------|------|------|------|
| γ - ray energy | 46.5 | 42.6 | 37 | 31 | 23.2 | 16.1 |
| Relative intensity | 1 | .015 | .016 | .015 | .025 | .06 |

The 16.1keV γ - ray is also doubted by Wu et al who conclude that it is probably the $L_{\gamma}X$ - ray line of energy 15.7keV. The decay scheme shown in Fig 18 is therefore postulated by Wu, Boehm and Nagel (1953).

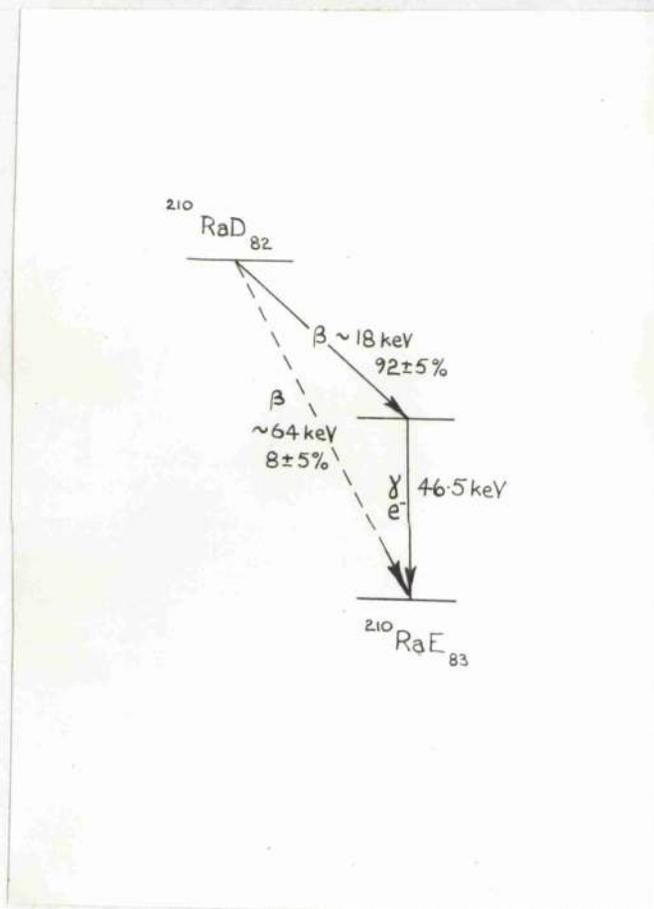


Fig 18. Decay scheme of RaD proposed by Wu, Boehm and Nagel (1953).

54.

The fairly recent views (Lee and Libby, 1939; Kinsey, 1948; Cranberg, 1950) that the sum of the conversion electrons and unconverted γ -rays of energy 46.7keV can account for only 75 per cent of the disintegrations is also re-checked by Wu whose value is 92 ± 5 per cent. Thus there still exists the possibility of a β -transition between ground states to account for the other 8 ± 5 per cent of the disintegrations.

5. FURTHER STUDY OF RaD DECAY SCHEME.

From the author's results, described in chapter 4, it was concluded ~~from~~ ^{K_αT} the complexity of the observed spectrum made it quite impracticable to attempt a separation of the β -transition between ground states. It appears therefore that the best contribution which can be made to the decay scheme of RaD, by means of proportional counters, is a re - investigation of the primary β -spectrum with a view to its more complete isolation without the rather complex analysis used in chapter 4. It is considered that this is possible by examination of coincidences between the β -particles and the unconverted 46.5keV γ -rays, which occur in about 3 to 4 per 100 disintegrations (Bramson, 1930; Gray, 1932; Droste, 1933; Stahel, 1935).

With this in view, the counter shown in Fig 19 was

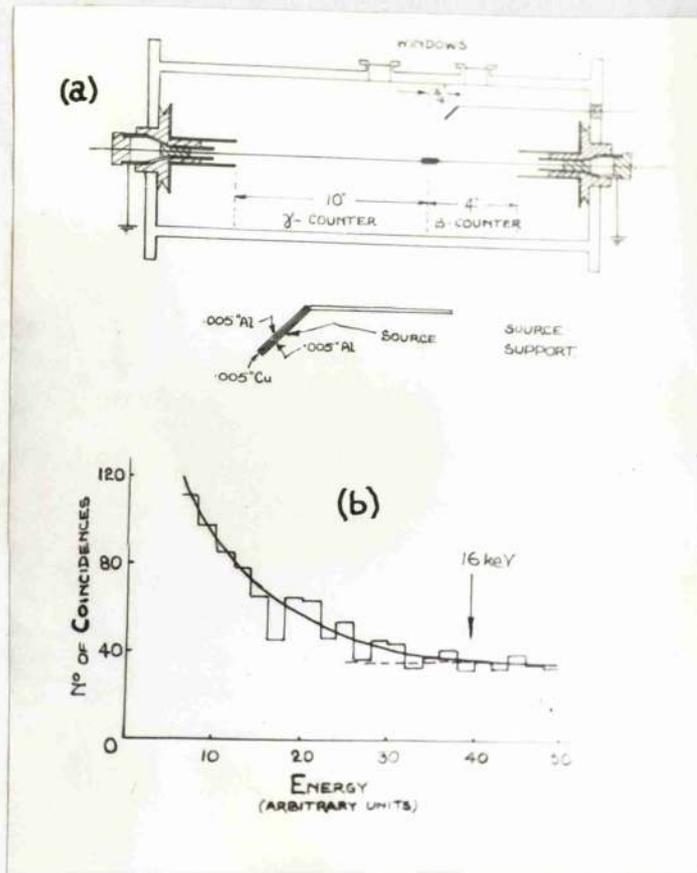


Fig 19. (a) Proportional counter used for isolation of primary β -spectrum of RaD.
 (b) Spectrum observed in coincidence with 46.5keV γ -ray.

constructed. The aluminium - lined brass counter, 6ins in diameter, was divided into two counters of fully operating lengths 4 and 10ins by means of a glass bead insulating the central wires from each other. The RaD source, in the form of chloride, was evaporated on to an aluminium (to minimise back - scattering effects) source holder .005in thick. Sufficient copper (.005in) was placed on the reverse side of the source holder to allow only the 46.5keV γ -rays to enter the γ -counter. To ensure that X - rays of copper excited in this absorber did not pass into the γ -counter, a further aluminium foil was placed behind the copper. The source was supported by a probe (Fig 19) held at a potential appropriate to its position in the electric field, the source holder being positioned towards the counter wire in order that any distortion of the field is in the nature of an increase rather than a decrease.

The position of the bead was chosen to give maximum stopping of the γ -rays in the γ -counter. The source was placed inside the β -counter to ensure that no β -particles can reach the γ -counter which therefore can detect only the γ -rays of energy 46.5keV. Efficiency of detection of the γ -rays was increased by using a gas filling at a pressure of 3 atmospheres

(10 per cent methane, 90 per cent argon). Under these conditions of the experiment the efficiency of detection was ~ 3 per cent.

The x - y plotting method was used to record the β - spectrum as a distribution of spots along a strip parallel to the axis and at a distance from it proportional to the γ - ray energy of 46.5keV.

6. RESULTS.

The counting rates in the β - and γ - counters were $\sim 4.10^4$ and 3.10^3 per minute respectively. The coincidence counting rate of ~ 20 per minute again required correction of the results (by the subtraction method) in view of the comparable chance coincidence counting rate of ~ 10 per minute.

The spectrum obtained is shown in Fig 19. There is little doubt that the β - spectrum has been isolated and the observed end point of about 16keV is in good agreement with the author's previously determined value. Further, the complete absence of a low energy cut - off at ~ 15 keV adds further support to the theory that the re - adjustment energy of the atomic electrons of the RaE atom is shared between electron and neutrino.

Unfortunately the statistics obtainable do not afford the more accurate comparison with Fermi theory that had been anticipated. The time taken for the actual recording of the results was six hours, and it is considered that little improvement can be made by this method. The main shortcoming of the experiment was the low efficiency of detection of the unconverted γ - rays.

7. SUGGESTION FOR FUTURE IMPROVEMENT.

It would appear that successful isolation of the β - spectrum, in the way envisaged, could be effected by combining the high efficiency of detection for γ - rays, provided by a scintillation counter, with the good resolution for the low energy β - particles, provided by the proportional counter. The RaD source could be placed on the inside of an aluminium window in the proportional counter, and the scintillation counter placed close to the outside of the window. Further, in view of the very sharp leading edge obtained with pulses from a photomultiplier, the difference in the bias levels of the two discriminators could be used to isolate the 46.5keV γ - ray from the rest of the electromagnetic radiations, thus simplifying considerably the electronic circuitry of the recording apparatus.

CHAPTER 7.THE EFFECT OF BACK - SCATTERING
OF ELECTRONS IN β - SPECTROSCOPY.INTRODUCTION.

In β - spectroscopy the use of solid sources, deposited on some form of source support, leads to some uncertainty regarding the effect of scattering of the β -particles or electrons, from the source - mounting, back into the counter. Apart from the effect on the counting rate, the phenomenon may cause distortion of the observed spectrum, and it is essential that the effect be taken into account.

Theoretically, the scattering of electrons has received considerable attention since the early theory of single scattering by Rutherford, Chadwick, and Ellis (1930). In the case of back - scattering, which involves large total angles of scattering (up to 180°), it is necessary to treat the problem on the basis of multiple scattering

of the particles. However the treatments of multiple scattering made by Williams (1939), Goudsmit and Sanderson (1940), Molière (1948), Snyder and Scott (1949), Butler (1950), and Lewis (1950), have all neglected back - scattering. More recently, Wang and Guth (1951) derived an expression to include back - scattering, but the functions are not practicable for numerical calculation. Furthermore when the total scattering angle is large (as in back - scattering), they are unable to convert their expression into any form suitable for numerical calculation. To date, therefore, no reliable data on the allowances to be made for back - scattering is available from theory and it is necessary to seek such information from experiment.

In the examination of sources of low activity, such as is involved in the study of radio - elements deposited on or near the wall of a proportional counter (as in the experiments of RaD), it is the back - scattering from the source support into a solid angle of 2π that is encountered. Experiments under conditions that are very roughly approximate to these have been made, but, with the exception of Brownell (1952), who used a proportional counter, these were made with Geiger counters.

91.

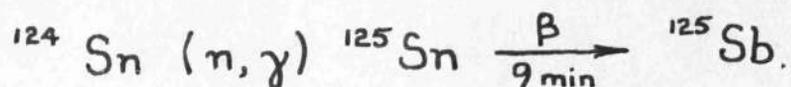
Consequently in the experiments of Burt (1950), Yaffe (1950), and Suzor and Charpak (1952), no close correlation with particle energy was possible. Indeed the sources used were almost invariably β -emitters and the variation of reflection with the average energy of the β -rays was measured. Agreement between the results of Burt (1950) and Yaffe (1950) is fairly close, but the more recent work of Suzor and Charpak (1952) shows marked disagreement. The latter workers indicate that the effect of back - scattering is much more serious than has been indicated by the earlier experiments. Furthermore they conclude that, contrary to previous belief, the effect of back - scattering increases with decreasing particle energy. The situation became even more unsatisfactory with the publication (in the course of the present experiments) of Christian et al (1952), which indicates rough agreement with the findings of Suzor and Charpak.

In view of this rather confusing state of knowledge it seemed useful to make fresh measurements with the aid of the proportional tube itself, and under conditions strictly comparable ^{with those obtaining in} the application of this instrument to β -ray spectrometry.

2. SOURCE.

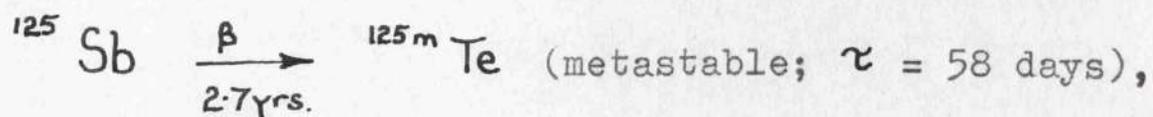
In order to obtain a better relationship between the effects of back - scattering and the energy of the primary electrons than had been possible by the earlier experiments with β - emitters, a source of ^{125m}Te was chosen. This source is known to have a spectrum consisting of two clear conversion electron peaks, of energies 105keV and 78keV, as well as a compound group of energy $\sim 27\text{keV}$ (Bowe and Axel, 1952). The source therefore provides simultaneously three roughly mono - energetic groups of electrons within the energy range 0 - 105keV. This range is of greatest interest in the present study of low energy disintegrations, and was the least accurately examined by Burt (1950) and Yaffe (1950). They have shown, in fact, that in this region the variation of back - scattering with energy is most rapid, whereas above this region a saturation value is reached (i.e. independent of further increase in electron energy).

The source was prepared by neutron bombardment of tin in the Harwell pile, the reaction being



After allowing four months for the irradiated sample to

age, the ^{125m}Te , formed by the decay,



was separated chemically by the method of Mandeville and Shapiro (1949). A very thin source ($\sim 10 \mu\text{gm}/\text{cm}^2$), almost carrier - free, was then obtained by thermal evaporation of the tellurium in vacuum. In most cases the source backing was a thin nylon film ($\sim 50 \mu\text{gm}/\text{cm}^2$).

An initial examination of the source was made in a proportional counter, 14cm in diameter and 30cm in fully - operating length, filled to a pressure of 2 atmospheres with methane (15cm mercury) plus argon. The source, deposited directly on to a spade - shaped aluminium holder 0.5mm thick, was supported inside, and half way along, the counter by a copper probe, parallel to the axis of the counter. The voltage applied to the probe was adjusted to a value appropriate to its position in the electric field in order to eliminate its disturbing effect on the field. The counting rate was $\sim 30,000$ per minute and the pulse distribution was analysed by displaying on a C.R.T. and recorded by the moving film technique.

The resulting spectrum shape obtained (Fig 20) is found to agree well with that expected from the decay scheme proposed by Bowe and Axel (1952) (inset Fig 20). According to the decay scheme, the 58 - day isomeric state decays to a stable ground state by means of two highly converted γ -rays in cascade, of energies 109.7keV and 35.5keV. Conversion of the former in the L- and M-shells gives peaks in the distribution at ~ 105 keV, while a peak at 78keV is obtained by K-conversion. Similarly mono - energetic groups of electrons at 31keV and 34.3keV are obtained by L and M conversion of the 35.5keV γ -ray. Evidence of all of these is shown in the observed spectrum (Fig 20). The peak at 27.4keV is due to the K X - rays. These are detected in the present arrangement with an efficiency of only 15 per cent but, since 16 per cent of the X - radiation undergo Auger transitions, the resulting Auger electrons of energies 25.8keV and 22.5keV are detected with 100 per cent efficiency and contribute to the somewhat complex distribution centred at ~ 27 keV.

The other peaks, or change in slope, indicated in Fig 20 can be explained by the integration, by the counter, of coincident events (the delay at the 35.5keV energy level is $< 10^{-9}$ sec). In particular, apparently in

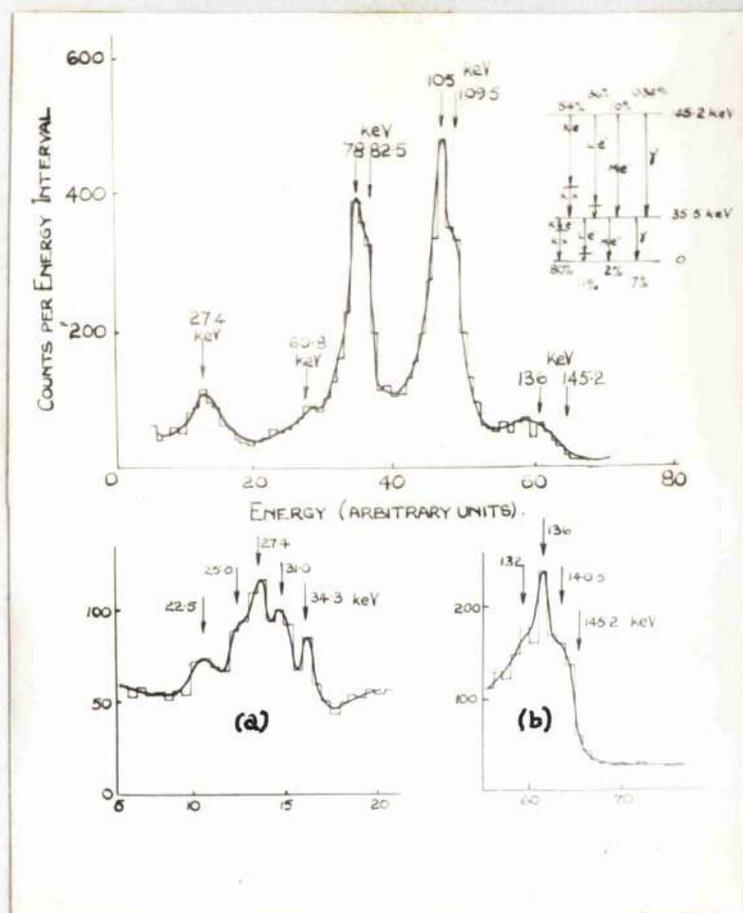


Fig 20. Electron spectrum of ^{125m}Te analysed by the proportional counter. Insert shows decay scheme (Bowe and Axel, 1952).

(a) and (b) Closer examination of the spectrum in the regions of 27 and 136keV respectively.

contradiction to the decay scheme, the intensity of the 78keV peak is less than that at 105keV. This is due to the fact that, in addition to electrons of energy 105keV, integration of a K X - ray with an electron of 78keV can give a pulse corresponding to a total energy of $(27.4 + 78) = 105.4\text{keV}$. Similarly, though of smaller probability, integration of an Auger electron with a 78keV electron also makes a contribution to the peak.

Taking into account the efficiency of detection of the K X - rays and the relative probabilities of the various integration processes possible (using the data shown in Fig 20), the ratio of the areas under the peaks at 78 and 105keV respectively was calculated as 0.79. This compares with an experimentally determined ratio of 0.77 and gives confidence, both in the postulated decay scheme and in the accuracy of the present measurements with the proportional counter.

An interesting check on the linearity of the energy versus pulse height relationship obtained with the proportional counter is obtained from the results of Fig 20. The energy values assigned to the various peaks and discontinuities of the curve are taken from a consideration of the decay scheme. By replotting these

assignments, in the form of a pulse - height versus energy diagram, a perfect linear relationship is obtained (Fig 21).

3. APPARATUS.

The proportional counter was designed to facilitate the examination of various scattering materials. The counter, shown diagrammatically in Fig 22, has a diameter of 14cm and a fully - operating length of 30cm. In the side wall an aperture, A, of diameter $\frac{1}{2}$ inch, is cut, and close to this is the source support, consisting of part of a nylon film in the form of a circle of the same diameter. This is held between two brass plates, Sh, each 0.015 inch thick and suitably perforated. These plates are screwed to the counter wall. A wheel, placed close behind the source holder, allows foils, F, of six different substances, or foils of the same substance in six thicknesses, to be rotated into position behind the source, this adjustment being made accurate by means of a ball and socket arrangement. The distance between the source and back - scattering foil is 0.025 inch. In order to confine the paths of the electrons to the vicinity of the source and the backing foils, the counter was placed in a magnetic field, of strength between 2000 and 4000 gauss, the direction of the field being perpendicular to

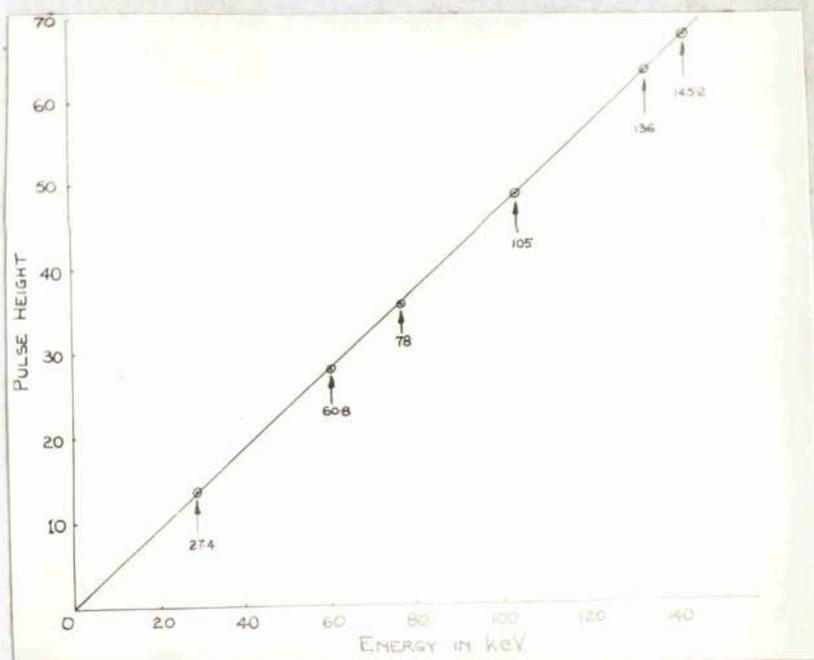


Fig 21. Linearity of proportionality derived from ^{125m}Te spectrum (Fig 20).

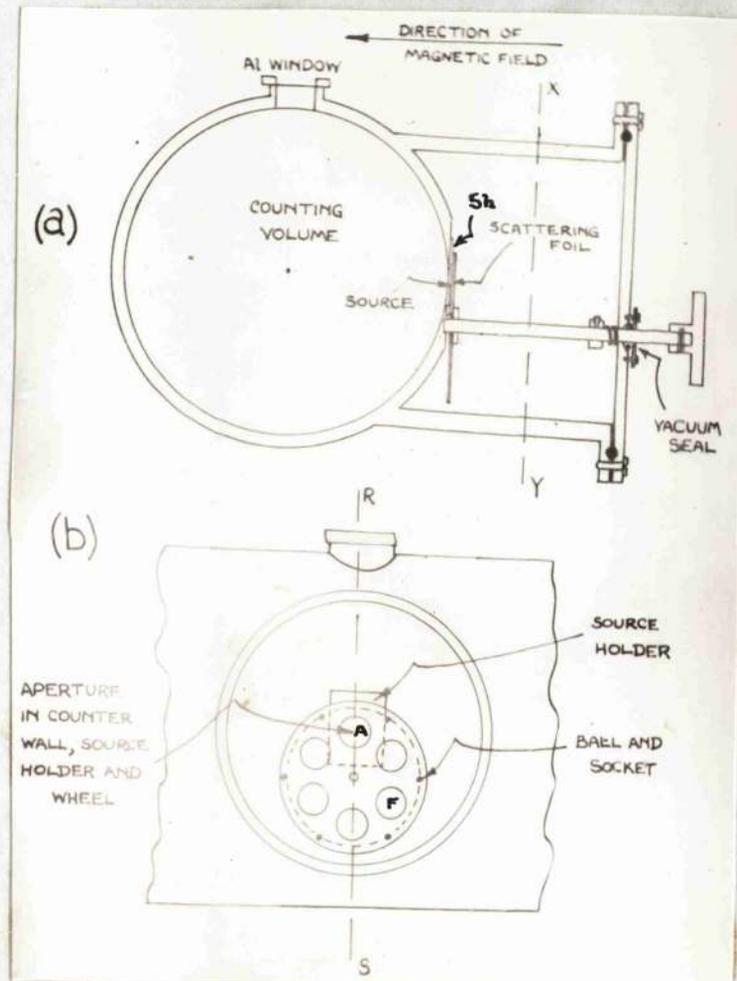


Fig 22. Diagram of counter. (a) Sectional end view (across RS), (b) Sectional side view (across XY). The direction of the applied magnetic field H is as shown.

It is known (Burtt, 1950; Yaffe, 1950) that P_x increases with increasing x until a saturation value is

reached at $x = s$. The corresponding value of P_x is the "saturation back - scattering coefficient" for the arrangement, the back - scattering is independent of gas pressure in the counter for pressures between $\frac{1}{2}$ and 2

atmospheres, the magnetic field ensuring that the higher energy electrons are completely stopped within the

counting volume. In this experiment no attempt was made to separate out the various energy components of ^{226}Ra . Walls of aluminium of thicknesses 0.002, 0.005, 0.001, 0.002, and 0.006 inch were used. The count rate with no aluminium foil behind the source was $\sim 10^4$ per minute.

4. COEFFICIENT OF BACK - SCATTERING.

The coefficient of back - scattering from a foil of thickness x is defined by

$$P_x = \frac{N_x - N_0}{N_0} \times 100 \%$$

where N_0 , N_x are the counting rates with no backing material, and with thickness x of backing material respectively, both of these being corrected for the natural background counting rate of the counter. (In the following experiments N_0 was taken as the counting rate with only thin nylon source support behind the source. Later results showed that the error introduced by this procedure is negligible).

It is known (Burtt, 1950; Yaffe, 1950) that P_x increases with increasing x until a saturation value is reached at $x = s$. The corresponding value of p , viz p_s , is the "saturation back - scattering coefficient" for the particular material and s is the "saturation thickness".

are tabulated below. (Shown by Yaffe and Justus that

5. VALUE OF SATURATION THICKNESSES.

In this experiment no attempt was made to separate out the various energy components of ^{125m}Te . Foils of aluminium of thicknesses 0.0002, 0.0005, 0.001, 0.002, and 0.006 inch were used. The count rate with no aluminium foil behind the source was $\sim 10^4$ per minute.

The values of P_x , found for each foil thickness, were obtained after correcting (a) for the background of the counter, and, (b) for the fact that part of the total count rate is due to X - rays, and is unaffected by

backing material. The resulting curve (Fig 23) gives a saturation thickness of 0.002 in aluminium and a saturation back - scattering coefficient of 11 per cent. Yaffe and Justus (1949) have given the following relationship for calculating saturation thickness:-

$$M_s^2 = 36R$$

where M_s = mass in mg/cm^2 required for saturation.

R = maximum range in (mg/cm^2) of the electrons.

For ^{125m}Te this gives a value of $M_s \doteq 26 \text{ mg}/\text{cm}^2$ for the

105keV group of electrons. For aluminium this corresponds to a thickness of 0.0037 in, a value considerably in excess of the thickness determined experimentally. Mass thicknesses considerably greater than the calculated value were used in subsequent experiments. The values used are tabulated below. (Shown by Yaffe and Justus that mass required for saturation is independent of Z).

TABLE 2.

| Back-scattering Material | Polythene | Aluminium | Copper | Silver | Gold |
|--------------------------------------|-----------|-----------|--------|--------|------|
| Atomic N°Z | 6 | 13 | 29 | 47 | 79 |
| Mass thickness in mg/cm ² | 45.6 | 41.0 | 45.2 | 53.4 | 48 |

6. VARIATION OF ρ_s WITH ATOMIC NUMBER Z.

Again, in this experiment, no examination is made of dependence of ρ_s on energy, i.e. the results are valid for the ^{125m}Te spectrum taken as a whole within the energy range ~ 8 and ~ 105keV.

Variation of ρ_s with atomic number was studied by

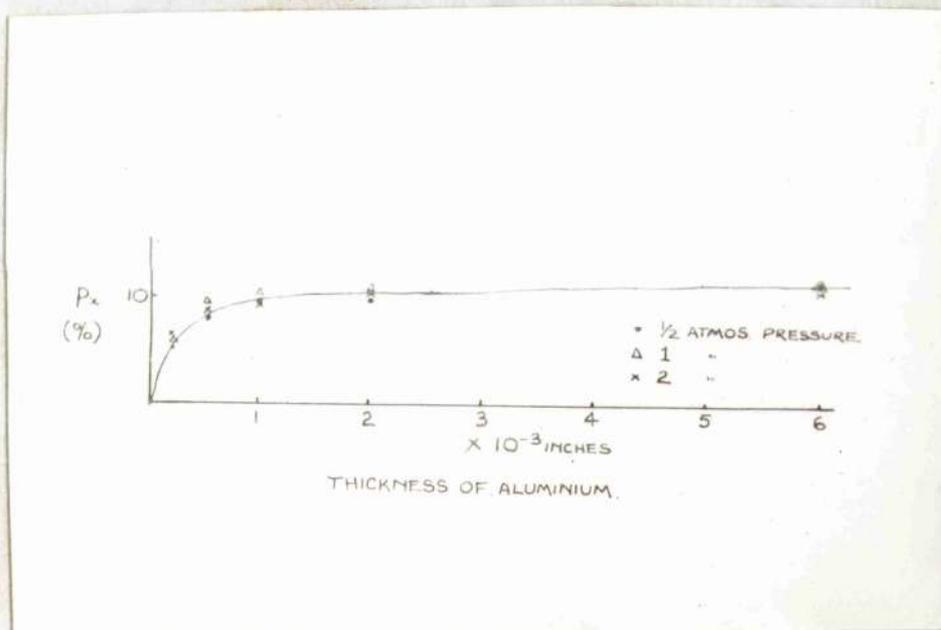


Fig 23. Variation of back - scattering coefficient P_x with thickness of back - scattering foil.

bringing in turn each of the materials tabulated in Table 2. The results (Fig 24) lie along a smooth curve and this curve is found, within experimental limits, to be independent of pressure of gas filling of the counter between $\frac{1}{2}$ and 2 atmospheres. This indicates that back - scattering of the electrons by the gas between the source and scattering material is negligible.

The experiment was repeated, with increased gas gain, so that all particles down to an energy of 1keV were counted. The resulting points fell accurately along the same curve. This shows that, at least above an energy of 1keV, there is no appreciable low energy part in the spectrum of back - scattering^{ed} electrons.

7. VARIATION OF p_s WITH ELECTRON ENERGY.

The counter was filled to a pressure of 1 atmosphere (10 per cent methane, 90 per cent argon) and placed in the magnetic field. The spectra of the pulses produced by a very thin source of ^{125m}Te were then measured in two ways.

- (A) With the source deposited on nylon but with no other material behind the source.
- (B) With saturating thicknesses of aluminium, silver, and gold brought behind in turn.

To facilitate subsequent analysis each separate run was

made in exactly the same time, the pulses being recorded by the moving film technique.

The procedure of analysis is understood with reference to the spectra (Fig 25) obtained with silver behind the source. Subtraction of curve A from curve B gives the shape and intensity of the spectrum of the back - scattered electrons. This subtraction is not very easily made for energies less than 30keV and this energy region was examined separately as described below.

It is now necessary to analyse the compound spectrum into two separate parts corresponding to back - scattering of primary electrons of energies 105keV and 78keV. Little assistance in tracing the correct shape of these component spectra is obtained from the theory of multiple scattering, which in most cases is limited to small - angle scattering and neglects completely scattering in the backward direction. In general, however, the theory of multiple scattering leads, for an incident monoenergetic beam of electrons, to an energy loss curve, which is approximately Gaussian in shape, centred on the position of mean energy loss, with a high - energy - loss "tail". This tail is only approximately exponential in shape and appears to be dependent on the material in which scattering takes place.

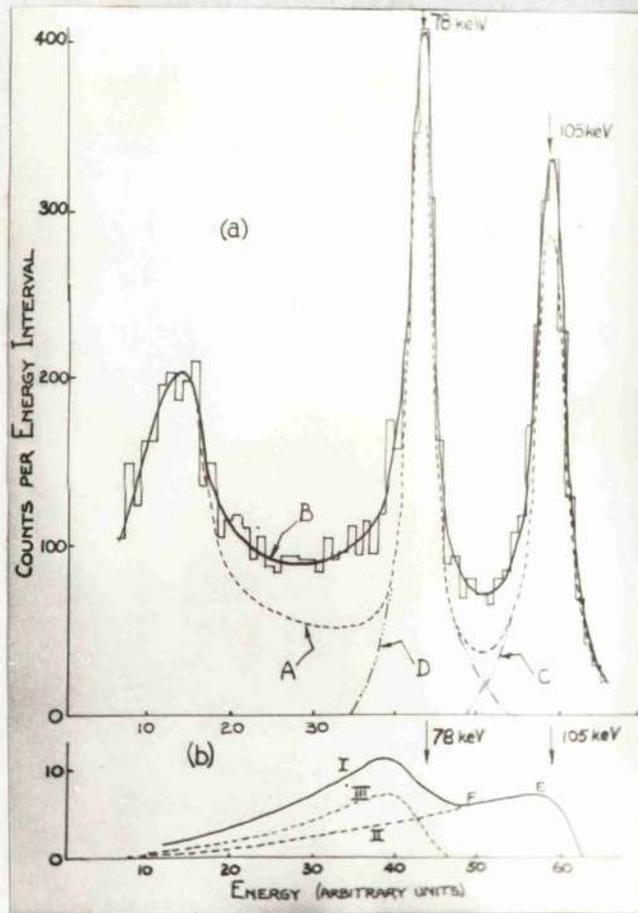


Fig 25. Analysis of the spectrum of electrons of ^{125m}Te back - scattered from silver foil.

The part EF (Fig 25(b)) of the back - scattered spectrum can be accounted for only by electrons of primary energy 105keV and can be drawn unambiguously from the experimental curves. The lower energy part of curve 2 is obtained by extrapolating exponentially. The resultant curve 2 then represents the shape of the spectrum for electrons of primary energy 105keV back - scattered by a saturation thickness of silver into a solid angle of 2π . The resultant curve 3, obtained by subtraction of curves 1 and 2, must be the corresponding spectrum for electrons of primary energy 78keV. (In order to make curves 2 and 3 of the same shape and still combine to give curve 1 it was found necessary to depart somewhat from an exponential tail). The coefficients of back - scattering are now obtained by dividing the areas under curves 2 and 3 by the areas under the corresponding peaks (C and D) in the spectrum obtained with no foil behind the source. This gives two points on a plot of p_s versus energy, for silver backing. The results were recalculated to take account of the fact that ~ 15 per cent of the peak at 105keV is due to integration, by the counter, of 78keV electrons with electrons of the lower energy group (~ 10 per cent contribution) or with X - rays (~ 5 per cent contribution). The resulting increase in the previous value of p_s for an energy of

78keV, at the expense of a slight decrease in the value for 105keV, was found to be almost negligible.

It is seen that only a small fraction of the area under Curves 2 and 3 lies below an energy of the order 34keV, which represents the upper - energy limit of the lower - energy complex group of electrons. The average coefficient P_s for this group, difficult to obtain by the above method in view of the closeness of curves A and B, can thus be found by a purely counting method without serious error due to electrons of energy less than 34keV originating from high energy loss of electrons of primary energies 105 and 78keV. For this measurement the discriminator bias was set to obtain:

- (1) count rate N_1 due to electrons of energy greater than 5keV
- (2) count rate N_2 due to electrons of energy greater than 34keV;

each of these being measured under both conditions:

- (a) no scattering foil behind the source, and then
- (b) saturation thickness of foil behind the source.

Then the coefficient of back - scattering is given by

$$P_s = \frac{(N_1 - N_2)_{(b)} - (N_1 - N_2)_{(a)}}{(N_1 - N_2)_{(a)}} \times 100 \%$$

This gave a third point, at the average energy of 28keV, on the ρ_s versus energy curve, after slight correction was made for the small part of Curves 2 and 3 lying below an energy of the order of 34keV.

It is found that a smooth curve can be drawn, with fair certainty, through these points (Fig 26). As a further check on this curve, use is made of the previous value of ρ_s obtained when the tellurium spectrum was treated as a whole (Fig 24). The average energy of the ^{125m}Te spectrum, as observed here, is close to 57keV. This further point is found to fit well on the curve, thus giving confidence in its validity.

The same procedure was applied with aluminium, silver and gold scattering foils, and the results are collected as a family of curves in Fig 26. The results can be re - plotted as a function of Z instead of energy (Fig 27). From the latter figure it is seen that the curves extrapolate fairly smoothly back to a value of $\rho_s = 0$ at $Z = 0$. This is interpreted as indicating that no appreciable scattering occurs from the thin nylon support. (If such an effect were appreciable, the extrapolated curves would, for $Z = 0$, cut the vertical axis at a negative value of ρ_s which would then have to

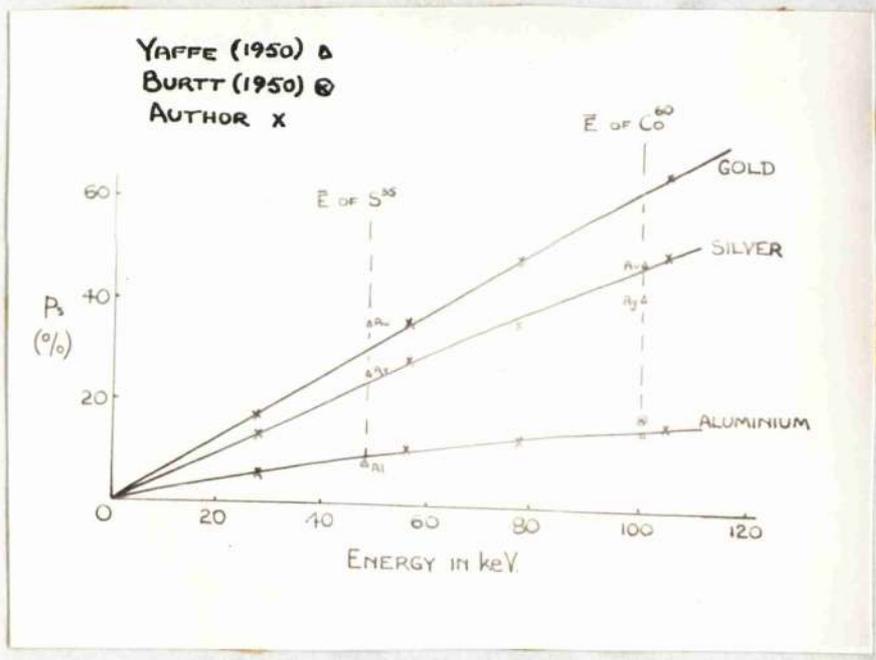


Fig 26. Variation of P_s with energy for various scattering materials.

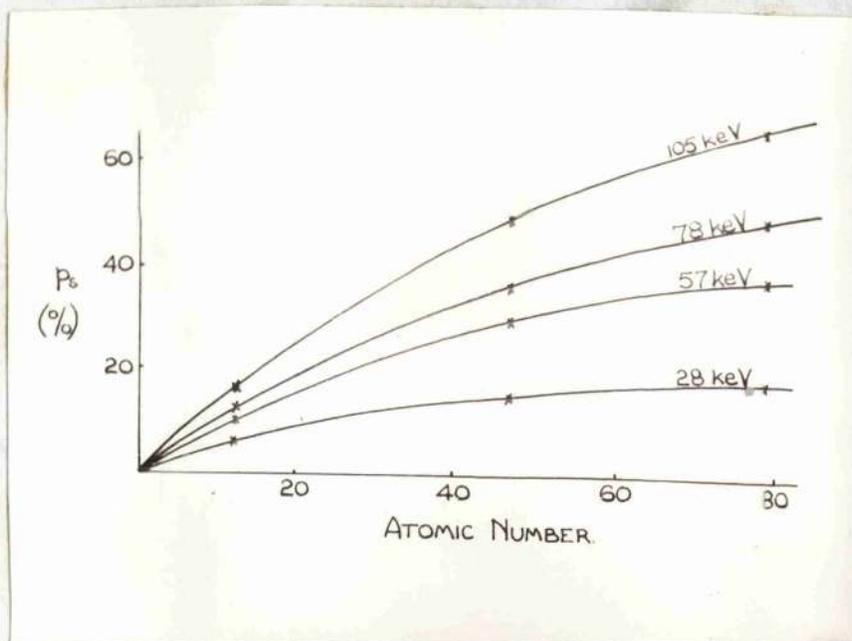


Fig 27. Variation of saturation back - scattering coefficient P_s with atomic number for various energies.

be added to the experimentally determined values as a correction factor).

It is interesting to note that that maxima of curves 2 and 3 of Fig 25 occur at lower energies than 105 and 78keV respectively. A shift of this type was predicted by Weymouth (1951) on the basis of multiple scattering of charged particles during transmission through a thick foil, and was in good agreement with the experimental results of Hornyak, Lauritsen, and Rasmussen (1949) obtained for electrons ejected from a thick lead converter by the γ - rays of ^{198}Au (of energy 411keV giving maximum energy of electron of 301.3keV, but showing a peak shift to (301.3 - 4.3)keV). In view of the difficulty of obtaining an exact shape of the peak in the spectrum of back - scattered electrons, by subtracting areas, the present results do not afford a rigorous check or extension to the theory of this peak shift phenomenon.

8. COMPARISON WITH RESULTS OF OTHER WORKERS.

All previous investigations on back - scattering of electrons from radioactive sources have been made by means of Geiger counters (with the exception of Brownell (1952)). These fall into two distinct groupings.

(a) Source placed outside thin end window. The work of Burt (1950) and Yaffe (1950) falls into this category.

The sources, mounted on formvar or nylon films, were placed close to the thin end window of the counter, and foils of different materials brought close behind the source. Continuous β -spectra were used and the variation of ρ_s with energy plotted as a function of maximum energy E_0 of the β -spectrum. The initial part of the curves, rising steeply with increasing energy, were not examined very closely and it is to this region that the present work particularly applies. In view of this marked variation with energy it would appear to be more correct to plot ρ_s , not as a function of E_0 , but of the average energy \bar{E} of the spectrum, since β -spectra with the same upper energy limit but with different shapes will have different average values of ρ_s .

A value of \bar{E} for the β -spectra of ^{35}S and ^{60}Co , obtained from the spectrum shapes observed by Cook, Langer and Price (1948) and by Fan (1952) respectively, is used to compare the author's results with those of Yaffe and Burtt. The comparison is shown on fig 26.

An alternative method of comparison is to use the curves of Fig 26 and the shapes of the β -spectra to calculate the mean value of ρ_s for ^{35}S and ^{60}Co , the resultant average value being weighted according to the

shapes of the β - spectra. To make this possible the curves of Fig 26 had to be extrapolated to higher energy values, and, to reduce error in this extrapolation, guidance was taken from the value of ρ_s obtained for ^{32}P ($\bar{E} = 685\text{keV}$) by Yaffe (1950). Objection could be made that such an extrapolation will give results biased towards those of Yaffe. This may be partly true in the case of ^{60}Co but, for the case of ^{125}S ($E_0 = 169\text{keV}$), only a small part of the spectrum lies within the extrapolated range and therefore no appreciable bias towards Yaffe's final result is introduced. The results of the calculations are shown in Table 3.

In general, although agreement of the author's results with those of group (a) is fairly good, the results obtained with Geiger counters are seen to be rather higher than those obtained by calculation from the ^{125}Te results. This is not unexpected in view of the difference in acceptance solid angle. In the proportional counter experiments, this has a value of 2π , somewhat greater than for the Geiger counter measurements, and it has been shown by Brownell (1952) that the intensity of back - scattered electrons is greatest in a direction normal to the plane of the scattering foil i.e. the

back - scattered spectrum will be more predominant in the case of the smaller acceptance solid angle.

(b) Source inside Geiger counter. This method, aimed at eliminating uncertainties due to absorption at the window of the Geiger counter, was employed by Suzor and Charpak (1952) and by Christian et al (1952). The former used sources mounted on thin aluminium foil at an aperture between two semi - cylindrical Geiger counters, and by a coincidence technique, deduced the variation of back - scattering coefficients with energy by introducing various thicknesses of aluminium foil in front of the source. Christian et al used a source placed on the inside of the counter end - wall. The results (Table 3) are in marked disagreement with those calculated from the experiments with ^{125m}Te. Further, analysis by Suzor and Charpak shows that ρ_s decreases with increasing energy, contrary to the evidence of Fig 26.

9. DISCUSSION OF DISCREPANCIES.

Generally, the agreement between the author's results and those obtained by method (a) is fairly good, but all of these results are in marked disagreement with those obtained by method (b). There would appear

TABLE 3.

| Source | ^{35}S | | | ^{60}Co | | |
|-------------------------------------|-----------------|----|----|------------------|-----|----|
| Scattering Material | Al | Ag | Au | Al | Ag | Au |
| By calculation from the results. | 8.6 | 23 | 30 | 14 | 41 | 49 |
| Method (a) | | | | | | |
| Yaffe | 8 | 27 | 36 | 14.5 | 40 | 46 |
| Burt | | | | 17 | ~50 | |
| Tobias | | | | 17 | | 53 |
| Method (b) | | | | | | |
| Christian et al | ~23 | 52 | 52 | 33 | 53 | 56 |
| Suzor and Charpak | 33 | | | 33 | | |

to be some definite explanation of why the introduction of a source inside a Geiger counter should lead to such a marked difference.

It is well known (Rudberg, 1936; Jonker, 1951) that, when a mono - energetic beam of electrons impinges on a target, there is obtained, in addition to a spectrum of back - scattered electrons, a spectrum of low - energy true secondary electrons of energies a few tens of electron volts. Such electrons would not be detected by means of the method (a) nor in the experiments with the proportional counter. It is possible, however, that they could be detected when the radioactive source is inside a Geiger counter (method (b)).

A calculation was made on the assumption that a continuous β - emitting source could be treated as the source of a number of mono - energetic beams of energies ranging from 0 to E_0 , the intensity of each beam being determined by the intensity of the β - spectrum at that particular energy. Known data on the measured secondary electron yield, δ , as a function of primary electron energy (Trump and Van de Graaf, 1949), and as a function of angle of emission (Müller, 1937), was applied in this way to the case of a thin source of ^{60}Co deposited on

aluminium backing. The results of these calculations indicate that emission of secondary electrons into a solid angle of 2π on the source side of the backing material could amount to 25 per cent of the primary β -particle intensity for this particular source and backing. Thus if at least a fraction of these secondary electrons can be detected by a Geiger counter the discrepancies in the results on back - scattering might be explained. Chapter 8 is devoted to an account of a search for these soft secondary electrons.

CHAPTER 8.EXAMINATION OF POSSIBLE SECONDARY
ELECTRON EMISSION ACCOMPANYING
BACK - SCATTERING OF ELECTRONS.1. APPARATUS.

The special Geiger counter designed for this experiment is shown diagrammatically in Fig 28. It consists of a brass cylindrical cathode, of internal diameter 1 in and length 2 ins, terminated at one end by a glass seal acting as an axial support for the anode. The anode is made of tungsten wire (0.008 in in diameter) and terminated by a glass bead. The source, deposited directly on a metal disc source - support, rests on an ebonite disc to insulate it from the walls of the counter. By means of brass terminals, passing through ebonite plugs, the potential of the source with respect to the cathode can be adjusted. A short copper cylinder, carrying twelve tungsten wires (each 0.008 in diameter) spaced 2mm apart to constitute a grid, fits tightly inside the

cathode and ensures that the grid is always at the same potential as the cathode. The distance between grid and source is 2mm.

Using this counter it was intended, by application to the source of a positive or negative potential with respect to the grid, to prevent the soft secondary electrons from entering the counter, or to accelerate them into it respectively.

2. EXPERIMENTAL RESULTS.

A source of ^{125m}Te (counting rate approximately 5000 per minute) was deposited on an aluminium disc 0.010 in thick. The counter was filled with a mixture of alcohol (5 per cent) and argon (95 per cent) and the anode voltage always kept constant at a value corresponding to the centre of the observed Geiger plateau. The position and length of this plateau was found to be independent of potential applied to the source. A measurement was made of counting rate as a function of voltage applied between source and grid.

With a view to separating out the soft secondary electrons from the primary or back - scattered electrons,

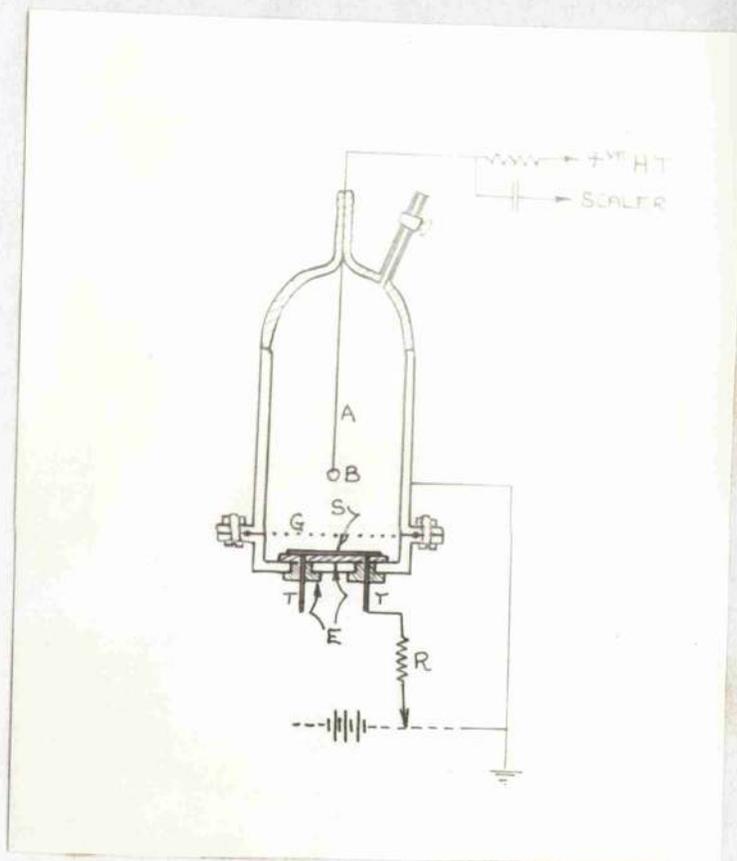


Fig 28. Diagram of gridded Geiger counter used.

A : anode B : glass bead G : grid

S ; thin source on disc - shaped holder

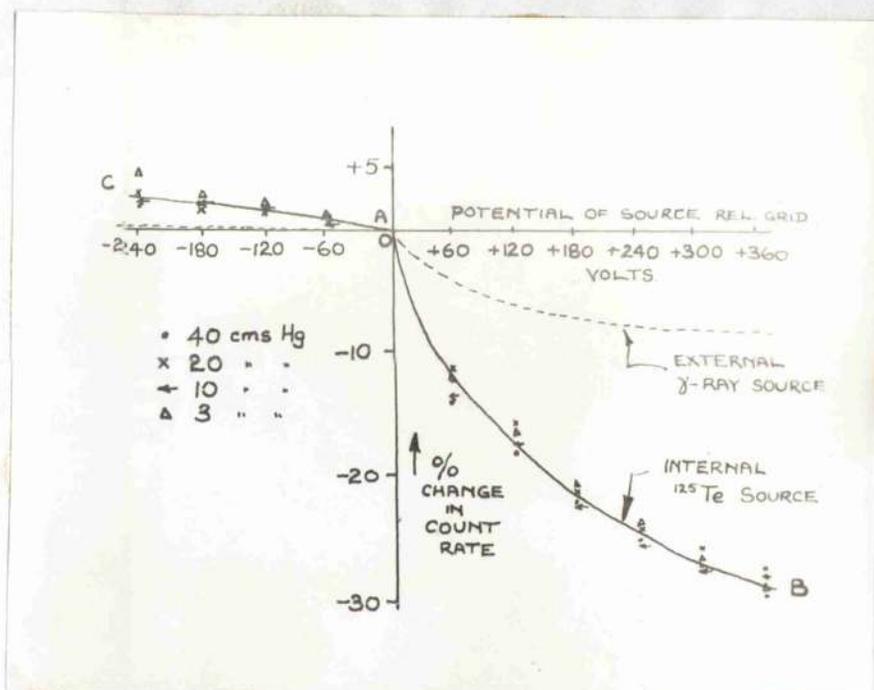
E : ebonite insulator

R : large resistance to prevent accidental drawing of large current in discharge.

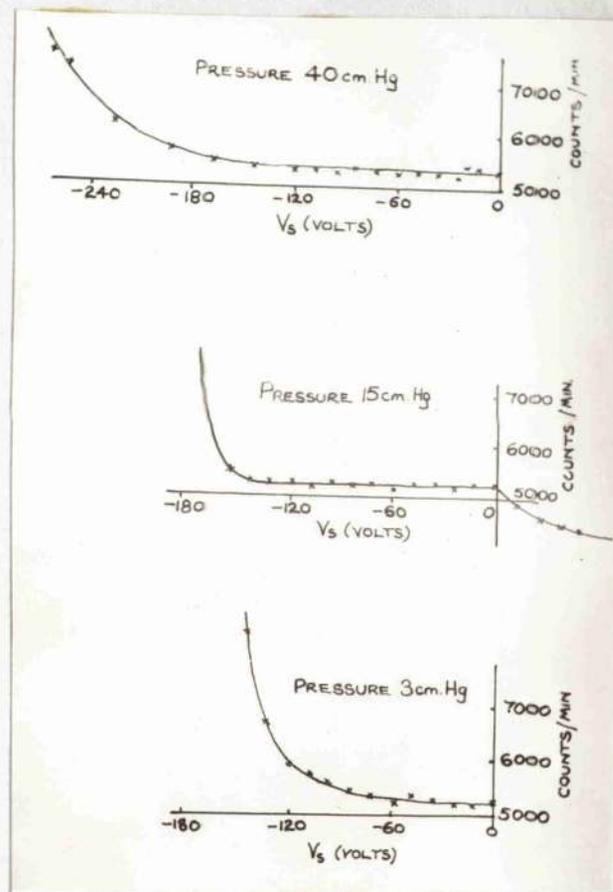
the experiment was performed with different pressures (3, 10, 20, and 40 cm. mercury) of gas filling. At the highest pressure the secondary electrons should not be sufficiently energetic to pass into the counter beyond the grid although the primary electrons would be able to do so. Reducing the pressure should have little or no effect on the primaries, but should increase the probability of the secondary electrons entering the counter. Thus the presence of secondary electrons should be shown by increase in the observed counting rate with decrease in pressure, and with increasing accelerating grid action.

The results obtained (Fig 29) were almost independent of pressure when the source is positive with respect to the grid. The slight indication of dependence on pressure, in the way visualised, obtained with increasing negative source potential was found, by closer examination (Fig 30), to be false, and due to an increasing tendency of the counter to break into discharge at lower source potentials as the pressure is successively decreased.

In view of the dependence of secondary electrons yield on the energy of the primary electrons and on the atomic number, Z , of the target material, the experiments



- Fig 29. Variation of counting rate as a function of potential applied to source. Source of ^{125m}Te deposited on aluminium.



- Fig 30. Closer examination of the variation of counting rate with pressure in the region of negative source potential.

were repeated with different sources (^{35}S and ^{60}Co) on backing materials of various Z values (aluminium, copper, and silver). Exactly similar results were obtained in all cases.

3. CLOSER EXAMINATION OF THE RESULTS OBTAINED WITH POSITIVE POTENTIAL APPLIED TO THE SOURCE.

Despite the diversity of conditions under which the preceding experiments have been made, the same per centage fall in counting rate was always observed with positive source potential (part AB of the curve in Fig 29). A possible explanation of this fact is that under these conditions a local decrease in electric field in the region of the grid is produced, with consequent insensitivity of the counter in this region.

This theory was checked by removing the source from the counter and replacing it by a copper disc exactly similar to the source mountings used, in order to preserve the same electrical conditions. A well - collimated beam of γ - rays from an external source of ^{60}Co was allowed to pass:-

- (a) through the end wall of the counter,
- (b) through the side wall of the counter level with the bead on the anode.

The results obtained in these two cases were identical, within experimental error, Comparison (Fig 29) of these results with the results obtained with an internal source shows the same general shape of curve but the fall in counting rate is reduced by a factor of about 4 in the case of the external γ -ray source. This is interpreted as being consistent with the hypothesis regarding loss of sensitivity of the counter, because, in the case of the external γ -ray source, most of the electrons detected are produced in the walls of the counter and can cause ionisation in regions of the counter which are of unimpaired sensitivity of detection.

It appears, therefore, that part AB (Fig 29) of the curves obtained with the various internal sources is due to local reduction in sensitivity and not to any retarding action of the grid on the slow secondary electrons. On this view it is of no assistance in the investigation of these secondary electrons.

4. CLOSER EXAMINATION OF RESULTS OBTAINED WITH NEGATIVE POTENTIAL APPLIED TO THE SOURCE.

The part AC (Fig 29) of the experimentally determined curves gives no indication of the presence of secondary electrons, either by accelerating grid action

or by variation of counting rate with pressure. Before concluding that this proves the non - existence of these secondary electrons, a more detailed study of the counter used was considered to be necessary to ensure that it is capable of detecting them efficiently if they are present.

For this purpose a heated tungsten filament was used as a source of low energy electrons. A tungsten wire, .006in. in diameter, was connected internally between the end terminals, at the same distance from the grid as the source had been. A copper disc, identical with the source support, was located directly below the filament to ensure a field distribution as nearly identical as possible to that of the previous experiments. The filament current was supplied by a 4 volt battery and the potential of the filament with respect to grid varied as before.

The operation of the counter was somewhat unstable, but by checking (a) the constancy of the Geiger plateau, with an external source of γ - rays (^{60}Co), and (b) the constancy of the counting rate with no potential difference between filament and grid, between each reading, the fairly reliable results plotted in Fig 31 were obtained. The form of these curves is what had been anticipated,

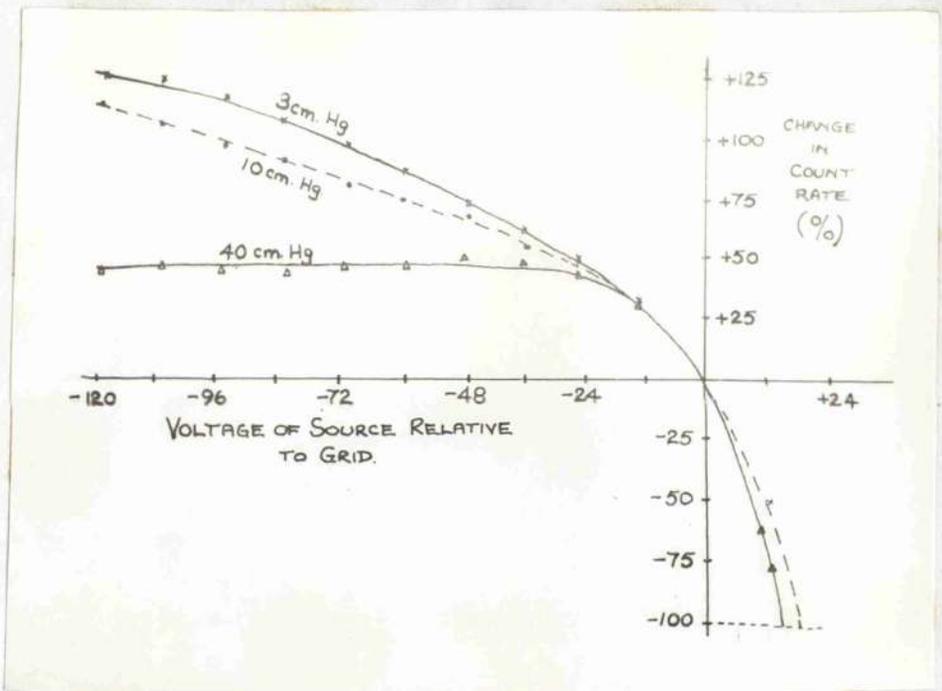


Fig 31. Percentage change in counting rate as a function of gas pressure obtained with thermal source of electrons.

namely a decided increase in counting rate with increasing accelerating field and with decreasing pressure.

Estimation of the efficiency of the grid, by measuring the counting rates obtained with and without the grid in position, proved somewhat difficult due to instability of counter operation, but it appeared to have an efficiency of between 50 and 100 per cent. It therefore appeared to be likely that the grid system used would be capable of detecting slow secondary electrons, but only if the counter itself were efficient for their detection.

A convenient counting rate of about 100 counts per second had been obtained by slowly increasing the filament current to a value of 1.35 amps. Application of known data (Spangenberg, 1948) on the thermionic emission characteristics of tungsten to this particular filament indicate an emission current of about $\frac{1}{10}$ μ amp, corresponding to a counting rate of the order of 10^{12} per second. This indicates that, under the conditions of these preceding experiments, the counter used is extremely inefficient for the detection of low energy electrons (~ a few tens of electron volts).

This marked inefficiency could possibly be due to the use of a glass bead on the end of the counter wire, a practice which reduces considerably the electric field between the anode and the end wall on which the sources have been positioned. In view of the rather important implications of this hypothesis, and the widespread use of Geiger counters of the "glass - bead" type, it was considered advisable to seek further evidence of its truth.

5. EXAMINATION OF THE INEFFICIENCY OF GEIGER COUNTERS IN THE VICINITY OF THE END WALL.

One way of seeking confirmation of the above hypothesis is to use a radioactive source emitting electrons of low energy and to position the source (a) on the end wall, and (b) on the side wall of a Geiger counter. In the absence of a source of this description a fair approximation to the requirements is obtained by using a source of moderately energetic electrons placed in a magnetic field of strength sufficient to cause the electron paths to spiral, confine them to the proximity of the source, and thus not penetrate far into the counter.

For this purpose the simple Geiger counter shown

122.

diagrammatically in Fig 32 was constructed. It consists of a brass tube, $4\frac{1}{2}$ ins long, and 1in diameter, terminated at the ends by good quality rubber stoppers R. The anode W, of 0.008in. diameter tungsten wire, is terminated by a glass bead G, the position of which, relative to the end wall, is, for the sake of comparison with the previous results, approximately the same as in the "gridded" Geiger counter used previously. To eliminate small pulses due to electrical discharge, the central wire is shielded near the end by a brass tube T, held at the same potential as the anode. The source, deposited directly on to a metal backing, was fixed in position at either A or B. To prevent errors due to insulation of the source from the cathode (Braden et al, 1948) the rubber stopper at A was rendered conducting with graphite. The counter was placed in a magnetic field as shown diagrammatically (Fig 32A, 32B). Gas filling was a mixture of alcohol (5 per cent) and argon (95 per cent) to a total pressure of 10cm mercury.

Several measurements of counting rate, with and without the applied magnetic field, were made in each of the positions shown in Fig 32, both with sources of ^{35}S (giving a β -spectrum of end point 169keV) and with ^{125m}Te . All of the results obtained were in good agreement and

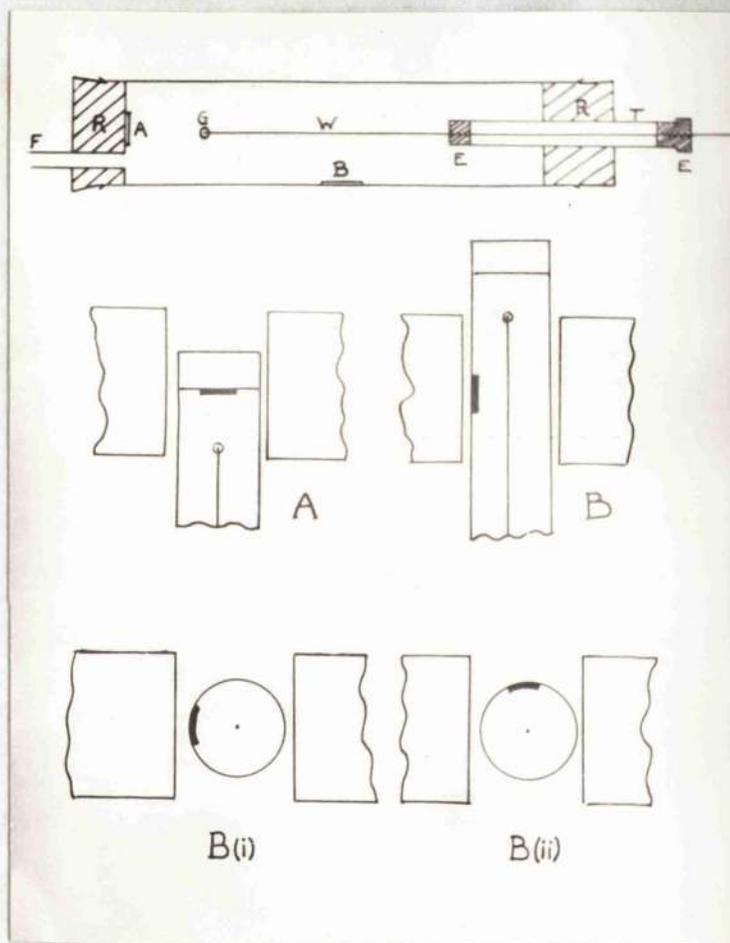


Fig 32. Geiger counter used in study of inefficiency for detection of secondary electrons. The various positions of the counter in the magnetic field are shown.

those shown in Table 4 are fairly representative:

TABLE 4.

Counting rate (per $\frac{1}{2}$ minute) obtained in the various arrangements (Fig 32) with sources deposited on aluminium.

| Source | Position A | | Position B | | |
|--------------------|------------|----------|---------------------------|---------------------------|-----------|
| | Field Off | Field On | Position B(1) Field on | Position B(2) Field on | Field Off |
| ^{125m}Te | 416 | 0 | 2904 | 2873 | 2895 |
| ^{35}S | 1001 | 135 | 5996 | 5988 | 5992 |

All of these results are corrected for the natural background counting rate of the counter (unaffected by magnetic field).

It can be deduced from the unaltered counting rate observed in position B, irrespective of application or removal of the magnetic field, that the counter is, in this arrangement, as sensitive for the detection of low energy particles as for those of moderate energy ($\sim 100\text{keV}$).

Transfer of the same source to position A gives only about 16 per cent of the previously determined counting rate, and on application of the magnetic field this figure reduces to zero (for ^{125m}Te) or to 2 per cent (for ^{36}S). (part, or all, of this 2 per cent could be accounted for by some of the electrons escaping from the effect of the magnetic field and entering the sensitive part of the counter near or beyond the glass bead).

It is concluded from these experiments that a Geiger counter of the type in which the anode is terminated by a glass bead, is extremely inefficient for the detection of low energy electrons emitted by a source in the vicinity of the end wall.

6. FURTHER SEARCH FOR SECONDARY ELECTRONS.

The conclusions of section 5 show that the method adopted (sections 1 and 2) would not establish the presence of soft secondary electrons, even if these were present. At the same time the results of section 5 indicate a method by which their absence or presence could be established, namely by using a radioactive source placed on the side wall of the counter in conjunction with a modified grid system, cylindrical in form, and

120.
~~concentric~~ ^{coaxial} with the counter.

Since this would entail considerable re - design of the counter, an alternative method, more readily applicable, is used. It is based on the fact that, while a Geiger is capable of detecting free electrons of energy down to zero, a proportional counter need not be operated to do so. Thus by using a source located on the side wall of a counter tube, a measure of the difference in counting rates obtained with the same tube, operated both as a Geiger counter and as a proportional counter, should give a measure of the secondary electron yield for the particular source and backing material used.

7. EXPERIMENTAL PROCEDURE AND RESULTS.

The counter used was that described previously (Fig 32). When used as a proportional counter, the negative high tension voltage was applied to the case and the tube T earthed. By firing the K X - rays of silver into the counter and examining the resultant array of pulses on a cathode ray tube, a measure was obtained of the energy of particle being counted. Use of a discriminator then ensured that only pulses corresponding to particle energies greater than 1keV were counted. A counter filling consisting of methane (8cm mercury) plus argon

(54cm mercury) was found to be suitable. For Geiger operation an argon - alcohol mixture (ratio 20:1) at a pressure of 10cm mercury was used. In each case correction was made for the dead time τ of the Geiger counter (large compared with that of the proportional counter) using the value of τ determined experimentally (Curran and Craggs, 1949).

Various sources and source - mountings were used:-

(a) Source of ^{60}Co deposited on copper.

To reduce uncertainties arising from low energy primary electrons, a source of ^{60}Co was chosen because of the negligible portion of its β - spectrum lying below 1keV. Self absorption of the secondary electrons in the source itself is reduced by preparing it in the thin uniform manner obtainable by thermal evaporation in vacuum. A plateau was plotted for both modes of operation (Fig 33) and from these curves the following results are taken:

Counting rate (proportional operation) = 6,400 counts per
minute.

Counting rate (Geiger operation) = 6,450 counts per
minute.

(b) Source of ^{35}S deposited on copper.

Because of the fact that the secondary electron emission yield increases with decreasing electron energy (Trump et al, 1949), ^{35}S was used as a source (the maximum energy of its β -spectrum is 169keV compared with 310keV for the ^{60}Co β -spectrum). Thus a greater difference in counting rate is to be expected. The results:-

Counting rate (proportional operation) = 12,550 counts per
minute.

Counting rate (Geiger operation) = 12750 counts per
minute,

show a counting rate about 2 per cent greater in the case of Geiger operation. In view of the uncertainty of the fraction of the ^{35}S β -spectrum lying below 1keV and not counted in proportional operation, and of the accuracy of the experiment, this difference can be accounted for on grounds other than proof of the existence of secondary electrons.

(c) Source of ^{125m}Te on tin.

In view of the X - rays emitted by this source, the two types of gas filling were chosen to give approximately the same stopping power for X - rays. These fillings were:-
Proportional : 15cm mercury of argon + 4cm mercury of
methane.

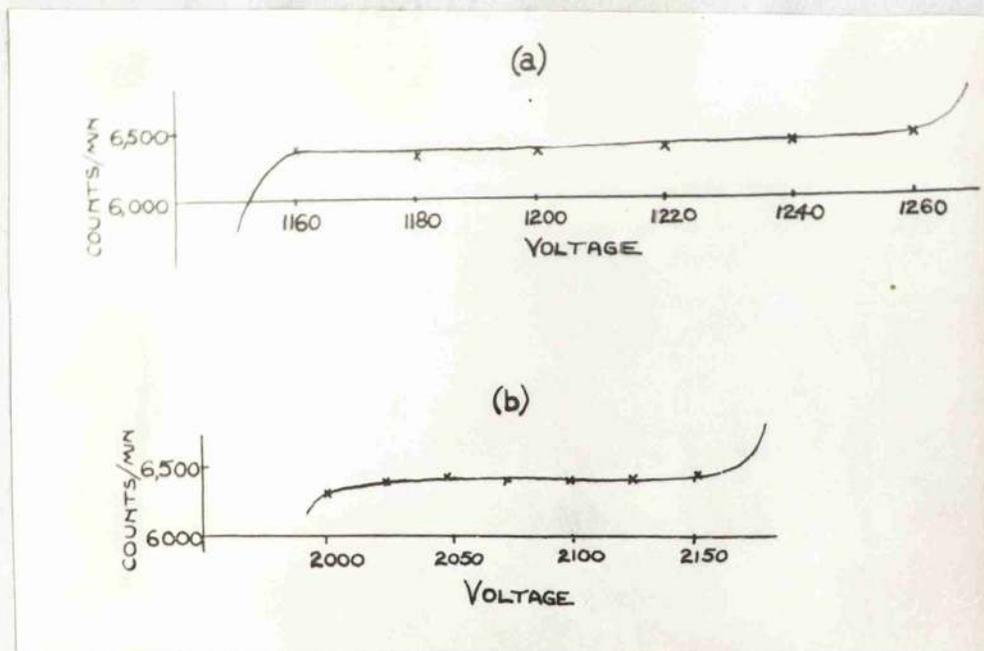


Fig 33. Plateaux obtained with source of ^{60}Co deposited on aluminium.

(a) Geiger operation of Counter.

(b) Proportional operation of Counter.

Geiger : 15cm mercury of argon + 1.5cm mercury of alcohol.

The corresponding counting rates of 3820 per minute and 3890 per minute gives a greater (< 2 per cent) counting rate for Geiger operation, but again this is within experimental error.

(d) RaD + E + F source on silver backing.

None of the above results gives as large a difference in counting rates as could be expected from calculations based on known data on secondary electron yields. As a check that radioactive sources do not give any appreciable yield of secondary electrons, use was made of the established fact that the secondary electron yield is of the order of 10 times greater for α - particles than for β - particles. In the absence of a pure α -particle source, a source consisting of an equilibrium mixture of RaD + RaE + RaF was used.

The experiment, complicated by the absence of a pure source of α - particles, consists of three parts.

(1) The proportional counter plateau shown in Fig 34(a) is that obtained with α - particles of RaF alone.

Assurance that only α - particles, and not single electrons,

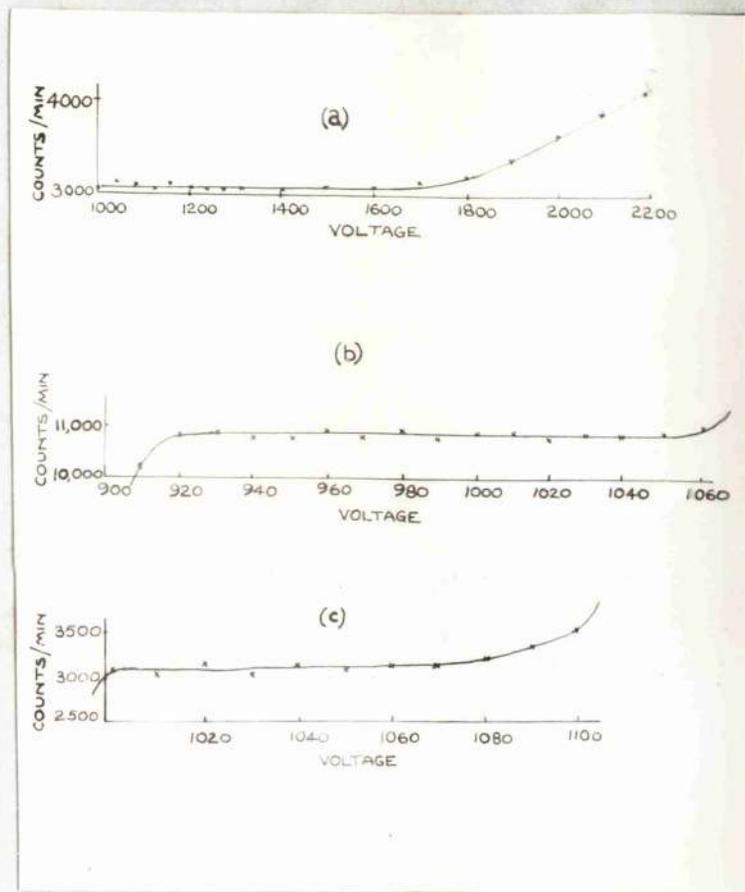


Fig 34. Plateaux obtained with a source of RaD + E + F on silver backing material.

were being counted was obtained from the fact that an external γ -ray source (^{60}Co) made no change in the counting rate. Beyond the part B of the curve single electrons began to be counted. Counting rate due to α -particles alone is 3100 per minute.

(2) In fig 34(b) is shown the Geiger plateau obtained with the same source. This gives a counting rate for all the particles of RaD + E + F, plus possible secondary electrons as 10,900 per minute.

(3) By repeating this experiment (Geiger operation), with the source covered by a thin (0.001 in) aluminium foil, to absorb the α -particles of RaF and the soft β -particles and conversion electrons of RaD, only the β -particles of RaE were detected, the counting rate being 3,100 per minute.

The fact that the three sources are in equilibrium, and the counting rates due to RaE and RaF are identical, enables calculation of the counting rate due to RaD. If the decay of RaD were by the emission of a single β -particle, the counting rate would also be 3,100 per minute. Since, however, in view of the very high conversion of the succeeding γ -ray, each disintegration gives rise to two particles, the counting rate is increased by a factor of almost exactly $\frac{1}{2}$ i.e. to 4,650

counts per minute.

The contribution to counting rate by secondary electrons is then given by (total count rate from experiment (2)) - (sum of separate count rates of RaD, RaE, RaF) i.e.

$$10,900 - (4650 + 3100 + 3100) \text{ per minute} \\ = 50 \text{ counts per minute.}$$

which is certainly within the limits of experimental error.

8. CONCLUSIONS ON SECONDARY ELECTRON EMISSION.

From all of the experimental results of section 7, and particularly those obtained with α -particles, it is fairly evident that the practice of depositing the source on some sort of backing material, does not introduce any appreciable error due to emission of secondary electrons. The secondary electron emission yield from the source itself, or from the source support, is not as marked (if indeed it is present at all) as could be expected from calculation from known data. This is no doubt due to the nature of the surfaces. It has been found (McKay, 1948) that the emission yield is extremely dependent upon the obtaining of a smooth surface from

which emission occurs. In the experiments described, therefore, there is no doubt almost complete absorption of the low energy secondary electrons in the source support or in the source itself. Nevertheless the sources used (between ~ 20 and $50 \mu\text{gms/cm}^2$) are fairly representative of the sources used in normal investigations on radioactivity.

The established absence of secondary electrons, although generally reassuring for work on low energy β -spectroscopy, removes the proposed possibility of explaining the results on back - scattering of electrons obtained by Suzor and Charpak (1952) and Christian et al (1952). The results of the former, so different from those obtained by the author, remain unexplained. However the work of Christian et al was done using a source positioned at the end wall of a Geiger counter. In view of the result shown by the author that such an arrangement is subject to low sensitivity of detection, their figures must be regarded with some caution.

9. GENERAL DISCUSSION OF THE AUTHOR'S RESULTS ON BACK - SCATTERING; SECONDARY ELECTRON EMISSION, AND ON THEIR APPLICATION TO β -RAY SPECTROSCOPY

The results obtained on back - scattering are

re - assuring in view of the important part played by scattering in β -ray spectroscopy. In many recent designs of spectrometer the useful angle of emission has been greatly increased, but no systematic study of scattering in these cases has been made. Much of the earlier work on spectroscopy was done with magnetic semi - circular instruments, in which the emission was relatively restricted, say, into $\pm 6^\circ$ around the perpendicular to the source. Even in these circumstances absorption and scattering led to spurious results in many instances. It has been shown in recent years that the use of very thin sources on thin supporting foils (generally of organic material rendered conducting in some suitable way) leads to correct results down to rather low particle energies. Here both source and support have a thickness equivalent to some $10 \mu\text{gm}/\text{cm}^2$. The introduction of spectrometers of radically different design, for example the proportional tube spectrometer, has meant that the solid angle of useful emission is greatly increased. Values of 2π and 4π have been employed. In these very different circumstances, the effects of reflection require still more thorough examination.

It has been shown by the author that thin sources

mounted on thin supports, preferably of low Z - value materials, are very satisfactory for such instruments. Even when thick supports of materials of low or of medium Z - value are employed, the total amount of back - scattering is not excessively large, and as regards spectrometry, its effect is evident, mainly in a certain amount of "tailing" on the low energy side of homogeneous electron lines. There is no evidence of any appreciable amount of production of very low - energy secondary electrons. This means that good spectroscopy can be done with thin sources on thick supports of plastic or other light material e.g. aluminium. The problem of employing sources of intermediate thickness, from 0.1 to 1.0 mg/cm^2 , say, arises in the investigation of substances of low specific activity or of radio - elements of very long life. The author's results show that distortion of the spectrum in such cases is not very serious, particularly since the percentage back - scattering decreases with decreasing energy of the particles and many such sources emit β -rays of relatively low energy. When the source consists of materials of high Z - value the effect of scattering in the source itself must be carefully considered owing to the increase with increasing Z - value

In work on absolute source strength the value of the back - scattering coefficient is of primary importance. Here again mounting of the source on a thick light support, for example on the aluminium cylinder acting as the cathode of the proportional tube, does not lead to intense back - scattering, particularly when the particles are of low average energy. less than 100keV, say. This technique has been employed in important experiments on the half life of natural radio elements, some of great value in dating geological specimens. As an example of the application of the author's results there is the work on rubidium, due to Curran, Dixon, and Wilson (1952). The value for the coefficient of back - scattering (7.5 per cent) assumed in their estimation ($\tau = 6.15 \times 10^{10}$ years) of the half life of ^{87}Rb has been criticised by Charpak and Suzor (1951) using their own value of 33 per cent to give $\tau = 7.6 \times 10^{10}$ years. From the author's results a value of 8 per cent for the back - scattering corresponds to the mean energy of the β -rays of ^{87}Rb . This value yields a half - life τ of 6.16×10^{10} years, in good agreement with the original published result of Curran, Dixon, and Wilson. Independent measurements on the half life of ^{87}Rb have been made by MacGregor and Weidenbeck (1952) who corrected

for back - scattering by extrapolating results obtained with decreasing thicknesses of backing material, and by Lewis (1952) whose method is free from uncertainties introduced by back - scattering. Their values of $\tau = 6.13 \times 10^{10}$ years and 5.90×10^{10} years respectively, would seem to indicate preference for the author's results on back - scattering rather than those of Suzor and Charpak. The result obtained recently by Flinta and Eklund (1954) is likewise consistent with this view.

145.

R E F E R E N C E S.

- Angus, J., Cockroft, A. L. and Curran, S. C. 1949
Phil. Mag. 40,522
- Backus, J. 1945 Phys. Rev. 68,59.
- Balfour, J. G. 1954 J. Sci. Instrum. 31,395.
- Bannerman, R. C. and Curran, S. C. 1952 Phys. Rev. 85,134.
- Bannerman, R. C., Lewis, G. M. and Curran, S. C. 1951
Phil. Mag. 42,1097.
- Black, D. H. 1925 Proc. Roy. Soc. A109,166.
- Bothe, W. and Geiger, H. 1925 Z. Phys. 32,639.
- Bothe, W. and Maier - Leibnitz, H. 1936a Z Phys. 102,143.
- Bothe, W. and Maier - Leibnitz, H. 1936b Z Phys. 104,604.
- Bothe, W. and Von Baeyer, H. J. 1935 Z Phys. 95,417.
- Bowe, J. C. and Axel, P. 1952 Phys. Rev. 85,858.
- Braden, C. H., Slack, L. and Shull, F. B. 1949
Phys. Rev. 75,1964.
- Bramson, S. 1930 Z. Phys. 66,721.
- Brownell, G. L. 1952 Nucleonics 10,30.
- Burt, B. P. 1950 Conference on absolute β - counting,
Nuclear Science Series, Prelim. Report
No. 8. (Washington : National Research
Council, 1950).
- Butler, S. T. 1950 Proc. Phys. Soc. A63,599.

- Butt, D. K. and Brodie, W. D. 1951 Proc. Phys. Soc.
A64,791.
- Charpak, G. and Suzor, F. 1951 C. R. Acad. Sci. (Paris)
233,1356.
- Christian, D., Dunning, W. W. and Martin, D. S. 1952
Nucleonics, 10,41.
- Cockroft, A. L. and Curran, S. C. 1951 Rev. Sci. Instrum.
22,37.
- Cook, C. S., Langer, L. M. and Price, H. C. 1948 Phys.
Rev. 73,1395.
- Cooper, E. P. and Rogers, F. T. 1950 Phys. Rev. 77,402.
- Cranberg, L. 1950 Phys. Rev. 77,155.
- Curran, S. C., Angus, J. and Cockroft, A. L. 1948
Nature 162,302.
- Curran, S. C., Angus, J. and Cockroft, A. L. 1949a
Phil. Mag. 40,53.
- Curran, S. C., Angus, J. and Cockroft, A. L. 1949b
Phil. Mag. 40,36.
- Curran, S. C., Cockroft, A. L. and Inch, G. M. 1950
Proc. Phys. Soc. A63,845.
- Curran, S. C. and Craggs, J. D. 1949 Counting Tubes :
Theory and Applications.
- Curran, S. C., Dixon, D. and Wilson, H. W. 1952
Phil. Mag. 43,82.
- Danysz, M. 1911 Le Radium 9,1.

- Deutsch, M. 1952 Physica 12,1037.
- Droste, G. F. Van. 1933 Z. Phys. 84,17.
- Dunworth, J. V. 1940 Rev. Sci. Instrum. 11,167.
- Ellis, J. W. 1922. Proc. Camb. Phil. Soc. 21,125.
- Ewan, G. T. and Ross, M. A. S. 1952 Nature 170,760.
- Fan, C. Y. 1952 Phys. Rev. 87,252.
- Feather, N. 1949 Nucleonics 7,22.
- Feather, N., Kyles, J. and Pringle, R. W. 1948 Proc.
Roy. Soc. A61,466.
- Feenberg, E. and Trigg, G. 1950 Rev. Mod. Phys.
22,399.
- Feingold, A. M. 1951 Rev. Mod. Phys. 23,10.
- Flinta, J. and Eklund, S. 1954 Ark. Fys. 7,401.
- Frilley, M., Gokhale, B. G. and Valdares, M. 1951
C. R. Acad. Sci. (Paris) 232,50 and 232,151.
- Goudsmit, S. and Sanderson, J. L. 1940 Phys. Rev. 57,24.
- Gray, J. A. 1932 Nature 130,738.
- Hebb, H. M. 1938 Physica 5,701.
- Hofstadter, R. and McIntyre, J. A. 1950 Phys. Rev.
78,619.
- Hornyak, W. F., Lauritsen, T. and Rasmussen, V. K. 1949
Phys. Rev. 76,731.
- Insch, G. M., Balfour, J. G. and Curran, S. C. 1952
Phys. Rev. 85,805.

- 192.
- Insch, G. M. and Curran, S. C. 1951 Phil. Mag. 42,892.
- Ivanenko, D. and Lebedev, v, 1950 J. Exp. Theor. Phys.
U.S.S.R. 20,91.
- Jaffe, A. A. and Cohen, S. G. 1952 Phys. Rev. 86,1041.
- Jaffe, A. A. and Cohen, S. G. 1953 Phys. Rev. 89,454.
- Jonker, J. L. H. 1951 Philips Res. Reports 6,372.
- Kinsey, B. 1948 Canad. J. Res. A26,404.
- Kobayashi, Y. 1953 J. Phys. Soc. Japan 8,440.
- Konopinski, E. J. 1943 Rev. Mod. Phys. 15,209.
- Langer, L. M., Motz, J. W. and Price, H. C. 1950
Phys. Rev. 77,798.
- Lee, D. D. and Libby, W. F. 1939 Phys. Rev. 55,252.
- Lewis, G. M. 1952 Phil. Mag. 43,1070.
- Lewis, H. W. 1950 Phys. Rev. 78,526.
- MacGregor, M. H. and Weidenbeck, M. L. 1952 Phys. Rev.
86,420.
- Maier - Leibnitz, H. 1936 Z. Phys. 104,604.
- Mandeville, C. E. and Shapiro, E. 1949 Phys. Rev. 75,897.
- Meitner, L. 1922 Z. Phys. 11,35.
- Mitchell, A. C. G. 1948 Rev. Mod. Phys. 20,296.
- Mitchell, A. C. G., Langer, L. M. and McDaniel, P. W.
1940 Phys. Rev. 57,1107.
- McKay, K. G. 1948 Advances in Electronics, Vol. 1.
- Molière, G, 1948 Z. Naturforsch 3a,78.

- Müller, H. O. 1937 Z. Phys. 104,475.
- Norling, F. 1938 Z. Phys. 111,158.
- Nuclear Data 1950. National Bureau of Standards
Circular 499.
- Purcell, E. M. 1938 Phys. Rev. 54,818.
- Richardson, H. O. W. and Leigh - Smith, A. 1937 Proc.
Roy. Soc. A160,454.
- Rose, M. E. et al. 1951 Phys. Rev. 83,79.
- Rutherford, E., Chadwick, J. and Ellis, C. D. 1930
Radiations from Radioactive Substances.
- Rudberg, E. 1936 Phys. Rev. 50,138.
- Schwartz, H. M. 1952 Phys. Rev. 86,195.
- Schwartz, H. M. and Edwards, R. R. 1951 J. Chem. Phys.
19,385.
- Snyder, H. S. and Scott, W. T. 1949 Phys. Rev. 76,220.
- Spangenberg, K. R. 1948 Vacuum Tubes.
- Stahel, E. 1935 Helv. Phys. Acta. 8,651.
- Suzor, F. and Charpak, G. 1952 J. Phys. Radium 13,1.
- Tsien, S. T. 1946 Phys. Rev. 69,38.
- Trump, J. G. and Van de Graaf. R. J. 1949 Phys. Rev.
75,44.
- Wang, M. C. and Guth, E. 1951 Phys. Rev. 84,1092.
- Weidenbeck, M. L. and Chu, K. Y. 1947 Phys. Rev. 72,1171.
- Weymouth, J. W. 1951 Phys. Rev. 84,766.

- Williams, E. J. 1939 Proc. Roy. Soc. A169,531.
- Wu, C. S., Boehm, F. and Nagel, E. 1953 Phys. Rev. 91,319.
- Yaffe, L. 1950 Conference on Absolute β -Counting.
Nuclear Science Series, Prelim. Report No. 8.
(Washington, National Research Council).
- Yaffe, L. and Justus, K. M. 1949 J. Chem. Soc. 5,341.