NEW METHODS OF BETA AND GAMMA RAY SPECTROSCOPY

by

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PREFACE

In this thesis I have described a recent method of measuring the energies of beta and gamma radiations, using proportional tube spectrometers. The introduction contains a short account of the theory of beta decay with an indication of the methods of comparing this theory with the experimental data. This part has been largely drawn from Moon (Artificial Radioactivity).

In Part I the experimental techniques which have been employed for investigation of beta ray spectra are described. After a general discussion of these methods the use of the proportional tube spectrometer is considered, and its advantages noted. In this section I have drawn freely from Frisch (Progress in Nuclear Physics) and Curran and Craggs (Counting Tubes).

In Parts II and III I have described the experimental work performed and discussed the significance of the results obtained. It should be noted (see end of Preface) that the results of all these investigations have been published. While the matter here is original, much of the work has been carried out in collaboration, and only the following portions of this thesis describe work which has been carried out solely by myself:-

(a) all the work described in Part II, Chapter I;

(b) the construction of the electrometer valve circuit described in Part II, Chapter II;

(c) the preparation of the radioactive gas ($H_2 S^{35}$) and the investigation of its properties described in Part III, Chapter I;

(d) all the work described in Part III, Chapter II.

While all the experimental work was carried out by myself, frequent discussion with Dr. S.C. Curran considerably influenced my mode of attack on the problem.

I must thank Professor P.I. Dee and Dr. S.C. Curran for their sustained interest and encouragement throughout the work, and also Mr. D.L. Pursey for helpful discussion on several points of the theory. I also wish to acknowledge the invaluable help of the Department of Scientific and Industrial Research who awarded me a grant for my first two years of study.

G.M.I.

Publications

1). "Calibration of Proportional Counters", Insch (1950).

2). "The mean energy expenditure per ion pair produced by slow electrons in various gases", Curran, Cockroft and Insch (1950a).

3). "Beta and Gamma ray spectroscopy with Proportional Counters in Magnetic fields",
Curran, Cockroft and Insch (1950b).

4). "The beta spectrum of S^{35}", Cockroft and Insch (1949).

5). "The beta spectrum of H^{3}", Insch and Curran (1951).

6). "The beta spectrum of RaD", Insch, Balfour and Curran (in the press).

CONTENTS

INTRODUCTION

Fermi Theory of Beta Decay

One of the most important forms of spontaneous disintegration of a nucleus is that which results in the emission of a beta-particle. The study of this process is important, both for investigation of the energy levels of atomic nuclei, and for examination of the nuclear interaction responsible for the emission. This transformation is characterised by a continuous distribution in the energy of the emitted particles, first demonstrated by Chadwick (1914), and explained by the hypothesis of Pauli that the beta particle was responsible for removing only a part of the energy, while an undetected particle of half integral spin, known as the neutrino, carried away the remaining energy. This hypothesis, together with that of Heisenberg that the neutron and proton were two states of the same particle, formed a basis for a theory of beta decay formulated by Fermi. Although some modifications have been made to this theory, it still forms the foundation for theoretical examination of the experimental results obtained in beta-ray spectroscopy.

Fermi assumed that the process was, in some ways, analgous to the transition of an atom from one excited state to another with the emission of a photon. In the case of an excited atom, the magnitude of the

probability per unit time of emission of a photon depends on the initial and final states of the atom, i.e. it depends on whether or not the selection rules are obeyed, and distinguishes between "allowed" and "forbidden" transitions. In this case the process is described in terms of a "trilinear" interaction. This indicates that the energy due to the interaction is proportional to the product of three fields corresponding to the three participating states

$$e \rightarrow e^1 + h\nu$$

Beta decay, which involves four participating states, is described as a "quadrilinear" interaction

$$N \rightarrow \rho + e + \nu$$

Because of this difference, and the fact that the beta particle is affected by the Coulomb field of the nucleus, Fermi's equation for the probability per unit time of emission of a beta particle is more complicated. However, the distinction between "allowed" and "forbidden" transitions is still present.

Fermi's equation for the probability that in a time dt a single nucleus shall ~~limit~~ emit a beta particle of total energy between W and W + dW is

$$N[W]\,dW.dt. = \{G^2/2\pi^3\}\,|X|^2\,F[Z,W]\,p\,W(W_o-W)^2\,dW.dt.$$

$$\text{or } N[p]\,dp.dt. = \{G^2/2\pi^3\}\,|X|^2\,F[Z,W]\,p^2(W_o-W)^2\,dp.dt.$$

This equation only holds when the mass of the neutrino is negligible in comparison with that of the electron.

p is the momentum of the electron measured in m_0c units corresponding to a total energy W measured in m_0c^2 units. $p\,W(W_0-W)^2$ is a statistical factor representing the probability per unit energy range of sharing the total energy W_0 between the electron and the neutrino in such a way that the electron has an energy W and the neutrino an energy (W_0-W). $F(Z,W)$ is a function giving the effect of the Coulomb field of the nucleus on the energy distribution for an allowed transition. It emphasises fast positrons and slow electrons. The factor $|X|^2$ contains nuclear matrix elements and, in the case of forbidden transitions, energy dependent correction factors. The strength of the forces responsible for the transition is expressed by the factor $G^2/2\pi^3$.

It is difficult to decide which type of interaction between the heavy particle and the beta particle and neutrino gives a correct description. There are five possible types of interaction:-

Scalar, Polar vector, Axial vector, Tensor, and Pseudo Scalar.

There is no theoretical reason to prevent the interaction from being a superposition of two of these types. The polar vector interaction was originally assumed by Fermi, by analogy with photon emission. His selection rules put rather rigorous restrictions on the spin changes permitted in beta

transitions. Gamow and Tellar have suggested the Tensor interaction, and at present this seems best for explanation of the experimental results. Their selection rules are less rigorous and permit certain spin changes not allowed by Fermi selection rules.

Relationship between half life and transition energy

To a first approximation it is found that the half life of a beta disintegration is inversely proportional to the fifth power of the total energy

$$t = K/W_o^5.$$

An examination of the values of the constant K, for beta particles from various nuclei, shows that beta disintegrations separate into groups having approximately the same value of K. These groups are defined as "allowed", "first forbidden" and so on with increasing values of K. Forbidden transitions are those which take place with much reduced, but not zero,probability and correspond to a violation of the selection rules. A physical picture of the angular momentum rules can be obtained by considering that, in the Fermi case, the electron and the neutrino can leave the nucleus with opposite spins, while in the Gamow Tellar case they are permitted to leave with parallel spins. $|X|^2$ decreases when the angular momentum change exceeds the allowed amount, and the much smaller value of $|X|^2$ corresponds to the relatively small

probability that the particle will leave a nucleus and carry with it orbital angular momentum. Each unit of orbital angular momentum reduces the probability by a factor of 100. Further information on spin changes can be found in the excellent review of the subject by Konspinski (1943).

A more refined form of classification of beta disintegrations which depends on Fermi theory is achieved by forming the product ft where f is given by

$$f\ (Z,W_0) \ = \int_1^{W_0} F(Z,W)\, p\, W(W_0-W)^2\, dW.$$

The total probability per unit time that a beta ray of any energy shall be emitted (the decay constant λ) can be expressed as

$$\lambda = K,f = \frac{0.693}{t}$$

$$\text{Hence } ft = 0.693/K,.$$

According to Fermi theory all allowed transitions should have the same values of ft. First forbidden transitions should have values of ft 100 times greater, and so on. In the Gamow-Tellar interaction the general behaviour is the same, but the numerical values will be a little different. Unfortunately, a comparison of the experimental values with these predictions is not sufficient to ascertain which form of the theory is correct, because G^2 is involved in both forms and is theoretically undetermined. These predictions are not very

precisely fulfilled and should not be interpreted too rigidly. However, they form a useful guide.

Comparison of experimental results with theory

a) Allowed transitions

A very useful approximation for the comparison of experimental results with theoretical predictions lies in the application of the Kurie plot. Since $|X|^2$ is a constant for allowed transitions we can write

$$N[W] = C^2 F(Z,W) p W (W_0-W)^2 \quad \text{where } C^2 \text{ is a constant.}$$

$$\therefore \quad (N[W] / p W F(Z,W))^{\frac{1}{2}} = C (W_0-W).$$

Hence if $(N[W] / p W F(Z,W))^{\frac{1}{2}}$ is plotted against W a straight line, known as the Kurie plot, is obtained which intersects the energy axis at W_0. This is extremely useful for comparison of results with theory as shall be seen later.

b) Forbidden transitions.

We have stated that, in the case of forbidden transitions, $|X|^2$ contains energy dependent correction factors. The matrix element denoted by X can be composed of a number of terms. Each of these can have its own effect on the shape of the distribution and can also have its own selection rules. The relative magnitudes of these terms are

not accurately predicted by theory. However, the introduction of suitable correction factors allows us to compare the theoretical predictions with experimental results.

The validity of Fermi theory must be judged by its ability to explain the experimental results. Measurements on the shape of "allowed" spectra cannot distinguish between the five types of interaction, but in the case of "forbidden" spectra, since the matrix element X is normally energy-dependent and the nature of this energy-dependence is related to the particular interaction form assumed, it may be possible to choose a form of interaction by accurate measurements on spectral shapes. Measurements on "allowed" spectra, particularly in the low energy region, are of importance, mainly because Fermi theory has not been thoroughly tested for the slower electrons, due to experimental limitations. We would emphasise the importance of the disintegration of tritium, since the measurement of the spectrum of the beta particles from this isotope plays the same role in the theory of beta decay as the measurement of the hydrogen spectrum in quantum theory.

PART I

CHAPTER I

Experimental Techniques Employed in the Investigation of Beta Spectra.

1) Absorption

One of the most straightforward methods of examining the radiations from a radioactive source is the study of the absorption curve obtained when the rays or particles are passed through matter. In the case of beta emission, it is possible to obtain much useful information by careful measurements on the absorption of the electrons in light materials, such as aluminium. Feather (1938 a and b) employed an arrangement of counter and absorbing screens. He used sources of several sq. cm. area, comparatively weak in intensity, held at a few cm. from a thin window counter. The beta rays were passed through absorption foils placed close to the source of radiation. Feather proved, using the beta radiation of RaE as a standard of comparison, that the maximum energy and the approximate shape of the energy spectrum of the beta particles under investigation could be obtained. The technique is suitable for examination of weak sources or sources of finite thickness, a satisfactory method for correcting the maximum energy of the beta particles for the thickness of the source

being employed. The absorption curve can yield information on conversion electrons superimposed on a continuous beta ray spectrum. These are often recorded as ~~knits~~ kinks on the curve, but careful measurement of the change of slope is essential for good results.

Measurements on the radiation from Na^{24} (Feather and Dunworth 1938) and Uranium Z (Feather and Bretscher 1938) demonstrate the usefulness of the absorption method.

2) Magnetic Resolution

However, the passage of an electron through matter is accompanied by continual loss of velocity and small random changes of direction caused by collision with atoms or molecules. This property causes a homogeneous beam of electrons, initially well colimated and of uniform energy, to lose homogeneity of energy and direction in passage through material. It is, therefore, preferable to measure electron energies in some manner which employs a path through an evacuated chamber. This is achieved in magnetic spectrometry, where the beta rays from a source are resolved in a magnetic field perpendicular to the path of the particles. This type of resolution has, in the past, yielded the most accurate results in the examination of beta ray emission, especially over the upper energy range of the spectrum. The manner in which the beta particles are brought to a point or

line focus varies with the different instruments employed, and we shall now consider the types of magnetic spectrometers which have so far been used.

a) Semicircular focusing spectrometer.

This type of focusing is the oldest used in magnetic beta ray spectroscopy, being introduced in 1913 by Danysz. It involves the selection of a narrow beam from the particles leaving a short line source in directions nearly perpendicular to a uniform magnetic field. All particles having a given momentum come to a line focus which has a sharp outer edge. The angular width of the beam is defined by a number of apertures and only the central ray completes a semicircle before arriving at the focus. The other rays which lie in the central plane will traverse a path either larger or smaller than a semicircle, while these deviating from this plane travel along a flat helix. The method is characterised by its high resolving power. It is limited in that the focusing action is in one plane only, which results in a reduced intensity when compared with the other focusing methods. It is usually employed when it is important to obtain absolute values of the momentum, since only a uniform field can be accurately measured. If the calibration with particles of known momentum is available other instruments are often preferred.

b) Solenoid Spectrometer

This instrument, first used by Tricker in 1924, employs helical focusing. The source is in the form of a small disc, placed perpendicular to a uniform magnetic field. The beta particle, leaving the source at a small angle to the field direction, come to an axial focus after describing a complete turn of an elongated helix. The method was developed by Witcher (1941) who focused the electrons on a ring perpendicular to the axis. Recent work by Frankel (1948) and Persico (1948) shows that the optimum working conditions of this instrument are only possible for a certain mean angle of emission. Working under these conditions the method compares favourably with the performance of the semicircular focusing instrument, but difficulties due to the amount of copper required, and the fact that the counter and the source are in a high field region, have tended to make this instrument unpopular.

c) Magnetic lens Spectrometer

The method of focusing employed in this type of spectrometer is essentially the same as that used in the Solenoid case, but the magnetic field is more limited in space. The evacuated chamber has a source at one end and a Geiger counter at the other, with an annular ring baffle between them to select the electron trajectories. The instrument is characterised by good efficiency for electron

collection, and localisation of the magnetic field which permits both the source and the Geiger counter to be in relatively field free regions.

This instrument has advantages over the semicircular focusing instrument, in that the efficiency for collection of electrons is relatively high, the source material can be distributed over the surfaces of thin films covering an area larger than that generally used in the latter instrument, and the source can be located relatively far from any scattering material, except that of the source holder itself. However, the instrument does not utilise the central part of the beam in the focusing, since the resolving power is proportional to the diameter of the central shutter. In addition, it is normally used with a poorer resolving power than that obtained in the semicircular instrument.

d) Double focusing spectrometer

This form of device for focusing electrons was originally proposed by Siegbahn and Swartholm (1946). They emphasise that the advantages possessed by the electron lens type of spectrometer are counter-balanced by the fact that the central screening is detrimental to the focusing on account of spherical aberration. In the focusing of the semicircular method this difficulty is avoided, but the two directional focusing of the electron lens

is lost. They used an inhomogeneous magnetic field of cylindrical symmetry with an electron path perpendicular to the lines of force giving a two dimensional focusing. Thus they incorporated several of the useful properties of the semicircular and lens methods. The momentum-current relationship is non-linear, however, and further disadvantages lie in the necessity to profile the pole pieces for correct field distribution and the fact that both the source and counter are in a high field region. However, for most purposes these disadvantages are not serious, and this method of focusing represents a real advance over the magnetic lens spectrometer in resolution.

e) Radial inhomogeneous field semicircular focusing spectrometer.

A spectrometer of this type is used by Langer and Cook (1948). They have an annular magnet and employ a mean radius of curvature of 40 cm. This makes it possible to use larger source areas than normally employed in semicircular focusing instruments without sacrificing the good focusing action.

f) Prolate spheroidal magnetic field spectrometer

The possibility of building an axially symmetric spectrometer of wide angle, using a prolate spheroidal field, has been discussed by

Richardson (1949). His analysis shows that such an instrument should have a resolving power comparable with that attainable in the case of semicircular focusing in a uniform field, with the added advantage of a wide angle. The good action of this type of instrument has been reported by Braid and Richardson (1951).

Performances of magnetic spectrometers.

The utility of a beta ray spectrometer will generally depend upon the following three criteria:

1) The range in energy over which the instrument can be used,
2) the resolution,
3) the transmission.

Concerning (1), in the case of magnetic spectrometers there is no real difficulty in achieving accurate energy measurements on high energy electrons, but, as shall be discussed later, measurements on low energy electrons present a formidable problem.

Concerning (2) and (3), the transmission will be defined as the percentage of all electrons within a given momentum range emitted by the source which is recorded at the focus, while the resolution is measured by expressing as a percentage difference in momentum the half width of the curve for monokinetic electrons. Both vary with the source

area and with the type of focusing employed. However, it is reasonable to say that magnetic spectrometers are normally used with a resolution of the order of 1 or 2 percent, a transmission of the order of 1 percent or less, and a source area of the order of a sq. cm. These values will be sufficient for comparison with the performance of the proportional tube spectrometer to be discussed later.

Detection of the particles

The two main methods of detecting the electrons after resolution in the magnetic field are the photographic plate technique and the use of Geiger counters. The photographic plate technique, normally used only for monokinetic electrons, has the advantage of requiring only a single exposure. However, the plate is non-linear for high and low intensities, making qualitative work difficult, and is also relatively insensitive. Thus, if it is not possible to obtain strong sources of high specific activity, the use of this technique presents difficulty. However, it has been used, notably by Ellis, Helmholtz (1941) and Cork (1949). Ellis used it in his well known investigations of the spectra of the natural radioactive bodies with such high accuracy that his results have since been used as standards for the calibration of magnetic

spectrometers - the accuracy of his best work (Ellis and Skinner 1924) is quoted as 1 part in 500. Cork overcame the difficulty of source strength by using permanent Alnico magnets and making exposures for as long as several weeks. Recent developments in this technique, enabling the detection of single electrons (Barriman 1948), now provide great advantages. However, the plate must be subjected to tedious microscopic examination after exposure.

The Geiger-Muller counting tube present a detector of much greater sensitivity, and enables the relative intensities of lines or of different parts of a continuous spectrum to be accurately measured. The line focus experienced in the semicircular type of spectrometer is ideal for use with a cylindrical counter, the entrance for the particles being controlled by a slit parallel to the axis of the tube. In the case of the lens spectrometer, bell shaped counters with circular apertures have been used with success (Copp and Greenberg 1943). Care must be taken to ensure that the pressure of the gas in the counter is sufficient for the detection of the most energetic particles. In addition the counter window must be thin enough to admit as many of the particles as possible.

The Geiger counter has two main disadvantages. First, due to the finite width of the slit, the resolution of the instrument is lowered, and secondly the counter can only record the integral of a small part of the spectrum at one time necessitating the varying of the magnetic field, and the recording of a large number of observations to complete the whole spectrum.

Limitations of magnetic spectrometers.

Despite the many and ingenious improvements which have been made in the field of magnetic spectrometry in recent years, measurements on the low energy regions of beta spectra are still subject to considerable possible error. The beta particles have to penetrate the thin window which separates the evacuated chamber from the detecting tube. In doing so the very low energy electrons invariably suffer considerable absorption, even when extremely thin organic films are used for the window material. Consequently, it is reasonable to doubt the accuracy of any measurements on electrons of energy less than about 20 KeV, although many such electrons do penetrate into the counter volume. Recently, Langer and Cook (1948) have attempted to accelerate the low energy particles by application of a difference of potential between the detecting slit and the Geiger counter, but without much success so far. Similar work has been performed by Backus (1945)

and Butt (1950). Using his post-focusing electron accelerator, Butt found that electrons resulting from secondary emission were detected in the counter, in the absence of a source, when the accelerating voltage exceeded a certain value. However, despite this limitation, the method is useful and a small percentage of electrons having energies as low as 1 or 2 KeV can be detected in the counter.

If the absorption in the counter window were the only reason for distortion of spectra in this energy region, all might be well, since correction can be made for this - at least to a certain extent. However, there are other problems. For example, some of the beta particles will lose energy in the source itself, while others may be completely absorbed. This is true, even for very thin sources. Again, even when the source is mounted on a thin film, there is a certain amount of "back scattering" which results in particles being attributed less energy than they had on leaving the source. In addition, the primary beta radiation expels secondary electrons of lower energy from the source. All these limitations help to "pile up" the low energy end of the spectrum, sometimes causing quite a violent distortion.

It becomes obvious from these considerations that, for ideal spectrometry work, large area sources of high specific activity should be used with spectrometers of high transmission. But here again

another limitation is obvious, since all magnetic spectrometers have a small transmission, resulting in the necessity for sources of a comparatively strong nature.

Finally, it should be mentioned that care must be taken to ensure that the source is not insulated electrically from the body of the spectrometer (see Braden et al. 1948). If it is, the spectrometer body and the source form, in effect, a small capacitance with a very high resistance leakage path between them. The beta emission developes a positive charge on the source, thus gradually establishing a considerable potential difference between the source and vacuum chamber, resulting in the deceleration of the electrons.

It should be emphasised that, despite these limitations, which apply mainly in the low energy region, magnetic spectrometry not only offers the most accurate means of measuring the majority of beta spectra, but also has, in the past, played a very valuable role in this field.

Before passing on to discuss some of the other methods which have been applied in beta ray spectroscopy it should be mentioned that Korsunsky et al. (1945 a and b) have examined inhomogeneous magnetic fields (a), and four pole arrangements (b) and have claimed increased transmission for a given resolving power.

3) Electrostatic spectrometers.

The use of electrostatic fields is not favourable for most beta spectra examinations because of the very high voltages which are required. However, Backus (1945) has employed a spectrometer using electrostatic focusing to investigate the lowest energy positrons and electrons from Cu^{64}, his measurements extending from 50 KeV down to about 10 KeV. He employed the 127^{o} focusing method of Hughes and Rojansky (1929) and examined spectra down to 4 KeV with a very thin windowed counter.

The upper energy limit of the beta spectrum of tritium, H^{3}, was measured by Watts and Williams (1946) using a very straightforward system. They applied accelerating or retarding potentials between a source, which was placed opposite a counter, and the counter itself, until the beta rays just failed to penetrate the thin window. Electrons accelerated from a hot filament were used in a similar fashion to determine the stopping power of the window. However, the method gave a result of about 11 KeV, which is not in agreement with the latest value (Curran, Angus and Cockroft 1949 b).

Very recent work by Hamilton and Gross (1950) has been the building of a hemispherical spectrometer where electrons move radially outward, encountering a retarding radial electric field in the region between two hemispherical grids. The integral beta spectrum is obtained by measuring the

current at a collector as a function of the retarding potential. The use of fine mesh grids and a large ratio of grid diameter to source size results in good momentum resolution. Secondary effects are minimised by using graphite electrode coating, an electron - absorbing backstop behind the source, and a large grid to act as a supressor for the collector. The method seems promising, despite a certain amount of back scattering at low energies and difficulties due to field emission, observed in the absence of a source.

4) Cloud chambers.

Due to collisions with gas molecules suffered by the electrons in their passage through the chamber, and their consequent changes of direction and loss of velocity, the instrument is not a very accurate one for beta spectra investigations, and should not be used when other methods are available.

5) Coincidence measurements.

Mention should be made of the measurement of complex beta spectra. When the nucleus is left in an excited state by beta emission, and this is followed by gamma radiation, coincidences between quanta and beta rays are detected up to a beta ray energy corresponding to the upper energy limit of the partial spectrum. Bothe and Maier-Leibnitz (1936) used a magnetic spectrograph to resolve the

beta particles from a radioactive source, and recorded coincidences between quanta and beta rays, as a function of the beta ray energy, with a gamma ray counter near the source. By this means two spectra superimposed upon each other can often be separated. The gamma ray intensity of the source can yield information on the intensity of the partial spectrum.

6) The proportional tube spectrometer.

As has already been mentioned, there are two major disadvantages associated with normal spectrometers when used for measurements in the low energy region of beta spectra:

a) uncertainties arise due to straggling and reflexion of electrons;

b) the absorption in the thin window of the Geiger tube detector makes correction of the data at low energies a somewhat empirical procedure.

These two disadvantages were overcome by Curran, Angus and Cockroft (1948) who introduced their radioactive source into a proportional counter in gaseous form, providing them with what is effectively an infinitely thin source. In conjunction with the counter they used a high gain amplifier of low noise level (Curran, Angus and

Cockroft 1949 a). It is suitable at this point to consider the performance of their spectrometer, since part of the work to be reported here deals with improvements which have been made to this performance.

a) <u>The range in energy over which the instrument can be used</u>.

In the case of the proportional tube spectrometer, the lower limit to the measurable energy corresponds to the production of one ion pair in the gas. However, the statistical fluctuation in the number of electrons released by very soft radiations is pronounced and statistical variation in the value of the gas amplification must also be considered. Because of this the resolving power is poor for measurements in the very low energy region (see next paragraph). On the high energy side, the maximum electron energy which can be faithfully recorded is limited by the path length in the gas. It is essential that a very high percentage (> 98 percent say) of the beta particles expend all their energy in the gas, and not in the wall of the vessel. Using large, high pressure counting tubes, electron energies up to ~ 200 KeV can be examined without violation of this condition (Angus, Cockroft and Curran 1949).

b) The resolution.

The pulses produced by homogeneous particles, expending their energy in the counter gas, vary in amplitude due to statistical fluctuations which occur in the number of ion pairs formed in the initial ionizing event, and in the value of the gas amplification.

Let us consider the case of a proportional tube spectrometer being used to measure the energy, E, of a homogeneous group of particles. When one of these particles produces m initial electrons in the counting volume, m independent avalanches will occur. Now, if the mean value of the initial ionization be $\bar{m}$, and its variance be w, it can be shown (see Wilkinson 1950) that the variance v_p in the total number of electrons formed in the multiple avalanche is

$$v_p = w \bar{A}^2 + v \bar{m}$$

$$= w \bar{A}^2 + (\bar{A}^2 - \bar{A}) \bar{m}$$

where A is the gas amplification, $\bar{A}$ its mean value and v its variance.

The relative variance is obtained by expressing this variance in terms of the square of the mean ionization produced

$$v_p / (\bar{m} \bar{A})^2 = w/\bar{m}^2 + 1/\bar{m} - 1/\bar{A} \bar{m}.$$

The relative variance in the number of initial electrons is given by $w/\bar{m}^2$.

For large values of A

$$\nu_p/(\bar{m}\,\bar{A})^2 = w/\bar{m}^2 + 1/\bar{m}.$$

If the initial ionization had a normal Poisson distribution, $w = \bar{m}$, and the contributions to the line width arising from fluctuations in the initial number of ion pairs formed and in the statistical variation of the gas amplification would be equal. However, when homogeneous particles expend all their energy in the gas $w < m$ (see Fano 1947). Therefore, at first sight it would appear that the gas amplification provides the greater contribution to the variance. However, the above result assumes that, for the electrons in the avalanche, the instantaneous probability of an ionizing collision is merely a function of X/p, where X is the field strength measured in volts/cm. and p is the gas pressure measured in mm. of Hg., and not on the previous experience of the electron concerned. In the high field region, close to the wire of a proportional tube, this assumption is not correct. The experience of one ionizing collision reduces the instantaneous probability of another such collision below the value obtained from consideration of the value of X/p alone at that position. This results in a smaller straggling than the value quoted

above. That the straggling is indeed less has been demonstrated by Hanna, Kirkwood and Pontecorvo (1949) and by Curran, Cockroft and Angus (1949). The latter authors detected single slow electrons liberated at the wall of a proportional tube by ultra violet quanta, and investigated the straggling within a single avalanche. They conclude that the variation introduced by the gas gain alone, and that caused by the fluctuation in the initial number of ion pairs released in the gas, were about equal in magnitude and less than predicted theoretically.

Unfortunately, the precise value of w is not known, and the value of $\bar{m}$ for particles of known energy is also subject to doubt. Because of this, the resolution of a proportional tube used for the measurement of particle energies is only approximately known. That the approximation is quite good has been demonstrated by the work of Curran, Cockroft and Angus (1949).

The value of $\bar{m}$ depends on the average energy lost per ion pair formed in the counter gas as a result of the absorption of the particles. Let this average energy be V

$$\bar{m} = E/V.$$

Since the measurements of the values of V for slow electrons in the various counter gases are not in agreement (see Part II, Chapter II) it was decided to determine these values as accurately as possible.

This would at least provide data for accurate assessment of $\bar{m}$.

Of course, a knowledge of the values of V in the various gases is important for other reasons and these are discussed by Wilkinson (1950).

c) Transmission.

For electrons released in disintegrations occuring in the counting volume, the transmission is almost 100 percent. This counting volume normally extends over $\sim$ 80 percent of the tube, but it is subject to an end-effect (Curran, Angus and Cockroft 1949 a) which limits the region of true proportionality. Such a tube allows comfortable examination of sources of total activity as low as 10^{-9} curies. A 4π acceptance angle has been achieved using thin sources mounted on thin nylon backing located at the centre of the counting volume. Such sources are held in position by probes which have the voltage appropriate to their position in the field, thus avoiding field distortion.

Application to the measurement of soft quanta.

The investigation of low energy gamma radiation (energies $<$ 200 KeV) is important for a number of reasons. For example, it seems very probable that low energy gamma rays are emitted in several disintegrations and that, in some cases, these radiations have so far escaped detection. Also, there are a number of transitions giving rise to low energy

radiations which do not appear to fit any established decay scheme. The decay of RaD is a good example of such a disintegration.

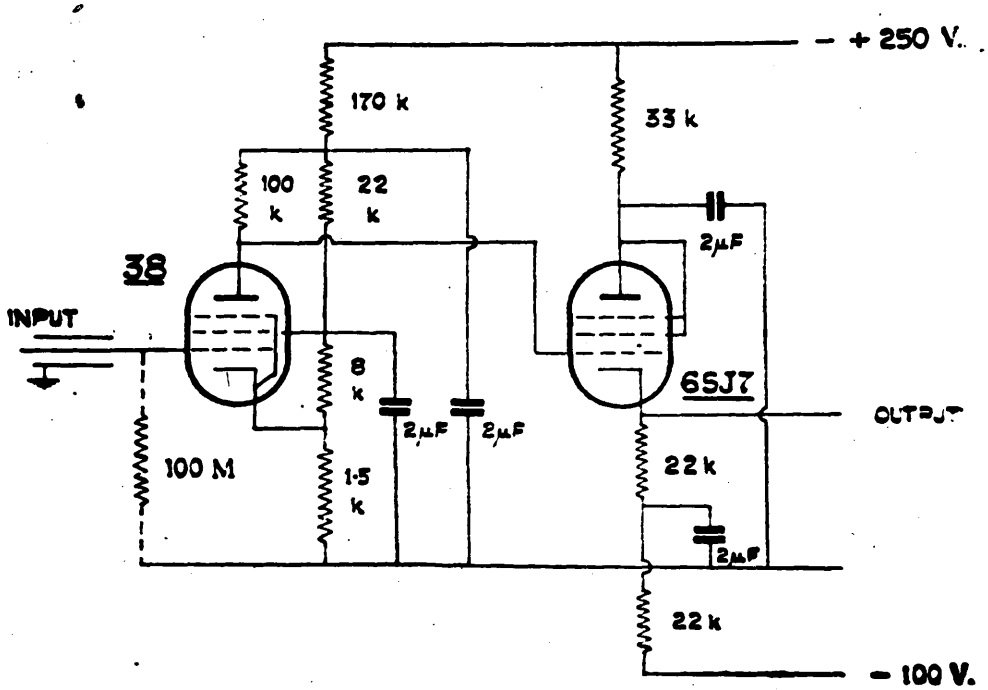
Energy measurements in the X-ray region are also important. The identification of a K-capture process involves the determination of the energy of the X-rays resulting from the capture of the extra-nuclear electron. This measurement is often achieved by the method of critical absorption which is satisfactory in the absence of other low energy radiation. However, coincidence measurements between the X-rays and gamma rays are often desired, and an improved method for the measurement and detection of the X-rays is indicated. The measurement of X-ray energies is also important in the investigation of an isomeric transition. The gamma ray, emitted in the transition, usually exhibits a high internal conversion coefficient, resulting in the liberation of characteristic X-radiation (see next chapter).

The proportional tube spectrometer is a powerful tool for examination of quanta of these energies. A proportional counter, fitted with a thin aluminium window to allow the passage of the quanta into the counting volume, and a cathode lining of aluminium to ensure a minimum photo-absorption and maximum Compton absorption at the wall, has been used by Curran, Angus and Cockroft (1949 a). Using argon as the counting gas the quantum is absorbed to produce photo-electrons and Auger electrons, with total

energy very close to the energy of the quantum, and the extremely high sensitivity achieved makes the technique invaluable.

Limitations of the proportional tube spectrometer.

At the commencement of this work the proportional tube spectrometer had definite limitations in its performance. We have already seen that the resolution to be expected under given operating conditions was not accurately known, and that the range in energy over which the instrument could be used was limited. In addition to these limitations, the gas multiplication and sensitivity of the counting tube were affected near the ends of the wire by the distortion of the electric field, known as "end-effect" (Curran, Angus and Cockroft 1949 a). The method of correcting for this used by Angus, Cockroft and Curran (1949) was not altogether satisfactory. Their procedure consisted in dividing the central wire of the counter into two sections of unequal length joined together by a glass bead; the histograms of the pulse distribution were measured for each end and the undistorted spectrum was obtained by subtraction. However, the number of pulses counted had to be considerable to avoid poor statistics. Finally, the methods available for calibration of the energy scale had not been completely examined (see next chapter).



Head amplifier circuit.

Fig.I - Head Amplifier Circuit.

Apparatus.

Throughout the experiments to be reported here the amplifier system and recording apparatus described by Curran, Angus and Cockroft (1949 a) were used. The main amplifier, which was of the push-pull type, was designed to give a good signal-to-noise ratio. The head amplifier is shown in fig.1. No grid leak was used when maximum sensitivity was desired, but this occasionally led to difficulty at high counting rates due to biasing-off in the first valve. The introduction of a 100-megohm resistance in the grid circuit overcomes this difficulty, but the signal-to-noise ratio suffers a reduction. The output voltage of the amplifier was used to deflect the spot of a cathode ray oscillograph in a horizontal plane. 35 mm. film (R55 Kodak) was moved vertically through a camera at uniform speed to give a permanent photographic record of the spectrum being investigated. The camera lens had an aperture of f/1.8. It was found that between 30 and 40 pulses per inch of film was most convenient. This method has advantages over the pulse discriminating circuit method in that a permanent record over all energy ranges of interest can be obtained in a relatively short time. This allows a considerable relaxation in the requirements of voltage stability. The attendant disadvantage is the labour involved in the analysis of the film which is done in the following manner. The film is

projected upon a screen, which is subdivided into 120 amplitude intervals, each $\frac{1}{3}$ cm. in height. The histogram of the radiation is built up by counting the number of pulses which fall into each energy interval. The pulse spectrum is normally calibrated by superimposing the pulses resulting from the detection in the counter of homogeneous X-radiation.

CHAPTER II.

The object of the research.

We have seen that the conventional type of spectrometer is not suitable for measurements on low energy electrons, due to straggling and reflexion of the electrons and the absorption in the thin window of the Geiger tube detector. The method of overcoming these disadvantages using the proportional tube spectrometer has been shown. However, this instrument has its own limitations (already mentioned) and it is the first object of the present research to improve the performance of this type of spectrometer. This is to be accomplished as follows:

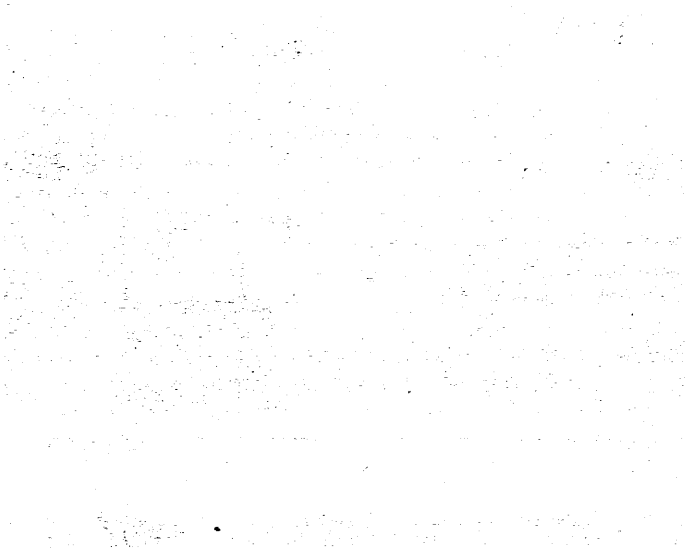
a) the methods available for calibration of the energy scale will be improved,

b) the average energy lost per ion pair produced by slow electrons expending their energy in various counter gases will be measured to provide better data for calculation of the resolution of the tube,

c) the range in energy over which measurements may be made will be increased by suitable use of magnetic fields.

Concurrent with this work, Cockroft and Curran (1951) corrected the end-effect using field adjusting tubes, and counters free from end-effect were always used after their experiments.

The second object of this work is the accurate measurement of the shapes of three beta spectra, using, whenever necessary, the improvements mentioned above.



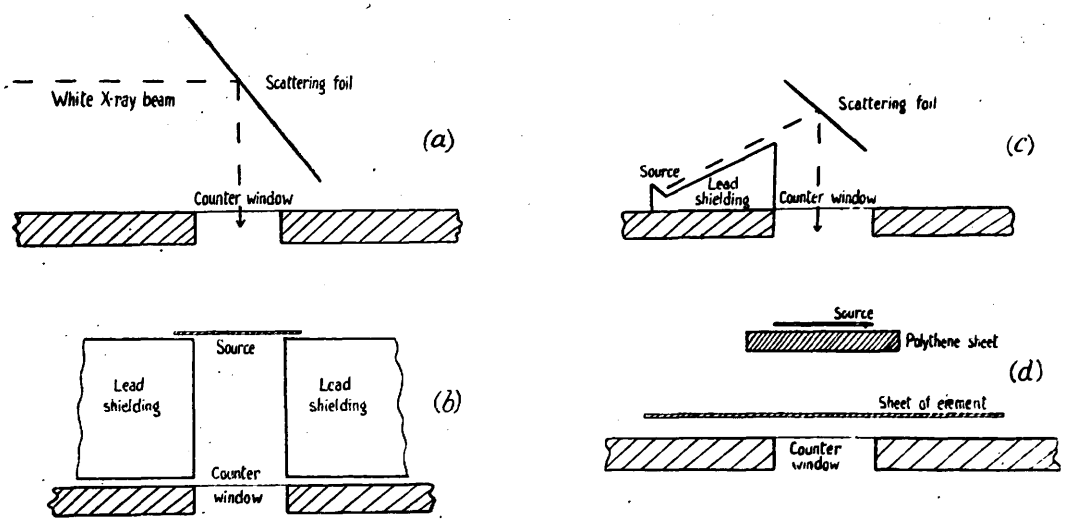


Fig.2 - X-ray Geometries.

PART II

Improvements to the Technique

CHAPTER I

Calibration of proportional counters by the excitation of fluorescence radiation with radioactive sources.

When the shape of a beta continuum is being examined using the proportional tube spectrometer, the energy scale of the pulse spectrum must be calibrated. So far, no methodical examination of the techniques available for calibration purposes has been made, and it is the object of the present work to examine these methods. Rothwell and West (1950 a) have already used radioactive sources for calibration of proportional counters, but the chief method described here is original and it has wide application.

In the early investigations using proportional tube spectrometers, the beam from an ordinary A.C.-operated X-ray machine, adjusted to operate at a small output intensity and which emitted white radiation, was allowed to fall on a thin foil and the scattered radiation was detected in the counter at an angle to the beam direction (fig.2(a)) (Curran, Angus and Cockroft 1949 a). This radiation entering the tube consisted mainly of the fluorescence X-rays of the scattering element. However, this method is not ideal. The X-ray machine is cumbersome, may give rise to spurious pulses (due to electrical interference),

TABLE I

Isotope	Half life	Transition	Approx. energy of X-rays	Associated radiation
In^{114}	50 days	isomeric	24.1 KeV	1.98 MeV $\beta + \gamma$'s
Sn^{113}	105 days	K-capture	24.1 KeV	γ's and e^-
Ge^{71}	11 days	K-capture	9.2 KeV	negligible
Fe^{55}	4 days	K-capture	5.9 KeV	none if pure

and calibration is accompanied by an increase in the "background" of the counter, due to the detection of the white radiation. In addition, the tube in ageing fails to self-rectify adequately and, hence, the pulses appear grouped on part of alternate half-cycles of the mains supply. For these reasons, new calibration techniques were explored and normally preferred. Pile-activated sources, which decayed by K-capture, or which showed nuclear isomerism or strong internal conversion of their gamma radiation, were found suitable as agents for the production of beams of homogeneous X-radiation or electrons. Sometimes the sources were used directly, but two methods of greatly extending their usefulness were established. The sources which have been most commonly applied in the course of several studies are shown in Table I.

Elimination of the effects of hard beta and gamma radiation.

The sources should have a reasonable half life for prolonged use. They should preferably be thin, particularly when associated with hard gamma radiation. Experience has shown that isotopes which have long period beta activity may be comfortably handled. The beta particles are absorbed by suitable thicknesses of polythene, which has good transparency for X-radiation. For example, in the investigation of In^{114} (see below) the source was covered by a layer of polythene, 6 mm. thick. This

completely absorbed the hard beta radiation (of energy 1.98 MeV) and only reduced the X-radiation (energy 24.1 KeV) by a relatively small fraction ($\sim 1/4$). When the required soft calibrating X-radiation is associated with gamma radiation (of energy say $\sim$ 100 KeV) some shielding is useful in reducing the undesirable gamma ray flux through the counter to a minimum (fig.2(b)). However, this method can only be applied with advantage to relatively soft gamma rays. In the case of still harder gamma radiation ($\sim$ 0.25 MeV upwards) the source is most conveniently examined when placed really close to the counter window. This simple geometry results in the best solid angle for soft radiation passing into the counter and makes use of the higher efficiency of the counter for such radiation. Usually it is possible to secure a ratio of at least 10 : 1 in efficiency of detection in favour of the soft X-radiation. To obtain optimum results when examining the source in this way, care must be taken to eliminate the "end-effect" of the tube, since a small fraction of the radiation detected penetrates to the region of variable gas gain.

Production of modified radiation.

Calibrating agents which gave homogeneous X-radiation covering the whole range from, say, 3 KeV to 50 KeV in small energy increments were found extremely useful. Since this coverage cannot be

accomplished readily by use of radioactive sources alone new methods were devised. These involved the excitation of the desired radiation by homogeneous X-rays of higher energy originating from a radioactive source. This process is one of high efficiency and it was achieved in practice either by allowing the initial radiation to fall on a thin sheet of the appropriate element and detecting the excited radiation emitted from the front face (reflection (fig.2(c)), or by placing a suitable thickness of the element between the source and the window (transmission). In the former case, the fluorescence radiation greatly predominates in intensity within the counter and its homogeneity is nearly independent of the thickness of the reflecting sheet. In the second case (transmission) a suitable thickness of the sheet can readily be calculated. For example, consider a source emitting homogeneous X-rays of wavelength λ_1, exciting a radiation of wavelength λ_2 in a sheet of the chosen element placed between the source and the counter window. Let this sheet have mass absorption coefficients μ_1 and μ_2 corresponding to λ_1 and λ_2. Let it be of thickness ℓ and density ρ.

It can be shown (see Appendix 1) that the number of fluorescence X-ray quanta entering the counter (fig.2(d)) when I_o quanta fall on the sheet is given by

$$N = K_1 I_o \left[e^{-K_2} - e^{-K_1} \right] / (K_1 - K_2)$$

where $K_1 = \mu_1 \rho \ell$ and $K_2 = \mu_2 \rho \ell$.

The number of unmodified X-rays transmitted through the sheet to the counter is

$$T = I_o e^{-K_1}$$

and the ratio of these is

$$R = N/T = K_1 \left[e^{K_1 - K_2} - 1 \right] / (K_1 - K_2) \quad \text{----- (1)}$$

Examination of equation (1) shows that the ratio R can be made as large as required (> 100 say) by using sufficient thickness ℓ , provided that $\mu_2 < \mu_1$, a condition which is always satisfied in the usual experimental arrangements. (Furthermore this argument is strengthened by the fact that the modified radiation is usually the more efficiently detected). However, in practice large ℓ values may necessitate strong sources and, in some cases, it may be difficult to secure adequate intensity. On the other hand, if $R \sim 1$ is regarded as suitable (peaks due to modified and unmodified radiations of equal intensity) the method can be used with very weak sources (see fig.3).

Regarding the reflection method (fig.2(c)), it is sufficient to note that it can be readily applied in almost every case, but it may be inferior to the transmission method when very weak sources are employed.

These methods were applied to excite the fluorescence radiations of a number of elements

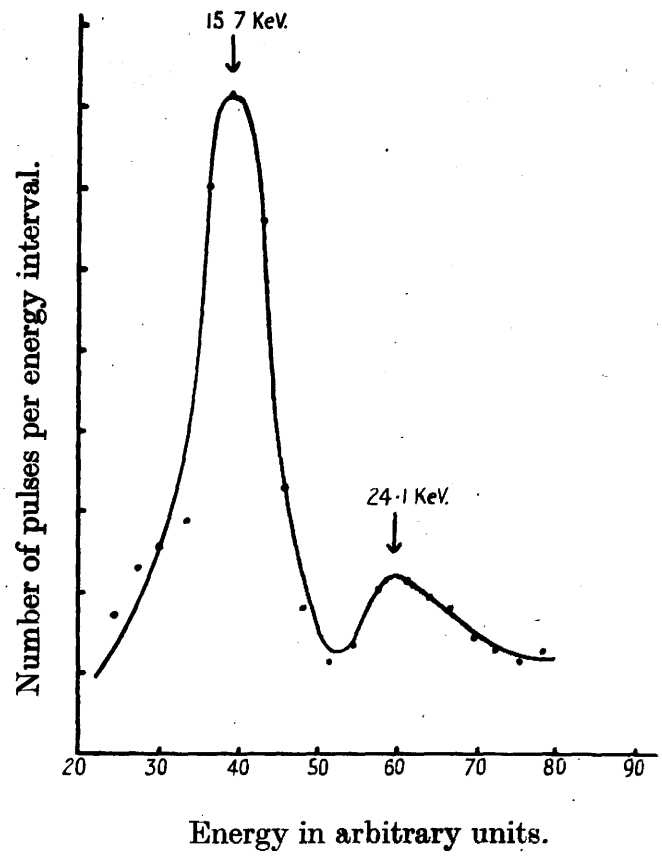


Fig.3. - Transmission case

TABLE II

Source	Excited element	E_o in KeV	E_m	Type of geometry	Histogram
In^{114}	Zr	24.1	15.7	Transmission	Fig.3
In^{114}	Cu	24.1	8.05	Reflection	Fig.4(a)
Ge^{71}	Ni	9.2	7.5	Reflection	Fig.4(b)
Ge^{71}	Fe	9.2	6.4	Reflection	Fig.4(c)

(see Table 2). In this table E_o is the approximate energy of the initial X-radiation falling on the element and E_m is the approximate energy of the modified radiation.

With the simple geometry demonstrated in Fig.2(c) it was possible to detect as modified radiation $\sim$ 5 percent of the total radiation emanating from the source. Hence, a source strength of $< 1\,\mu$ curie total activity proved sufficient. Indium and Germanium sources were found adequate for excitation of the fluorescence radiations from less than 5 KeV upwards, which considerably increased the number of homogeneous X-ray calibrating agents available within this energy range.

Some further work in this technique is necessary for use with proportional counters in magnetic fields. Proportional tubes will be used to measure particle energies $>$ 1 MeV with the aid of magnetic fields (see Chapter III). These examinations demand calibration radiations of several hundred KeV energy. Sources which decay by gamma transitions (not associated with beta activity) and which show strong internal conversion, should be studied (e.g. Sn^{113}; gamma ray energy 392 KeV, and $\sim$ 70 percent internal conversion). Such sources would be introduced into the counter.

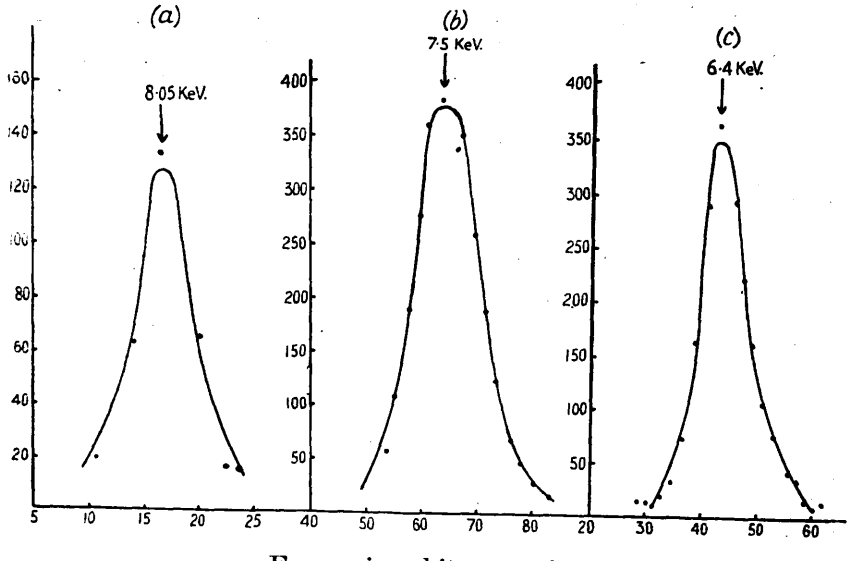


Fig.4 - Reflection case

Identification of isomeric transitions.

The primary phenomena externally observable in an isomeric transition are the emission of gamma radiation and usually conversion electrons. Accompanying the latter is a K X-radiation which is detected with high efficiency and accuracy by a proportional counter. Since this X-radiation is frequently difficult to identify with certainty, the method described here is an important aid. The isomeric transition in In^{114} was studied and the $K\alpha$ fluorescence X-ray histogram is shown in fig.5, curve A. At the same time, the $K\alpha$ fluorescence radiations of cadmium (energy 23.1 KeV), Indium (24.1 KeV) and tin (25.2 KeV) were excited and compared with the radiation arising in the nuclear process. The three histograms are shown in fig.5, curves B, C and D. This demonstrates at once that the known gamma ray, energy 192 KeV, which is strongly converted, is emitted prior to any transition in which Z changes. The importance of this method for the accurate identification of isomeric processes is obvious.

Measurement of the Auger effect in argon.

In the course of this work, argon was used as the absorbing gas. For the energies investigated it is superior to the gases of greater atomic number, such as Xenon and Krypton, which have a relatively small Auger effect, causing homogeneous X-radiation to yield two peaks of comparable intensity which can

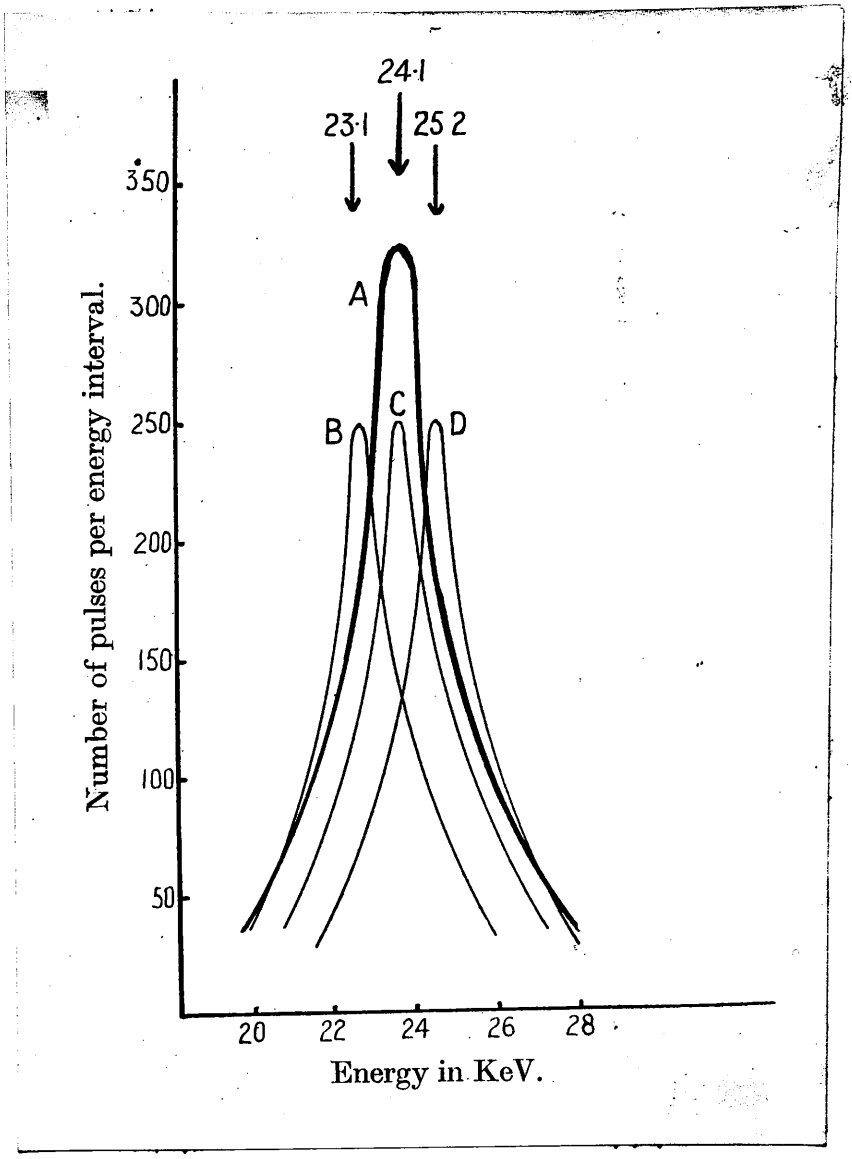


Fig.5 - Identification of an isomeric transition.

lead to uncertainties. One peak corresponds to the case when the X-ray escapes from its own atom and is not detected. The Auger effect in argon is high and an independent value for this effect was obtained. This was accomplished by studying an X-ray histogram obtained when the fluorescence K X-rays of iron were excited by a germanium source. Two peaks were evident, one due to the fluorescence K α X-radiation of iron of energy 6.4 KeV, and one to the photo-electrons (energy 3.4 KeV) from the K level of Argon when the X-ray escaped and did not form an Auger electron (see Fig.6). The effect on the smaller energy peak, due to the 3 KeV X-radiation caught in the gas, was calculated and the "background" spectrum of the counter was subtracted from the histogram. Using this the Auger effect in Argon was found to be 94 percent in reasonable agreement with previous work (Compton and Allison 1936).

Throughout this work a counter $5\frac{1}{2}$" internal diameter was used with a filling of 60 cm. Hg. of Argon and 15 cm. Hg. of methane.

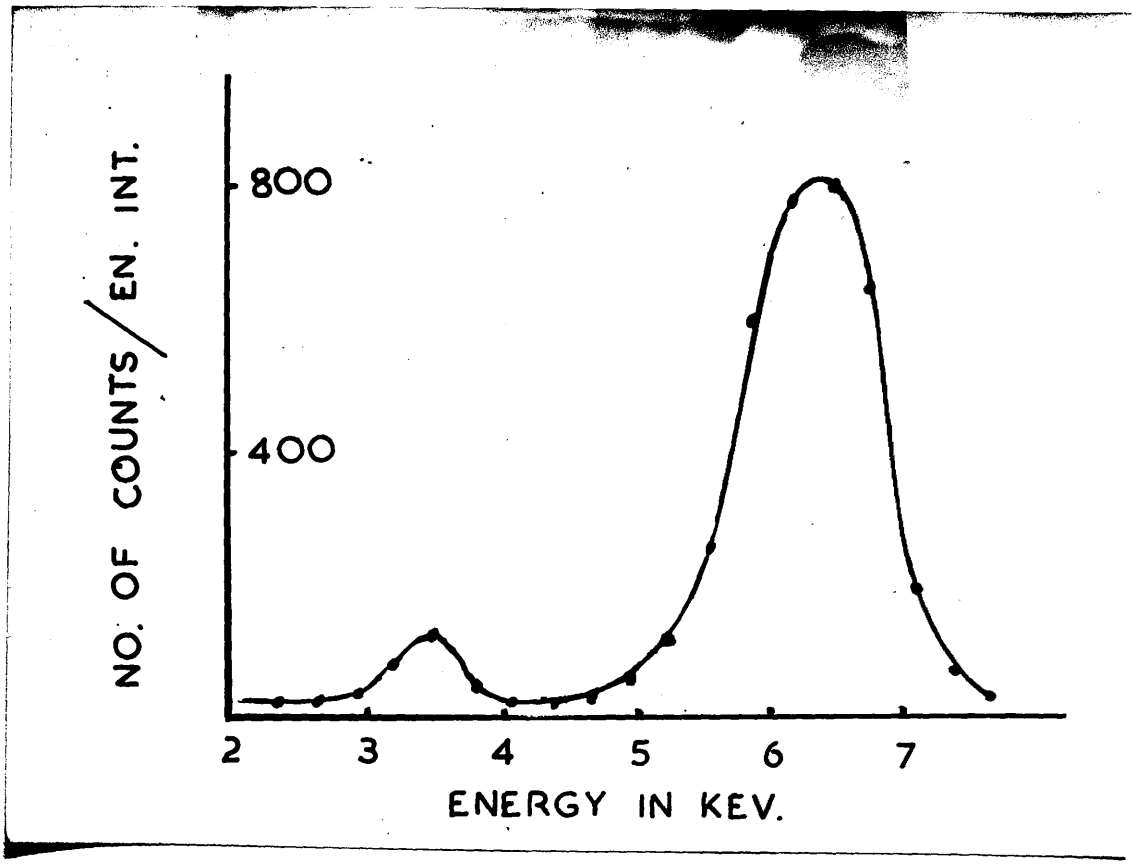


Fig.6 - Measurement of the Auger effect in Argon.

CHAPTER II

The resolution of the proportional tube spectrometer.

The energy expenditure per ion-pair for slow electrons in various gases.

We now deal with the measurement of the average energy lost per ion-pair produced by slow electrons in various gases, since the calculation of the resolving power of the proportional tube spectrometer necessitates a knowledge of this (see part I). The gases investigated were argon, methane, ethylene, air, nitrogen, oxygen and carbon dioxide.

Many investigations have been pursued to determine the energy expenditure per ion-pair produced for various ionizing particles (see Compton and Allison 1936; reviews by Gray (1944) and Binks (1936), and recent values tabulated in MDDC - 1017). However, there is little data directly obtained for electrons of known energy which achieves reasonable accuracy. The methods employed have been, in general, unreliable, particularly for measurements with low energy electrons. Determinations with monokinetic electrons have been made by Eisl (1929) and Gerbes (1935) for air and by Micademus (1946) for argon. Since the results are not conclusive it was decided to make an effort to obtain accurate values for electrons in the gases listed above. Included in the selection are the gases most commonly

used in proportional counting techniques. As the experiments progressed, it became obvious that the proportional counter lent itself well to this work, and a satisfactory accuracy was achieved.

The experiments commenced with an absolute measurement of the energy expenditure per ion-pair in a mixture of argon and methane. Two different methods were applied.

1) X-ray method

The nearly homogeneous X-rays resulting from the K-capture process in Cu^{64} were employed. We know that the average energy of such quanta, passing with a proportional counter, can be accurately measured, and that the number expending their energy in the gas can be ascertained with 100 percent efficiency. If the flux of quanta entering the tube and being detected is n/sec., and the average energy expended by the quantum, in the production of a photo-electron and Auger electron in the gas, is E electron-volts, the number of ion-pairs produced is n E/V per sec., where V is the energy expended in the production of each ion-pair. n and E are measured by operating the tube as a proportional counter, while the current i, produced by these ions, is measured by reducing the voltage applied to the case and operating the tube as an ionization chamber. V can be calculated since

$$i = n E/V \qquad \text{----- (2)}$$

As we shall see, one disadvantage of using this source lay in the presence of associated annihilation gamma radiation. A small fraction of these quanta was detected with the result that we did not have a completely homogeneous radiation. However, this lack of homogeneity was not serious, since the constancy of V as a function of the energy of the ionizing radiation has been demonstrated for the gas mixture used for this measurement (Curran, Angus and Cockroft 1949 a). It is, of course, necessary to measure the average energy accurately.

Unfortunately, the electrometer valve circuit used was not sufficiently sensitive to measure currents caused by counting rates where $n \sim 10^3$ c/sec., which represents a reasonable value for easy counting rate measurements. Consequently, a known fraction of the source producing the ion-current was used to measure the counting rate. In the case of the Cu^{64} X-rays this fraction was achieved by allowing the source to decay between the current and the counting rate measurements. If a fraction f of the source gives n quanta/sec. then

$$fi = n E/V \qquad \text{----- (3)}$$

A value of $f \sim 1$ percent was usually employed. This permitted both counting rate and current measurements to be obtained with reasonable accuracy and ease.

2) Beta ray method

This method employed the beta rays of

tritium. Since only a fraction of the X-rays are absorbed in the gas, constant pressure must be maintained throughout all experiments where they are used as the primary agents of ionization. In addition, the value of n must be measured for each gas investigated, since it is not possible to calculate the change of counting rate resulting from the use of a gas with a different value of mass absorption coefficient. This demands proper quantitative operation of the counter with all gases under examination. Some of the gases investigated, such as air, oxygen and carbon dioxide, are not suitable for proportional counter action, since it is impossible to obtain good counting plateaux on the characteristic curves. To overcome these difficulties a procedure was devised using the slow beta rays of tritium gas as the primary agents in the ionization process. A quantity of tritium was divided into two portions. One, giving a suitable ionization current, was passed into a chamber which was then filled with the gas mixture under investigation. The other, which gave a suitable counting rate, was passed into a tube which was to act as a proportional counter to give a value of n. This tube was then filled with an argon-methane mixture for proportional action. The larger quantity of tritium was accurately related to the smaller quantity used for evaluating n, the ratio of the activities being determined by measuring either relative volumes or pressures.

The average energy of the beta rays forming the tritium spectrum is accurately known from recent work with the proportional tube spectrometer (Curran, Angus and Cockroft 1949 b), the value being 5.7 KeV (see also Slack, Owen and Primakoff 1949, and Jenks, Ghormley, and Sweeton 1949). The superiority of this method is at once evident. Any gas or mixture of gases, whether having electron-attachment coefficients or not, can be examined provided that it gives a saturation current in an ionization chamber. The absolute value of V can be measured for one gas and the values of V for other gases obtained by using definitely related quantities of tritium in each gas in turn. The partial pressure of tritium was kept well below any value likely to affect the accuracy of the results. Because of the short range of the beta rays, a counter of 2" diameter at atmospheric pressure is sufficient to ensure that all but $\sim$ 1 percent of the energy is expended in the gas in the production of ion-pairs. Higher pressures reduce this value even more, so that the transparency problem experienced in X-ray cases is overcome.

Apparatus

The electrometer valve circuit used for all current measurements throughout this work is seen in fig.7. The valve employed (D.B.M.6A) was carefully screened and a well shielded lead was taken from the collecting electrode of the ionization chamber to the

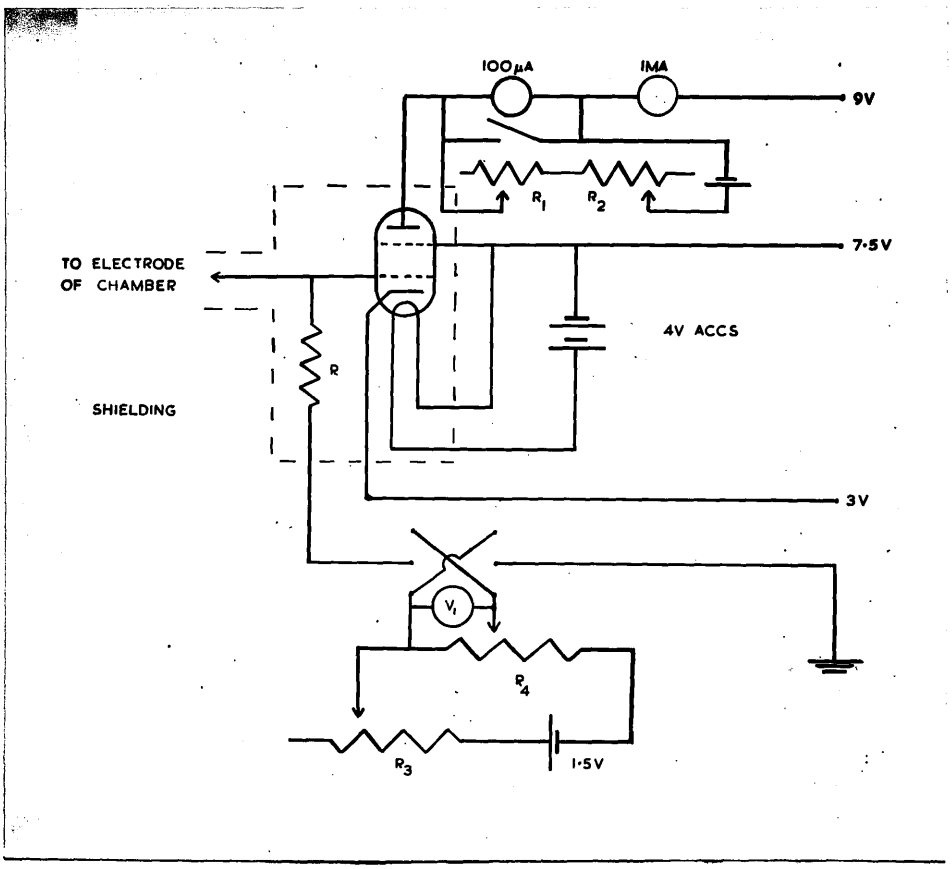


Fig.7 - Electrometer valve circuit.

grid of the valve. This lead had a high resistance leakage to ground, R. At the commencement of a current measurement, the grid was earthed and the sensitive 100 μA meter in the anode circuit was set at 50 μA, using the variable resistances R_1 and R_2, which had values of 20,000 ohms and 1,000 ohms respectively. The grid was then unearthed, which permitted it to attain the potential caused by the ionization current. This potential was positive or negative for collection of positive or negative ions respectively. By increasing R_4 from zero and making fine adjustment with R_3 this potential was cancelled and the sensitive meter reset to read 50 μA. The bias voltage, caused by collection of ionization current, was thus observed and measured on a large scale, standardised voltmeter V_1. Currents of about 10^{-12} amp. were comfortably measured.

The accuracy of the current measured depends on a precise knowledge of the value of the high resistance R. This value was determined in three ways:

(a) a bridge measurement was made, using a sensitive galvanometer and comparing R with a standard $\frac{1}{2}$ megohm resistance, using resistances of intermediate value as steps,

(b) an accurately measured condenser of 993 p.f.

was discharged through R in parallel with a gold leaf electroscope. The times taken to discharge through known calibrated potential differences were measured,

(c) resistor balancing on the electrometer valve circuit was also employed. It was found that

$$R = (1{\cdot}79 \pm 0{\cdot}03) \times 10^{11} \text{ ohms.}$$

The temperature dependence of R was not known. To ensure that R remained constant over long periods a calibration chamber was constructed. This consisted of a cylindrical ionization chamber with a radium source ($\frac{1}{2}$ mgm) supported at a known distance ($\sim$ 1") from it. The current in this chamber was observed before and after each measurement and it was found to remain essentially constant throughout the whole experiment.

Absolute value of V for argon using the X-ray method

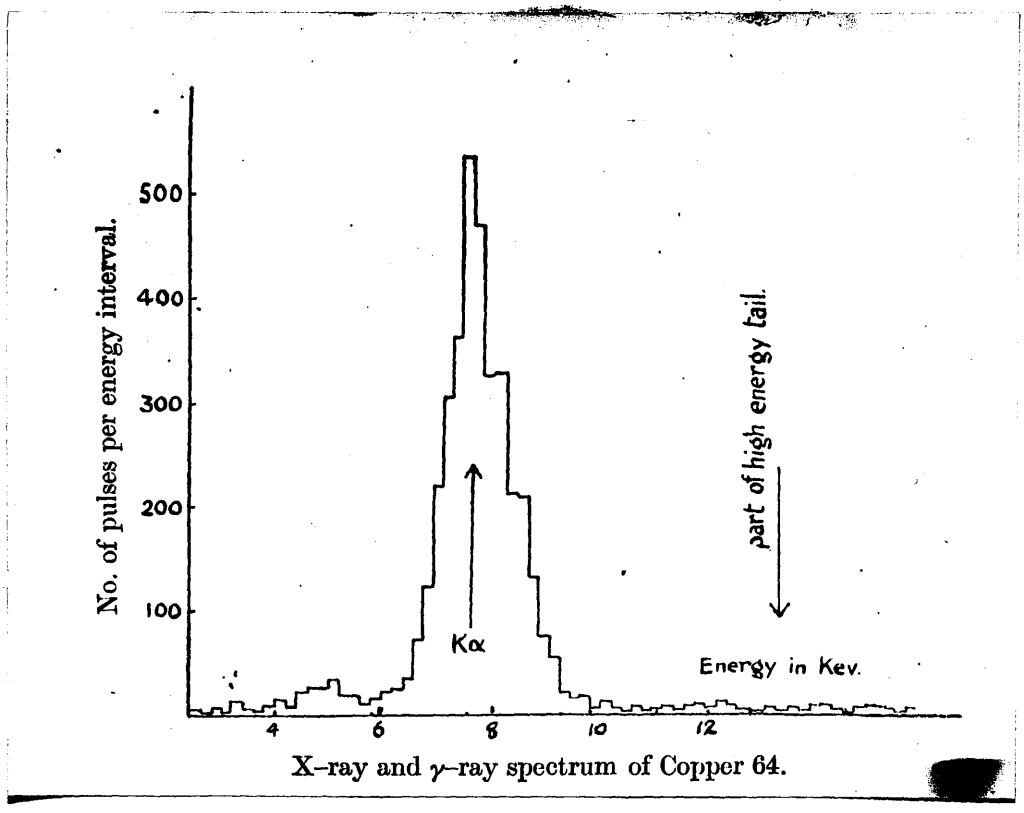
The extensive use of argon in counters, ion chambers, and cloud chambers makes it a suitable choice for the measurement of the absolute value of V. The source used to give the ionization was Cu^{64}, which was well suited for the purpose, having a half life of 12.8 hours and emitting the soft $K\alpha$ fluorescence X-rays of nickel (energy 7.5 KeV)

resulting from the K-capture process. A suitable value for f (equation 3) was obtained by allowing a lapse of three days between the current and the counting rate measurements, using the radioactive decay to give a suitable intensity for the latter.

The counter used was 4" in internal diameter and 20" in length. A small piece of radioactive copper was soldered to the inside of a central window and covered with polythene to absorb all electrons and positrons. The counter was then filled to atmospheric pressure with a 60/15 mixture of argon and methane, and the ionization current measured. During this measurement, it was found that the electrometer valve circuit was sensitive to movements in its vicinity, and shock absorbers were fitted under the mounting supporting the apparatus. This improved the performance but complete stability was only achieved after precautions had been taken to ensure that there were no air currents in the neighbourhood of the apparatus. The measurement of the ionization current was repeated a number of times with different values of voltage applied to the tube, and we feel confident that our results are not in error because of inaccurate current measurement. It was found that good saturation was attained with 360 volts on the tube, both for the collection of positive ions and electrons. Three days later the counter was

operated in the proportional region, the counting plateau being obtained with two scalers in series. The counting rate was found to be 133,000 c/min. on the main stretch of the plateau. A slightly higher counting rate was recorded at the lower edge of this plateau, but this effect was believed to be spurious. Similar effects have been observed when counting homogeneous pulses obtained in other investigations using homogeneous X-radiation, and, although no detailed investigations have been pursued, it is believed that the scaler is responsible for the anomaly. A film of the spectrum obtained from the source was analysed, and the average energy found to be 12.67 KeV (fig.8). This measurement was made when the source had decayed to give a counting rate of 20,000 c/min. To ensure that optimum conditions were achieved the counter was refilled both for the counting rate measurement and the investigation of the spectrum shape. Care was taken to ensure that the gas fillings and total pressure in each case were identical. The X-ray peak at 7.5 KeV, corresponding to the $K\alpha$ radiations from nickel, stands on a background of annihilation gamma radiation, which accounts for the high average energy. Using this data it was found that the value of V for a 60/15 mixture of argon and methane was 29.5 ev.

The experimental error is difficult to assess. The influence of the end-effect on the current and counting rate measurements has not been



X-ray and γ-ray spectrum of Copper 64.

Fig.8 - X-ray and γ-ray Spectrum of Copper 64.

taken into account, but the absorption of the X-rays emanating from the source, located at the centre of the tube, makes this less disturbing than usual. It is more likely that the total count was achieved than that complete saturation of the current was reached. (There was small but definite slope on the curve for current as a function of voltage). Considering all limitations the value

$$V = 29.5 \begin{matrix} +0.5 \\ -1.0 \end{matrix} \text{ ev.}$$

seems fairly reliable.

Absolute value of V for argon using the beta rays of tritium.

For this investigation two vessels, each 72" long and 1¼" in diameter were chosen. It was believed that the best method of obtaining a chamber with negligible end-effect lay in the use of long tubes with narrow bore, since the magnitude of the region at the end of the wire affected by the field distortion depends on the tube diameter. The vessels used here have since been employed to measure beta spectra, and their performance compared with that of properly end-corrected tubes (Cockroft and Curran 1951). These investigations have shown that the end-effect for these long, narrow tubes is indeed negligible. These

identical vessels were to be used, the first, with a central wire of 2 mm. diameter, for the measurement of the ionization current, and the second, for the counting rate measurement. This ensured that the counting vessel remained essentially free from tritium contamination, since the radioactive contact was always small, and did not have to be outgassed in the repetition experiments. The fraction of tritium which stuck to the wall was found to be less than 1 percent of the total gas in the tube. Later investigations have shown (Part III, Chapter II) that a preliminary "freezing" of the source before admission into the counter is sufficient to prevent this undesirable contamination, which is probably explained by the presence of tritiated water in the source used here.

The main source of tritium was mixed with a little hydrogen as carrier and passed into the ionization chamber. In the first experiment this was filled with a 135/15 mixture of argon and methane to a pressure of 2 atmospheres, and the ionization current measured. The thick central wire of the chamber enabled easy attainment of saturation current with 360 volts on the case for collection of both electrons and positive ions. The value of f in this case, $\sim 1/200$, was accurately measured by a manometer. This fraction was taken from the ionization chamber and passed into the counter which

was filled to atmospheric pressure with a 60/15 argon-methane counting mixture. These conditions proved ideal for counting plateau measurements, and the counting rate was found to be 40,000 c/min. In this case the pulses were not homogeneous and no spurious increase in the counting rate at the commencement of the plateau was observed. From these results, a value of 28.5 ev. was obtained for V. The experiment was repeated under essentially similar conditions, with a 138/10 argon-methane mixture in the ionization chamber at a pressure of two atmospheres, and the value of V thus obtained was 28.4 ev.

These values are not significantly different from the value of 29.5 ev. obtained using X-rays. They should be considered more accurate since the beta-emitting source is better suited for this work. However, we conclude that the values of V obtained using the beta-rays of tritium, which have an average energy of 5.7 KeV, and the photoelectrons and Auger electrons resulting from the absorption in the gas of the quanta from Cu^{64}, of average energy 12.67 KeV, are essentially the same. This conclusion is supported by earlier work demonstrating the constancy of V as a function of the energy of the electrons (Curran, Angus and Cockroft 1949 a). It will be shown later that the values of V for argon and methane are nearly equal.

Thus we conclude that for an argon-methane mixture of about 10/1

$$V = 28.5 \pm 0.5 \text{ ev.}$$

for electrons of energy between a few KeV and 50 KeV.

Measurement of V for other gases.

To measure the values of V for the remaining gases, the technique of directly comparing them with gases having known values of V was adopted. Since it was necessary for this investigation to subdivide a supply of tritium into a number of volumes of known relative value, the accuracy of what is known as the "sharing" process had to be examined. In this sharing process it was assumed that if a vessel containing tritium were opened into two evacuated vessels of equal volume at the same instant, the amounts of tritium in each of the two vessels would be equal. Since tritium can occasionally have a little tritiated water vapour associated with it, it does not follow that the sharing process will always hold in practice. However, an experiment was carried out to demonstrate that it did. Two evacuated vessels of known volume were simultaneously connected with a source of tritium and then shared in turn with a proportional counter. The counting plateaux, after adjustment had been made for the volumes of the original two vessels, were found to agree to within 1 percent, demonstrating

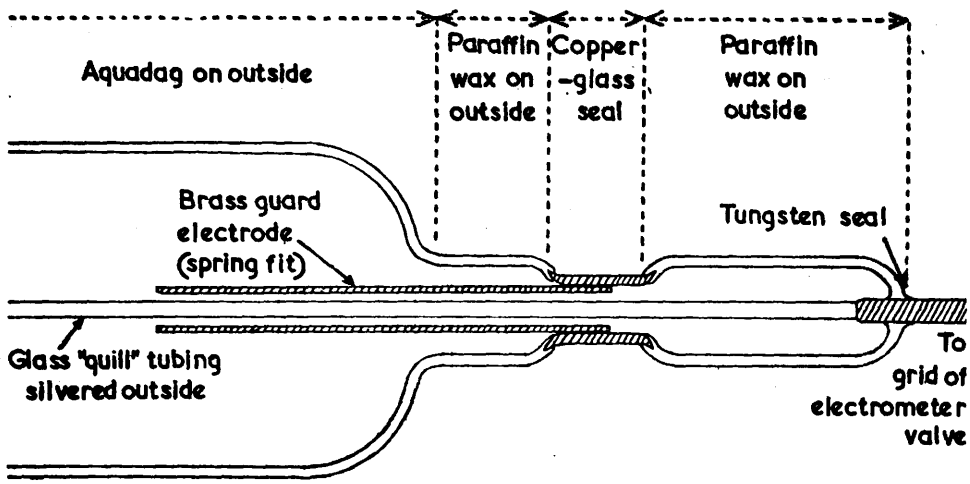


Fig.9 - Single-ended ionization chamber.

that this method of subdividing was satisfactory.

For the measurements with the other gases, a large supply of tritium was shared with six vessels, almost equal in volume. These could be further subdivided if necessary. It was found that when the tritium was left for a period in these vessels it tended to adhere to the walls, and this occasionally necessitated repetition of the experiment. However, the results given below are believed to be unaffected by this adhering property of tritium.

The chambers used for the comparison measurements were constructed entirely of glass except for the central wire and its grounding shield. Aquadag was coated externally on the glass envelope and this served admirably as cathode. Even pyrex glass of specific resistance 5×10^{16} ohms could be used (at least in thicknesses up to 1.3 mm). The central collecting electrode was in the form of a 2 mm. diameter glass tube, coated with silver. This was led out through an earthed guard ring at one end, the other end terminating within the tube (fig.9). Polarisation effects were experienced during long term operation, but were not serious and excellent saturation curves were obtained. Further information on the performance of this type of chamber has been published by Cockroft and Valentine (1950). It should also be mentioned that such vessels are well suited for outgassing in

work with gases of high purity or with gases or vapours which tend to be absorbed by the metals in common use.

The method adopted for measurements with the other gases was simple. A known relative amount of tritium was passed into the counter and to this was added a gas, having a known value of V, to give a total pressure of about 1 atmosphere. The ionization current was measured. Next an atmosphere of the gas to be investigated was added and the ionization current again measured. Thus the value of V for the second gas was determined. The value for air was obtained by this method, the standard reference gas being argon. Taking the value of V for argon as 28.5 ev. the value for air was by comparison 31.0 ev. Next an ionization chamber containing some tritium was filled to atmospheric pressure with nitrogen and the current measured. Oxygen was added to the vessel to give an air mixture, and the current re-examined. Hence, the value for nitrogen was found to be 32.0 ev. Finally, V for oxygen was found to be 28.8 ev. by adding more oxygen and making a 50/50 mixture of oxygen and nitrogen. It was assumed that the change in current from the value for nitrogen was proportional to the percentage oxygen content of the mixture. It was proved that there was no appreciable effect caused by mere pressure increase.

This was done by filling a chamber containing tritium with argon to pressures of 30, 50, 75, 100 and 150 cm. Hg., and observing that the current through the tube was essentially constant. The value of V for carbon dioxide was obtained by direct comparison with argon. It was assumed that the fall in current in changing from tritium + argon at one atmosphere pressure to tritium + argon + carbon dioxide at two atmospheres was proportional to the amount of carbon dioxide added. In this way it was found that for carbon dioxide V = 33.5 ev.

Methane and Ethylene.

The values of V for methane and ethylene were determined in more detail. The current resulting from some tritium in pure argon at atmospheric pressure was measured. Next increasing amounts of methane were added until a total pressure of two atmospheres was reached, the current being measured in each case. Then, starting with methane at one atmosphere, argon was added in increasing amounts. No appreciable change in the ion current was observed through the range pure methane to pure argon. Hence we consider methane to have the same value of V as argon, namely 28.5 ev., and that this value holds for any mixture of argon and methane.

A similar investigation was carried out

TABLE III

Gas	V in electron-volts
Argon	28.5
Air	31.0
Nitrogen	32.0
Oxygen	28.8
Carbon dioxide	33.5
Methane	28.5
Ethylene	30.6

using ethylene instead of argon. It was found that the value of V for ethylene was 30.6 ev. It was also evident that the addition of ethylene to argon raised the value of V from 28.5 ev. to 30.6 ev. linearly with increasing percentage of ethylene. The values for all the gases investigated are listed in table III.

Discussion

The tritium used in the experiments was usually diluted with a little hydrogen to act as carrier. In addition, the gases used were of commercial purity only, which meant that the argon contained about 1 percent of nitrogen and the methane about 2 percent of nitrogen. It is believed that other contaminants may be neglected. There was indication that the value for very pure argon could differ appreciably from 28.5 ev. but no serious attempt was made to investigate this.

Gerbes (1935) discussed a formula, based on the work of Eisl (1929) and Pigge (1934), for the variation of V with the energy of the incident particle in the case of air. He also quoted a value of 31.6 ev. for electrons of energy 60 KeV, but the predicted variation of V is not sufficiently large for us to criticise his formula on the basis of our result of 31.0 ev. However, the measured value of V for 0.3 MeV beta-rays is 32.0 ev., which suggests

that, in the case of air, V is almost constant between $\sim$ 3 KeV and 300 KeV. A value of 31.5 ev. appears to be the best mean value over this range.

Our results are not in complete agreement with those of Nicodemus (1946) for electrons of energy 17.4 KeV in argon. Our value of 28.5 ev., when compared with his value of 26.9 ev., gives a mean value of about 27.7 ev., which is probably just outside the limits of error (believed to be about 0.5 ev.). In view of previous work (Curran, Angus and Cockroft 1949 a, and Hanna, Kirkwood and Pontecorvo, 1949) it seems unlikely that there is any variation of V with electron energy in the range 5.7 KeV to 17.4 KeV.

Regarding some comparable measurements with α-rays and protons, it appears that the above values for electrons are smaller (Gray 1944) in the case of air (and presumably nitrogen), but larger for ethylene and argon (Schneider 1939). The values for carbon dioxide are nearly the same. Apparently there is no change of a regular nature involved here.

The resolution.

We have seen (part I) that the relative variance in the total number of electrons formed in the multiple avalanche is

$$\sigma_p^2/(\bar{m}\,\bar{A})^2 = w/\bar{m}^2 + 1/\bar{m} - 1/\bar{A}\,\bar{m}$$

where $\bar{m} = E/V$. In previous work on the resolution of the proportional tube spectrometer (Curran, Cockroft and Angus 1949), a value of 32 ev. has been assumed for V in an argon-methane mixture. Our present results give a value of 28.5 ev. for this, which increases the value of $\bar{m}$. However, since the precise value of w is not known, we can conclude only that the resolution is slightly better than would be expected from the results of previous work on the average energy expended in the production of an ion-pair - at least for argon and methane, the commonest of counter gases.

Recent Work.

The values of V for some of the gases listed in table III have been re-measured by Valentine, of this Department, (in the press), who used the electrometer valve circuit already described. He had the advantage of having at his disposal counters and ion-chambers fitted with field-adjusting tubes (Cockroft and Curran 1951) to eliminate end-effect. The well-defined collecting volume resulting from the use of these tubes improved the accuracy of absolute measurements, while the increased stability of his ion-chambers permitted the attainment of more consistent results in the relative measurements made with mixtures of gases. Valentine also measured the temperature dependence of the high resistance R. He found that

TABLE IV

Gas	V(Valentine's result) in ev.	V(our result) in ev.
Argon	27.0	28.5
Air	35.0	31.0
Nitrogen	35.8	32.0
Oxygen	32.2	28.8
Carbon dioxide	-	33.5
Methane	30.2	28.5
Ethylene	-	30.6

the coefficient of resistance with temperature was -2.3 percent/°C. between the limits of 15°C. and 20°C., the region of temperature within which all measurements were made. The values of V determined by him have all been normalized at 17.65°C., the temperature at which the absolute value of the resistance was measured. He emphasises that the temperature dependence of R was not considered in our work and that, while the use of the standard ion-chamber probably prevented large errors arising from the cause, it is possible that local variations in temperature led to small inaccuracies. A list of the values obtained by Valentine is given in Table IV, our values being included for the purpose of comparison.

CHAPTER III

Increasing the energy range of the instrument.

As we have already discussed, one of the major limitations of the proportional tube spectrometer is its inability to measure the energies of fast electrons ($>$ 200 KeV say) due to collisions of the particles with the walls of the vessel. When the only means of preventing the electrons from colliding with the walls lies in the use of large volume, high pressure tubes, the instrument is only of use for the investigation of low energy beta emitters. However, this limitation could be overcome if the particles were "wound up" in a magnetic field. It seemed that one of the best ways of achieving this was to place the proportional counter in a uniform magnetic field with its axis parallel to the lines of force, and to use high pressure to confine the range of the particles. The electrons would proceed down the length of the counter, rotating in smaller and smaller loops as they expended their energy in ionization. The source could be mounted on a thin nylon support, either at the end of the truly proportional counting volume (2π acceptance angle) or in the centre of the tube (4π acceptance angle). Such a source would be placed close to the axis of the counter. However, at the

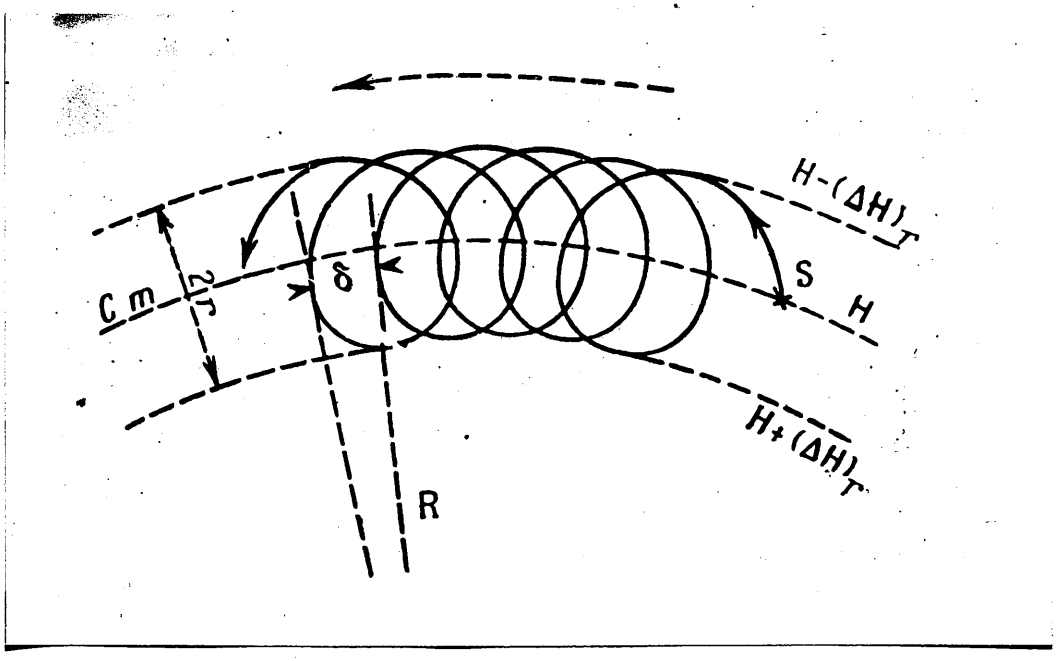


Fig.10 - Trochoidal path.

commencement of the investigation the work of Curran and Cockroft (1951) on the elimination of end-effect had not been carried out, with the result that the only proportional tubes at our disposal had an end-effect. Because of this, long tubes would have been necessary and, consequently, a large pole separation for the magnet, and this proved quite impracticable with the magnet at our disposal. The possibility of using an approximation to the elliptic form of pole face using short ion cylinders of different radii mounted on the axis of the magnet was also considered, but lack of field strength made this method unsatisfactory. In this case the counter axis would have been perpendicular to the axis of the poles. Finally, the answer was found in the type of field first studied by Thibaud, Cartan and Comparat (1938).

Thibaud considered the trajectory of particles emitted from a source placed in the fringing field of an electromagnet. If the particles travelled in an evacuated chamber and emanated from a source on the median plane with an initial velocity perpendicular to the lines of force, their path would be a trochoid (fig.10). Due to the radial field gradient ΔH there is a precession δ. For electrons the displacement caused by the precession will always be in the

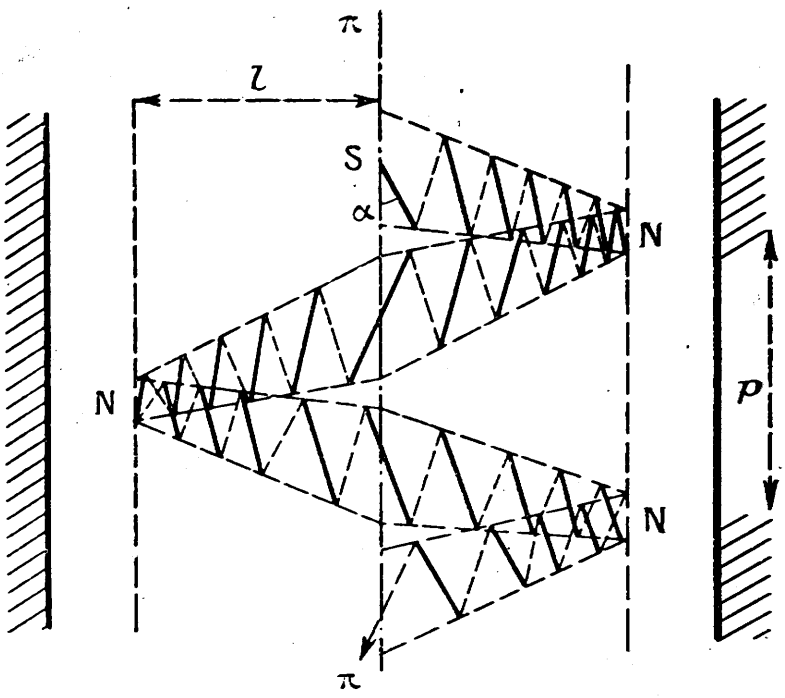


Fig.11. - Shape of the orbit of a particle in the general case.

same direction, irrespective of the initial direction of emission of the particles. It is obviously necessary to make the dimensions of the source small compared with ℓ to permit the escape of as many of the particles as possible. In general, the particles will be emitted in all directions and the path will depend upon the field variation in the transverse and radial directions. Thibaud has shown that a path of the form shown in fig.11. will result.

In our case the paths of the particles will be different, since the electron will continually lose energy along its path. However, the general movement will be similar and with an intense magnetic field with strong convergence of the lines of force, nearly 100 percent of the particles were prevented from reaching the walls of the vessel even when they had energies of the order of 1 MeV. The pressure in the counter need not be high, since the particles perform open loops and have a relatively small displacement per turn. Thus the total length of the path covered by a particle usually exceeds the total displacement from source to end of the track by a factor of more than ten. On the other hand, the complete escape of the particles from the source can be assisted by employing high pressure, since this ensures greater loss of energy per turn at a fixed field value. The importance of

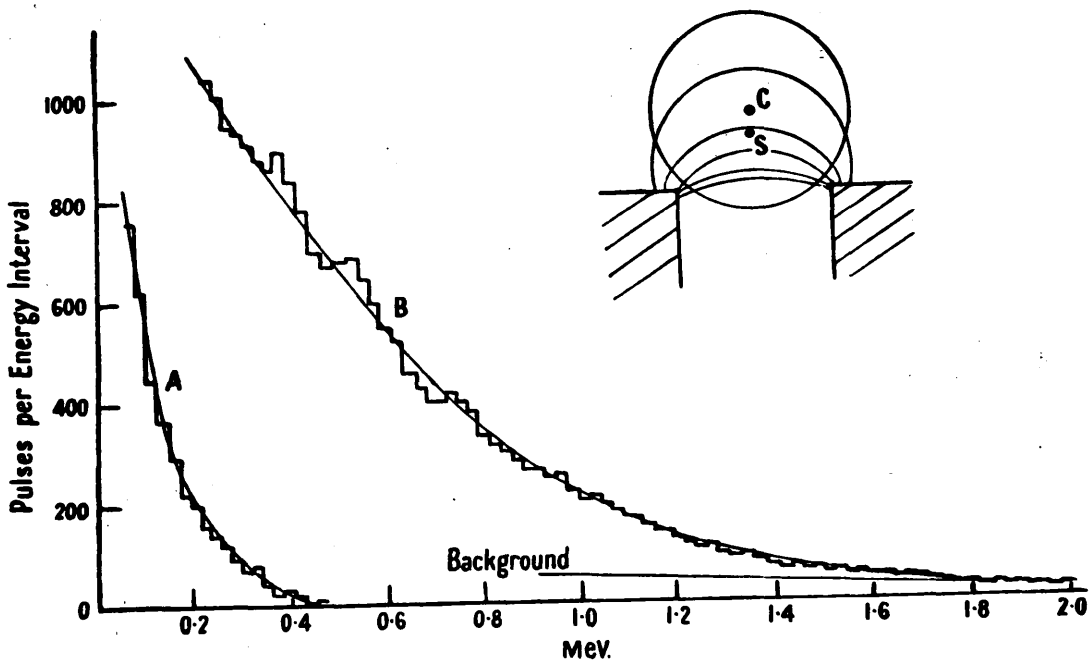


Fig.12. - Histograms of beta rays of P^{32}.

Curve A, no field; curve B, with field; can be normalised by making areas under them equal.

this is well demonstrated when high fields are employed in the analysis of energetic particles, since slower particles, if present, tend to turn back into the source. For the same reason high pressure is advantageous when large sources must be used.

An experiment with P^{32}

The measurement of the shape of the beta spectrum of P^{32} was regarded as a suitable means of testing the ability of the field to concentrate the particles. This source had the advantage of emitting high energy beta particles (the end-point of the spectrum is at 1.7 MeV) which are not associated with gamma radiation. In addition, the shape of the beta spectrum is well known. Siegbahn (1946), using a lens type beta spectrometer, has shown that it agrees fairly closely with the theoretical distribution for an allowed transition down to energy values below 100 KeV.

The proportional tube employed in the investigation, which had an effective counting length of 15" and was 4" in diameter, was situated in the fringing field of our large electromagnet (inset fig.12). The source was very weak, having a strength of only 5×10^{-9} curies and was deposited on the central part of the axial wire, C (inset fig.12). No special precautions were taken to ensure that the source was thin. The

central part of the wire was chosen to ensure that the end-effect of the counter had as small an influence on the shape of the spectrum as possible. However, we were not unduly disturbed by distortions arising from this cause, since our chief aim was not the accurate measurement of the shape of the spectrum but rather a test of the ability of the spectrometer to confine the majority of the high energy particles in the gas. The counter was filled with 60 cm. Hg. of argon and 10 cm. Hg. of methane, giving a total pressure of only 70 cm. Hg. Previous investigations had demonstrated the necessity of operating with the head amplifier in a relatively field-free region, thus preventing reduction in amplifier gain caused by the action of the field on the valves. A well-shielded lead of low capacity, which had a length of about 2 ft., was used to make electrical contact between the central wire of the counter and the grid of the first valve of the head amplifier.

The pulse distribution on the cathode ray tube was investigated with zero field. It was clear that the great majority of the beta rays reached the wall of the counter and expended only a small fraction of their energy in the gas. A photograph of the pulse distribution was secured and the result is seen in curve A, fig.12. It

should be noted that the end-point of this spectrum occurs at an energy value of about 420 KeV. On application of the field a very encouraging increase in the value of the average pulse size was observed. Above a fairly definite lower limit, which could be determined by visual observation, the growth of the average pulse size ceased and the spectrum shape seemed independent of the value of the magnetic field. We assumed that in this region the tracks of even the most energetic particles were being confined to the gas. The histogram of the pulses in curve B (fig.12) was obtained with a field of 5,000 gauss at the source. Comparison of curves A and B demonstrates the remarkable ability of this type of field to concentrate the particle tracks within the gas so that nearly 100 percent of them expend all their energy in ionization. The distribution shown in curve B (fig.12) agrees fairly closely with the theoretical Fermi distribution for an allowed transition down to about 0.9 MeV.

In this arrangement the low energy end of the spectrum is liable to considerable error. In addition to distortions arising from end-effect, some of the lower energy particles will undoubtedly collide with the central wire before expending all their energy, and the thickness of the source and source backing will lead to absorption and scattering.

However, as we have mentioned, the purpose of this investigation was not the accurate measurement of the shape of the spectrum of P^{32}, but an exploration of the possibilities of the technique, and in this respect the results are very encouraging. The fact that the complete histogram was recorded in less than two minutes with a very weak source demonstrates the usefulness of the method.

Application to gamma ray spectroscopy.

The system is admirably suited to the investigation of gamma ray sources, provided care is taken in the preparation of the source mounting. For example, in a case where the source shows strong internal conversion of its gamma radiation, it could be mounted on a thin nylon support and the photoelectrons examined. On the other hand, if the internal conversion coefficient is very low, the use of an external radiator is indicated. When particle radiation is associated with the gamma emission, a suitable covering material (say copper) is placed over the source, and the radiator placed over this material. Under these conditions the Compton distribution will be measured in addition to the photoelectric peak. The test source chosen for this work was Cu^{64}, the annihilation gamma radiation being examined. The source consisted of a small piece of Cu^{64} of total activity 3×10^{-7} curies within a copper-walled

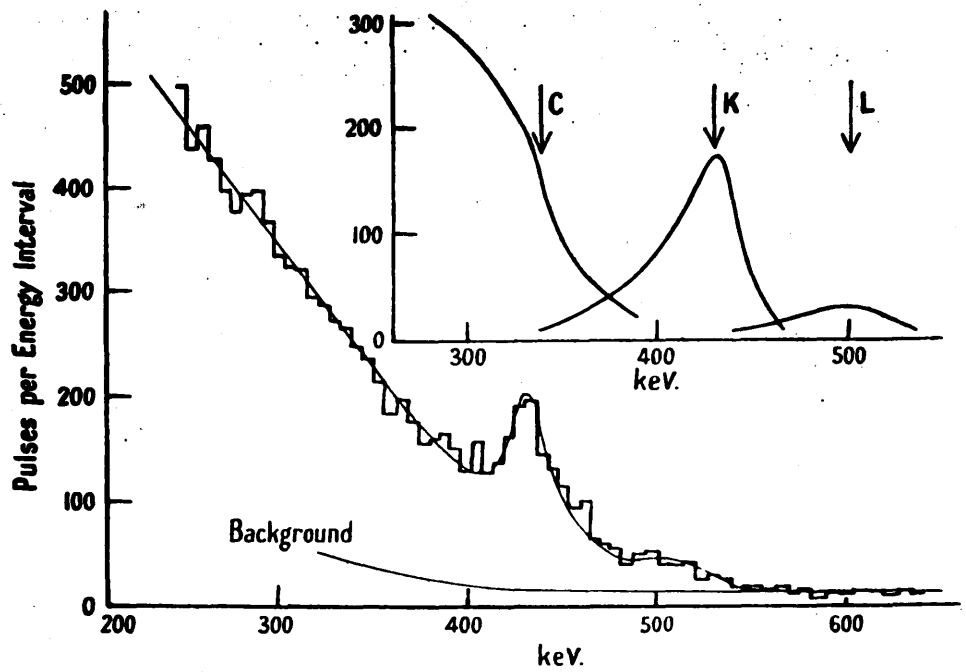


Fig.13. Histogram of secondary electrons produced by annihilation gamma radiation. Source of Cu^{64} in copper covered with gold foil (16 mg/cm^2). Inset figure shows resolution into Compton electrons (C); photoelectrons (K L).

capillary (200 mgm/cm^2) sheathed in gold foil (16 mgm/cm^2). The copper capillary absorbed all the particles (positrons and electrons) leaving the source. This was mounted on a probe running parallel to the central wire and maintained at the voltage corresponding to its position in the counter field (S of inset fig.12). The counter was filled with 15 cm. Hg. of methane and 145 cm. Hg. of argon. The spectrum shown in fig.13 was obtained in a few minutes with the magnetic field applied. The inset figure shows the separation of the histogram into its three components. The photoelectron peaks due to conversion in the L and K shells of gold are shown (energies 500 KeV and 440 KeV respectively) together with the Compton distribution. Each of these gives a quantum energy of 0.51 MeV showing that the annihilation radiation is being detected. High accuracy can be achieved - the resolution of the instrument in this energy range depends entirely on the thickness of the gold foil, since the intrinsic line width at 0.5 MeV is less than 1 percent.

Although no detailed examination of the effect of the magnetic field on the gas gain was undertaken, certain tests were made which showed that there was no appreciable change in the size of calibrating pulses produced by homogeneous X-rays absorbed in the gas for field values as

high as 10^4 gauss. This was expected since the ratio, R, of the magnetic to electric force in the vicinity of the wire is known:

$$R = 300\ H\ v/X\ C.$$

Taking values of $H = 10^4$ gauss, $v = 10^6$ cm/sec., $X = 10^4$ volts/cm., we find that $R = 0.01$.

These results are important. With a few modifications and improvements the proportional tube can now provide a compact beta and gamma ray spectrograph of solid angle 4π, capable of measuring energies over the whole range observed in radioactive disintegrations. The improvements have been made by Cockroft (unpublished) who used properly end-corrected counters (Cockroft and Curran 1951) with their axes parallel to the magnetic field.

Rothwell and West (1950 b) have also reported experiments in which magnetic fields were used to confine the particles in the counter volume.

PART III

Investigations of beta spectra

CHAPTER I

The results of examination on three beta ray spectra will be reported. Two of these, the spectra of H^3 and RaD, were examined with the latest type of proportional tube and we believe that our results represent the best that can be attained with this technique. The other, the spectrum of S^{35}, was examined in 1949 when the technique was not quite so well developed as it is today.

The beta spectrum of S^{35}

By the end of 1948 Cook and Langer (1948) and Cook, Langer and Price (1948 a and b) had carried out a number of investigations on the beta spectrum of S^{35} using their large, high resolution magnetic spectrometer. They reported an upper energy limit of 169.1 $\pm$ 0.5 KeV. in agreement with previous accurate measurements. They observed that the experimental data for the various sources analysed agreed with Fermi theory for energies above about 76 KeV, but that below this energy the experimental data for the various sources rose above the theoretical straight line and then fell rapidly for energies below 15 KeV. The attenuation of the counter window probably accounts in part for the rapidity of the fall.

Berggren and Osborne (1948), using a magnetic lens spectrometer to examine the spectrum, reported that the energy distribution was found to be within 3 percent of the allowed distribution for energies above 40 KeV., and that the end point was 169 ± 3 KeV. Albert and Wu (1948) investigated the spectrum with the Columbia University solenoid spectrometer, using eight different sources. Among these were extremely thin sources which they examined utilising the high geometric efficiency of their spectrometer. They claimed that their results agreed with those of Cook, Langer, and Price when relatively thick sources were used, but that, when the thinnest sources were employed, agreement with Fermi theory down to a lower energy limit of 16 KeV was achieved.

A few months after the above mentioned investigations had been made, Braden, Owen, Townsend, Cook and Shull (1948) published information on the discrepancies caused by source charging in beta spectrometers. Their work was particularly important, since most of the studies of beta spectra being carried out at that time involved the use of non-conducting source backing material. Such source charging effects would not only affect the energy measurements, but might also alter the effective solid angle of the spectrometer for particles of low energy, thus distorting the spectrum shape. It is impossible to single out

any measurement and condemn it because of source charging, but the results of experimenters who have used thin non-conducting source backing, without taking precautions against source charging, should be regarded with suspicion.

The disagreement with Fermi theory observed in the low energy region by the authors mentioned is difficult to understand theoretically. The half life of 87 days, together with the maximum energy of 169 KeV seems to establish the beta transition as definitely allowed (Konopinski 1943). In addition, the spectrum is simple and all investigations have resulted in a complete failure to detect any gamma radiation associated with the disintegration. It is, therefore, particularly important to examine the spectrum using the proportional tube spectrometer, since this instrument is better suited for measurement of low energy electrons than the magnetic spectrometers employed by previous workers. However, the use of proportional tubes is not without disadvantage, since the maximum energy of the beta particles is close to the limit for accurate measurement when high pressures alone are used to confine the particles in the gas - the work with magnetic fields had not been undertaken when this investigation was carried out.

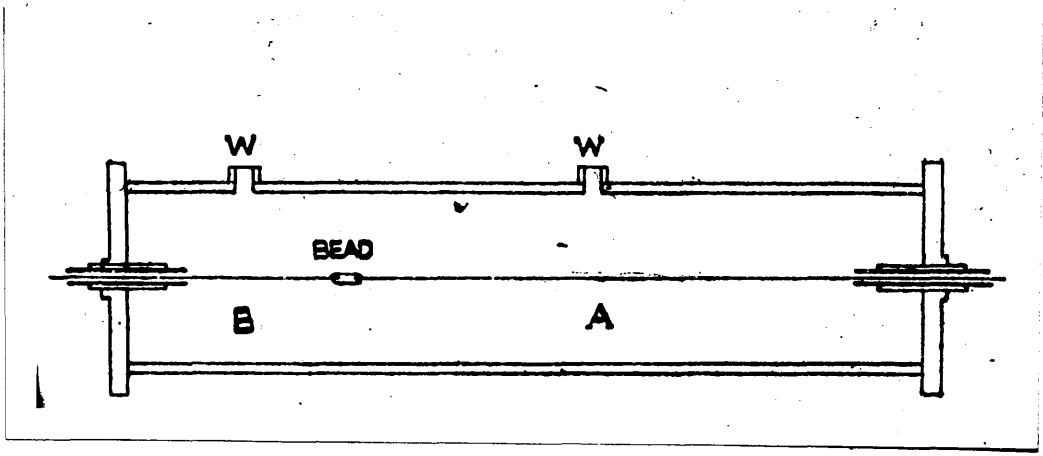


Fig.13a. - The counter used for investigation of S^{35}.

Apparatus

The counter used throughout the investigation is seen in fig.13a. It consisted of a copper tube, 75 cm. long and 14 cm. in diameter, with thick copper end plates supported by longitudinal steel rods to withstand a high pressure of gas. It was divided into two independently operating volumes, A and B, by a glass bead supported by the two portions of the central wire. The two windows, one for each counting volume, were ¼" in diameter and were covered by a sheet of aluminium 0.004" thick for the admission of soft calibrating X-radiations. The central wire in both portions of the counter was 0.004" in diameter and was divided by the glass bead so that the effective anodes were 45 cm. and 15 cm. long, giving a counter of effective length 30 cm. free from end effect (Angus, Cockroft and Curran 1949). The counter was filled with a mixture of methane at a pressure of 30 cm. Hg. and argon at slightly more than 5 atmospheres. It was operated at voltages between 5000 and 6000 volts, and under these conditions it behaved very satisfactorily, the number of spurious pulses being negligible.

Preparation of the source.

It was decided to introduce the source into the counter in the form of H_2S^{35}. However, before the manufacture of the radioactive gas was

attempted the effect of a small partial pressure of stable H_2S in the counting tube had to be considered. The two most probable disadvantages resulting from the use of this gas seemed to be:

(a) A small quantity of H_2S might distort the true spectrum shape due to its electron affinity, although this is believed to be small for electrons of the velocities experienced in the counter avalanche.

(b) A small fraction of the H_2S might possibly adhere to the counter wall. Such a situation would also lead to a distortion of the spectrum, due to some of the beta particles expending part of their energy in the wall.

The first possibility was examined by passing the $K\alpha$ fluorescence X-ray's of molybdenum through the central window of the counter when the gas mixture consisted of 1 mm. Hg. partial pressure of stable H_2S, together with an atmospheric filling of 15 cm. Hg. of methane and 60 cm. Hg. of argon. The histogram obtained was compared with a similar distribution resulting from the $K\alpha$ X-rays of molybdenum in the absence of the H_2S. The distributions were essentially the same, demonstrating that no appreciable electron affinity could be detected with this small partial pressure of H_2S.

The possibility of the H_2S adhering to the counter wall was examined most conveniently using the active gas.

The experiment was undertaken before Croatto and Maddock (1949) reported the production of H_2S^{35} directly in an atomic pile. The first two attempts to prepare the gas failed. The action of N/10 HCl on a small quantity of Na_2S^{35} in N/10 NaOH solution gave no result. It was assumed that the H_2S^{35} was dissolved in one of the stages. An attempt to produce the gas by passage of hydrogen over activated sulphur heated to about 400°C. also failed. Finally, the gas was prepared by heating a mixture of S^{35}, paraffin wax, and ignited asbestos, and collected in a liquid air trap.

Using the active gas it was found that a fraction of the H_2S adhered to the copper walls of the counter. A lining of aquadag reduced this to 10 percent of the total gas, while a stainless steel lining resisted any attack. However, some of the H_2S managed to penetrate behind the lining and the counting rate decreased somewhat during the experiment. Care was taken to ensure that measurements which had to be compared as regards intensity were secured within a short time interval, thus eliminating any appreciable error that might arise from this effect. The partial pressure of H_2S^{35} in the

counter was about 1 mm. Hg.

Pulse distributions.

The counting rates in the two volumes of the counter were 19,000 and 5,000 c/min., compared with background counting rates of 1400 and 400 c/min. respectively.

To observe the spectrum in detail from about 5 KeV to 60 KeV, the gas gain required was found to be $\sim 10^2$. This was used in conjunction with high amplifier gain and the pulse distributions from the volumes A and B were measured. It was found that the background spectrum was practically linear with a small rise at low energies; this was subtracted only from the final spectrum. The energy scale in this "high gain" case was calibrated by superimposing on the spectrum the fluorescence K α X-rays of silver, of energy 22.1 KeV, obtained by reflecting a beam of white X-rays from a silver foil of 0.001" thickness. Observations over the range 20 to 200 KeV were made with the same gas gain, but with the amplifier gain reduced to $\frac{1}{4}$ of its previous value. The pulse distributions from the volumes A and B were again measured. As an additional check on the performance of the amplifier in the two conditions the fluorescence K β X-rays of tungsten, of energy 66.7 KeV, were used as a calibrating radiation. These X-rays were fired directly into the counter through the

copper wall (the Kα group was strongly absorbed in traversing the copper). From the position of this X-ray peak it was evident that the value of $\frac{1}{4}$ for the ratio of amplifier gains was accurate to within 1 percent. A third set of readings was taken for the volume A alone with a view to examining the spectrum in the vicinity of the end-point. This was achieved by blacking out all pulses of energy less than 125 KeV and making a detailed study with good statistics of the pulses above this energy. The result is seen in the inset curve of fig.14. The resolution was not very satisfactory, mainly on account of the shape of the "background" spectrum, but a value of 168 KeV for the end-point seemed most consistent with the experimental data.

The two distributions arising from subtraction of the pulse distribution in the counting volume B from the comparable distribution in the counting volume A, one set obtained with high amplifier gain and the other with low amplifier gain, were fitted together. The distribution resulting from this was fitted to the inset curve of fig.14., and the final result is shown in fig.14. A Fermi plot of this experimental curve was made (fig.15). The theoretical Fermi distribution

$$S_{TH} = (W_o - W)^2 \, (mc^2 + W)^2 / \left\{ 1 - \exp \left(-2\pi\alpha Z (W + mc^2)/p \right) \right\}$$

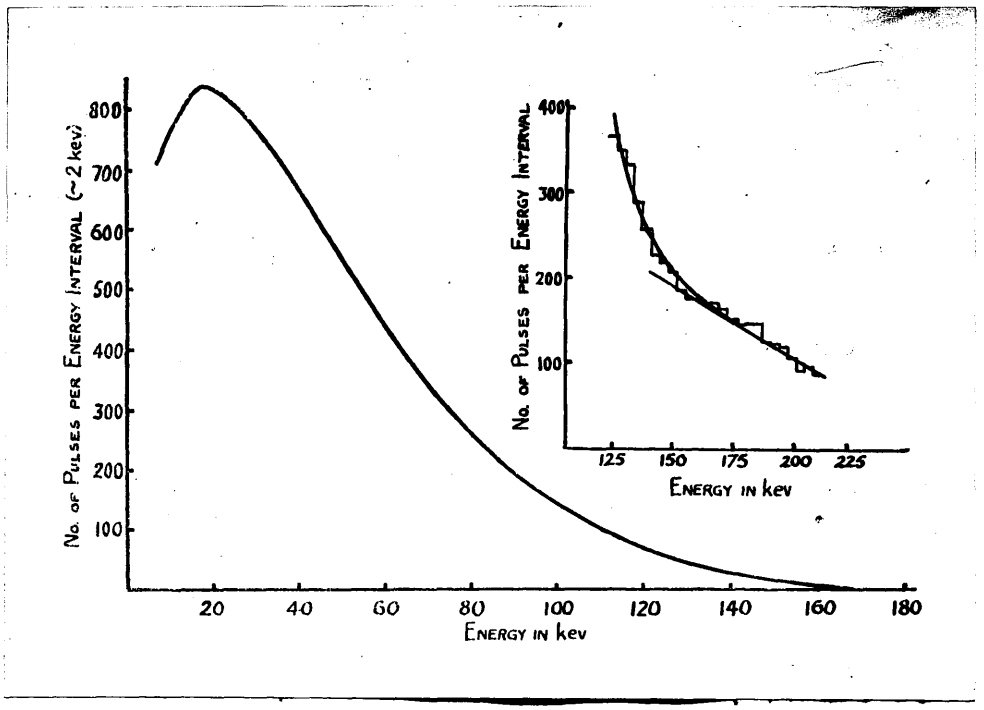


Fig.14. - The beta spectrum of S^{35} with the end-point shown in the inset.

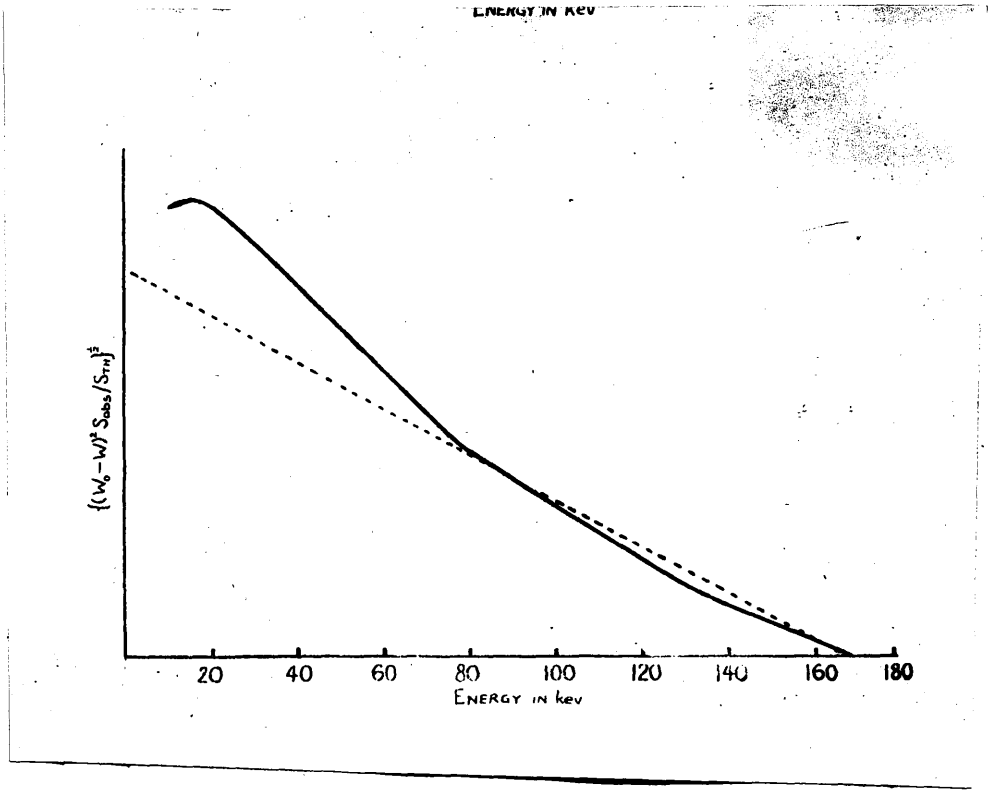


Fig.15. - Fermi plot of the spectrum.

was used. Here W = kinetic energy of the beta particle, mc^2 = the rest energy, p = momentum, α = 1/137 and W_0 = 168 KeV. The Coulomb part of the expression is essentially non-relativistic, but represents a good approximation even for energies of the order of the maximum energy of the spectrum.

Discussion

Our results are not in agreement with Fermi theory for the low energy electrons, even though the beta transition of S^{35} is allowed. The observed departure from agreement cannot be explained by experimental errors - we believe that the spectrum shape is accurate to within 10 percent down to an energy value of about 10 KeV.

It will also be noted that our agreement with the theoretical predictions is not altogether satisfactory in the region of higher energy. However, we are not unduly disturbed by this, since the technique employed leads to the introduction of small errors here. For example, our results were obtained by a subtraction method which is statistically poor in this region. Moreover, the relatively large range of the more energetic beta rays leads to quite an appreciable attenuation of measured intensity in this region, since a number of these particles reach the wall and do

not expend all their energy in ionization of the gas. This "wall-effect" will result in a displacement of some of the pulses into the region of lower energy. Calculation has shown that the disagreement with theory for the high energy particles could be attributed to such an effect. The resulting increase in intensity in the region of lower energy is relatively negligible, however, and the disagreement with Fermi theory here cannot be ascribed to this cause.

Our results are in reasonable agreement with those of Cook, Langer and Price (1948 b), and in somewhat sharp disagreement with those of Albert and Wu (1948). The agreement with Cook et al is quite good. Both their curve and ours have a maximum at about the same energy value ($\sim$ 16.5 KeV), and both sets of observations are in disagreement with Fermi theory for values below 76 KeV. At higher values of energy the agreement of our results is less good, due, we believe, to small errors arising from the causes already discussed. Our results are not as accurate as those of Cook et al in this region, but they are not inconsistent with their findings which show agreement with Fermi theory above 76 KeV. It is worth emphasising the good agreement of our results with theirs, for the technique employed by us was very different from that used by them. For this reason it seems unlikely that any form of systematic error can explain the deviations from the theoretical predictions observed in both cases.

On the other hand, the work of Albert and Wu is in disagreement with our results. They find that for thin sources the spectrum offers no disagreement with Fermi theory even at low energy values. They indicate that the excess of low energy particles found in the investigations of Cook et al might be a function of source thickness. However, other investigators have found no observable increase in the number of low energy particles as the thickness of the source was varied (Cook and Langer 1948), and other investigations resulting in straight line Fermi plots extending to quite low energies have been made even when extremely thick sources were employed (Levy 1947, Saxon 1948, and Alburger 1949).

Faced with this unsatisfactory state of affairs, Langer, Motz, and Price (1950) re-examined the spectrum shape. They concluded that the spectrum was in agreement with Fermi theory at least for energies above 50 KeV. Below this energy their curve shows some increase in the number of low energy particles. However, they believe that their results, coupled with those of Albert and Wu, suggest that the spectrum offers no contradiction to Fermi theory. They obtained a new value of 167 KeV for the end-point energy of the spectrum. Gross and Hamilton (1950 a and b) have examined the spectrum shape below 30 KeV with

their electrostatic spectrometer, and conclude that it is in agreement with Fermi theory in this region.

However, the author feels that the agreement with Fermi theory is not finally established. The experiment reported here had several disadvantages. The subtraction method employed and the collision of a certain fraction of the high energy particles with the walls of the vessel must result in certain inaccuracies. But these inaccuracies should not be sufficient to explain the difference between our results and those of Albert and Wu. Before any final conclusions are reached the spectrum should be re-examined using the proportional tube technique. A brief outline of the method which we hope to employ soon in a re-examination of the spectrum is given in Appendix II.

CHAPTER II

The beta spectrum of tritium

The beta decay of tritium is an allowed transition. The work of Curran, Angus and Cockroft (1949 b) has shown that between ~ 1 KeV and the upper energy limit (17.9 KeV) the spectrum is in good agreement with Fermi theory. However, a tentative suggestion of some deficiency in the number of beta particles in the low energy region was made by these authors. Theoretically, the intensity of the spectrum (particles per unit energy interval) at zero energy should be about 86 percent of the intensity at the maximum of the distribution. Cooper and Rogers (1950) have constructed a composite of the various published spectra and have concluded that all work on the spectrum indicates an excellent agreement with Fermi theory from 2.5 KeV to 18 KeV. The introduction of the improvements to the technique which will be described here makes it possible to examine the spectrum shape to lower energy limits than in the past without appreciable uncertainties arising from spuriously small pulses. These small pulses are those which have their origin in the part of the counter near the ends of the wire, since in this region the gas multiplication is reduced below the normal value unless field-adjusting tubes (Cockroft and Curran 1951) are employed.

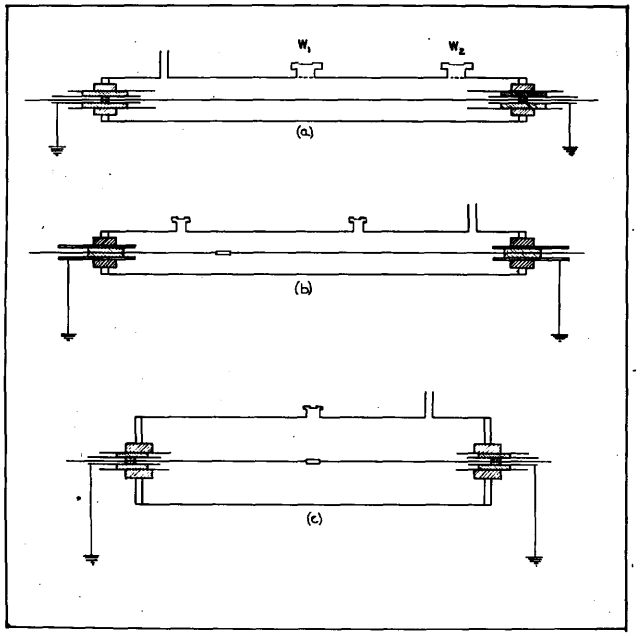


Fig.16. - The proportional counters used in the examination of the tritium spectrum.

Apparatus

A diagram of the counter normally used in this work is seen in fig.16a. The field adjusting tubes were held at the potential appropriate to their diameter, thus ensuring a minimum of "end-effect". The value of this potential was checked by ensuring that pulses from a homogeneous beam of X-radiation, fired through both windows W_1 and W_2 were of equal size at the centre and end of the counter. The counter was lined with a thin sheet of aluminium to minimise secondary effects at the cathode. The lining was perforated at the window positions to prevent any ionization in the window "neck" from being recorded at the central wire. The tube was 15" long and the counter parts are drawn to scale.

The pulses from the counter, after amplification, were recorded on a 35 mm. film. Using this technique, it was found difficult to examine the spectrum shape accurately below 300 ev. The thick stems of the higher energy pulses and the brightness of the baseline itself caused a fogging of the film in the very low energy region. To overcome this limitation, the pulses were measured using a single-stage pulse analyser. This consisted essentially of two scaling units type 1009A. The discriminator of one unit was used to control the first stage of both units. The insertion of a steady source of potential difference (V volts) between the grids of the first stages in

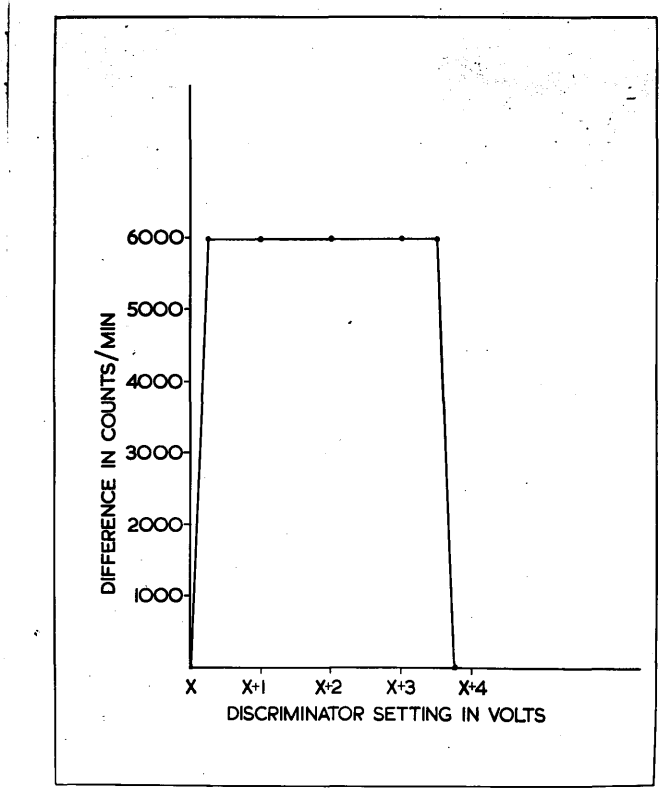


Fig.17. - The shape of the channel of the pulse analyser

each scaling unit supplied a channel width of V volts $\pm$ the small difference in the triggering potentials of each unit. The potentiometer on the discriminator, which determined the voltage X corresponding to the edge of the channel, was examined and found linear. To measure the shape and width of the channel, homogeneous pulses were fed into the analyser. Various values of the amplitude were employed covering the energy range to be measured. Graphs of the difference in the counting rate between the two scalers against the discriminator setting were plotted (fig.17) at various points throughout the range of the discriminator, and it was found that the channel width was constant and that the channel had "rectangular" shape.

The tritium source had argon associated with it to act as carrier. Before the gas was introduced into the counter its temperature was reduced to a value below the freezing point of water. Previous work with tritium gas in counters had shown that sometimes about 1 percent of the tritium stuck to the walls of the counter. Since it appeared likely that this was in the form of tritiated water, the preliminary "freezing" of the source would prevent this undesirable ~~containment~~ contamination from entering the counter. Measurements proved that no tritium stuck to the walls of the counter after this treatment.

Pulse distribution at low energies.

It will be noted that all the above precautions, both in counter design and handling of the source, were introduced to eliminate the possibility of affecting the true spectrum shape, particularly in the direction of moving higher energy particles towards the lower energy region.

Three separate measurements were made on the spectrum with the counter already described. In the first a small quantity of H^3, giving a counting rate of ~ 17,000 c/min. (the background counting rate was ~ 300 c/min.), together with a mixture of methane at a pressure of 16 cm. Hg. and argon at slightly more than 3 atmospheres, was introduced into the counter, fig.16(a). The spectrum shape between ~ 200 ev. and 2 KeV was examined, the pulses being recorded on 35 mm. film. The energy scale was calibrated by superimposing on the spectrum the fluorescence K α X-rays of Gallium (energy 9.2 KeV) arising from the K-capture of Ge^{71}. The amplifier gain was reduced by a factor of 4 to achieve this calibration. The pulse distribution from this measurement on the spectrum is seen in fig.18, and the intensity below about 1 KeV appears to be much less than expected on theoretical grounds. We have already mentioned the uncertainty of the curve below ~ 300 ev.

The experiment was repeated under almost identical conditions, using the pulse analyser to

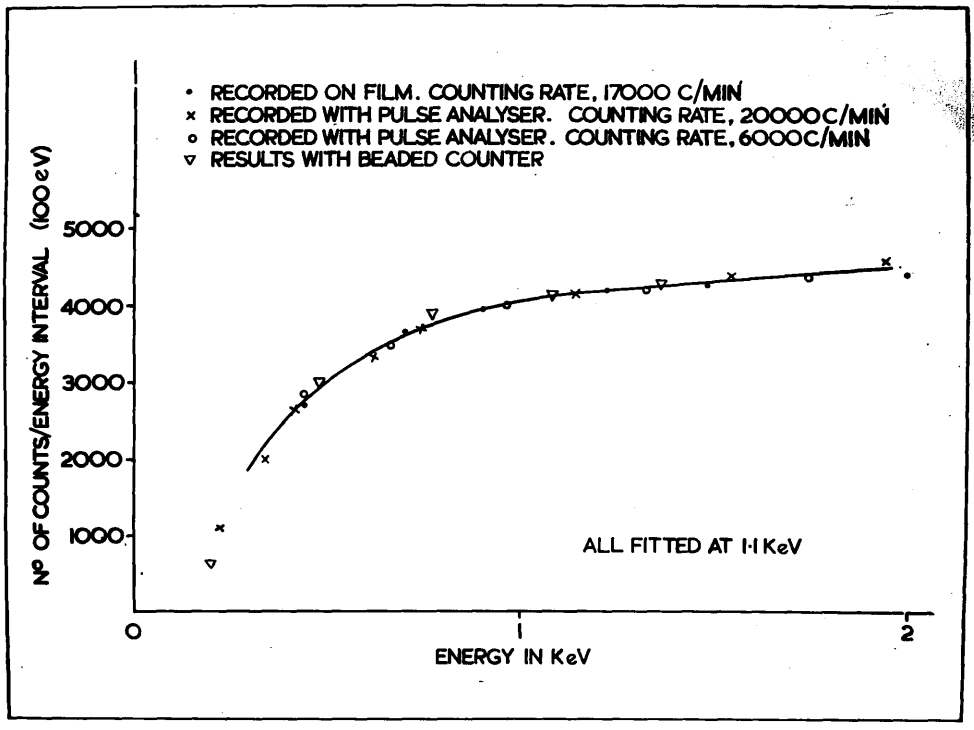


Fig.18. - The tritium spectrum at low energies.

record the spectrum shape. Two measurements were made, one with a counting rate ~ 6,000 c/min., and the other with a counting rate of ~ 20,000 c/min. (fig.18). These results were established with great care, and it was found that the various points repeated satisfactorily after several hours. In the case of the low counting rate, it was not possible to obtain readings below about 300 ev. because of poor statistics. The channel width was kept at 100 ev. throughout the experiments, and the energy scale was calibrated in the same manner as before. Since the results obtained from these determinations with different counting rates agree, it seems unlikely that the apparent lack of low energy pulses is due to any limitation of the amplifier. The particular limitation in mind was the possibility of affecting pulse amplitudes when the pulses occurred within too short an average interval.

In all the above measurements, the field-adjusting tubes were held at the potential appropriate to their diameters, thus ensuring a counting tube free from "end-effect". Curran, Angus and Cockroft (1949 b) have published a result obtained using a counter which was not "end-free" and their experimental curve did not demonstrate any tendency to fall steeply at low energies. To achieve a result comparable with theirs, the field-adjusting tubes were earthed and

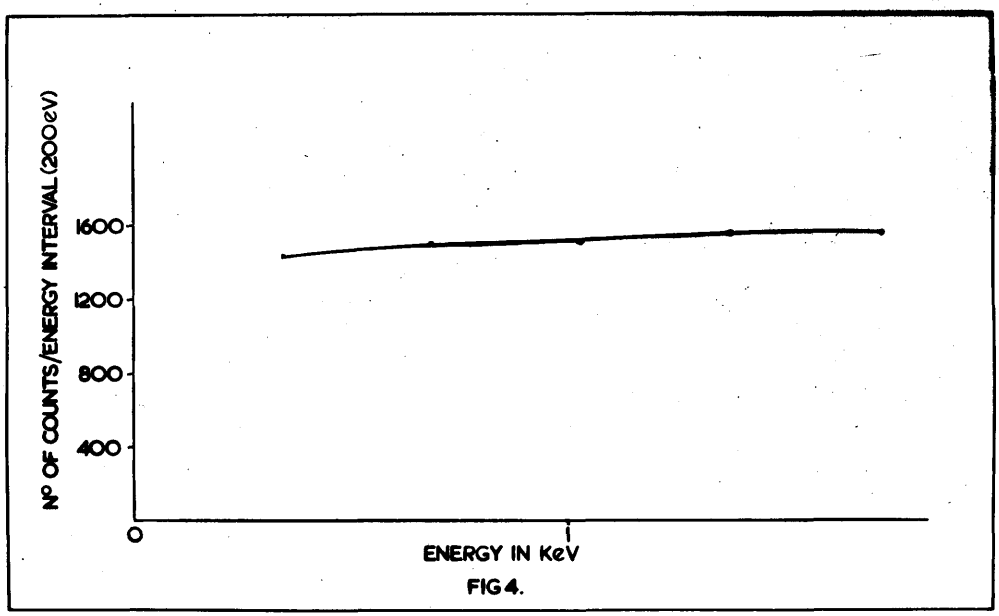


Fig.19. - The tritium spectrum at low energies in a counter which had an end-effect.

the spectrum shape examined (fig.19) in a manner similar to the two previous measurements. The counting mixture giving a counting rate of 20,000 c/min. was used. The result obtained is in fairly good agreement with that of Curran et al.

A "beaded" counter was built for further examination of the spectrum. This additional step was deemed advisable in view of the somewhat surprising result obtained. This counter consisted of a tube 2¼" in diameter and 24" in length (fig.16b). The central wire was divided into two sections, one 6" long and the other 14" long, insulated electrically from each other by a glass bead, 1 cm. long and 1 mm. in diameter. This provided two independent counting volumes having similar "end-effect", and, as we have seen, the difference between the spectrum in the long and short ends represents the distribution which would be obtained from a counter of length 8" free from "end-effect". Tritium was introduced into the counter, together with a mixture of methane at a pressure of 16 cm. Hg. and argon at 2 atmospheres. The counting rates were about 20,000 c/min. in the long end and 8,000 c/min. in the short end, compared with background counting rates of 600 c/min. and 200 c/min. respectively. The spectrum shape obtained from this examination is also seen in fig.18. The pulse analyser was again used, and the calibration was carried out in the same manner as previously.

Discussion

It should be emphasised that the utmost care was exercised throughout this work to reduce the probable sources of error to a minimum. After the first curve was obtained and its unexpected fall at low energies observed, the experiment was repeated under different conditions. The very good agreement between the results achieved by the differing methods employed is striking. We believe that the curves shown are accurate to within 10 percent down to an energy value of 400 ev., and that even below this energy the shape of the curve is significant. The results obtained are in disagreement with Fermi theory in the low energy region, since the theory predicts that the curve should meet the ordinate at an intensity value about 86 percent that of the maximum.

There are two possible explanations of the experimental results which remove disagreement with Fermi theory. (1) If the counter were not 100 percent efficient for detection of very low energy electrons or if the energy lost by the particles per ion pair produced varied with the energy of the ionizing particles, the true shape of the spectrum could possibly be distorted into a shape similar to that obtained. (2) If there were present, in coincidence with the beta particles, quanta or particles of low energy, the results could again be explained.

Fortunately, sufficient data is at hand to discuss the first possibility. The work of Kirkwood, Pontecorvo and Hanna (1948) together with that of Curran, Angus and Cockroft (1949 a) seems to establish that in argon, the energy expended in the production of an ion pair is constant down to very low energies. This has been discussed in Part II. Additional work carried out with the counter shown in fig.16 (a) has confirmed this result. The K and L X-rays of chlorine (energy 2.6 KeV and 200 ev. respectively) resulting from K- and L-capture in A^{37} are largely converted by Auger effect. It is found, by using a source of A^{37}, that the energy expended per ion pair by slow electrons in argon is constant down to 200 ev. It has also been found that the relative intensities of the two peaks are in agreement with the results of Pontecorvo, Kirkwood, and Hanna (1949). The low energy peak consists of two unresolved peaks, one due to the K-quantum escaping from the counter, involving the L_{II} or L_{III} shell (200 ev.) and one due to nuclear L-capture involving L_I electrons (280 ev.). From these results, it seems most unlikely that the counter used is failing to act proportionally down to the lowest energy values.

In connection with the second possibility, a homogeneous quantum radiation of energy $\sim$ 400 ev. in coincidence with the beta particles would explain

the spectrum shape. Such a radiation would give a peak (Curran, Cockroft and Angus 1949) of shape approximately represented by $x^{20} \exp(-x)$, and this radiation, together with the beta rays, would give a distribution similar to that observed. The origin of such radiation, however, is difficult to explain theoretically. We know that quanta of total energy about 60 ev. will be released in the de-excitation of the He^3 ion following beta emission from H^3. These quanta could give rise to about 3 ion-pairs per disintegration, but we require about 13 ion-pairs on the supposition that radiation is responsible for the shape of the spectrum observed. It is extremely improbable that the radiation has its origin in nuclear excitation of He^3 in the normal sense of the term. It is also possible that a number of low energy quanta (energy $\sim$ 50 ev.), released in coincidence with each other and the beta particles, could account for the results.

Coincidence measurements

The presence of quanta of energy $\ll$ 400 ev. is extremely difficult to establish in a coincidence measurement, due to the extreme absorbability of the radiations. However, the following method was adopted in an endeavour to ascertain whether or not such radiations were present. A counter of $5\frac{1}{2}$" diameter and 16" in effective length (fig.16 (c))

was divided into two similar but independently operating volumes by a very thin glass bead (~ ½ mm. in diameter and 1.5 cm. long). The counting tube was lined with aluminium sheet. Tritium was introduced into the counter with a trace of He to act as carrier and methane was added to give a total pressure of 1 cm. Hg. The counting rate was ~ 10,000 c/min. in each volume, and the counter operated satisfactorily at this low pressure with about 1.5 KV on the case, giving a high gas gain sufficient to detect pulses due to radiation of energy as low as 40 ev. The value of the mass absorption coefficient of methane for low energy quantum radiations does not appear to be accurately known, but from the data available it seemed likely that if quanta of energy 400 ev. were present, coincidences would be obtained. On the other hand, if the quanta had an energy value much less than 400 ev., no coincidences would be recorded. The counter was placed with its axis perpendicular to a magnetic field of sufficient strength to ensure that there were no $\beta - \beta$ coincidences due to an electron travelling from one counting volume to the other. The results obtained from this investigation were indecisive. As the counting tube volts were increased the two counting volumes lost their independence and coincidences due to "mutual firing" were recorded. The phenomenon increased in frequency as the gas gain was increased. A curve

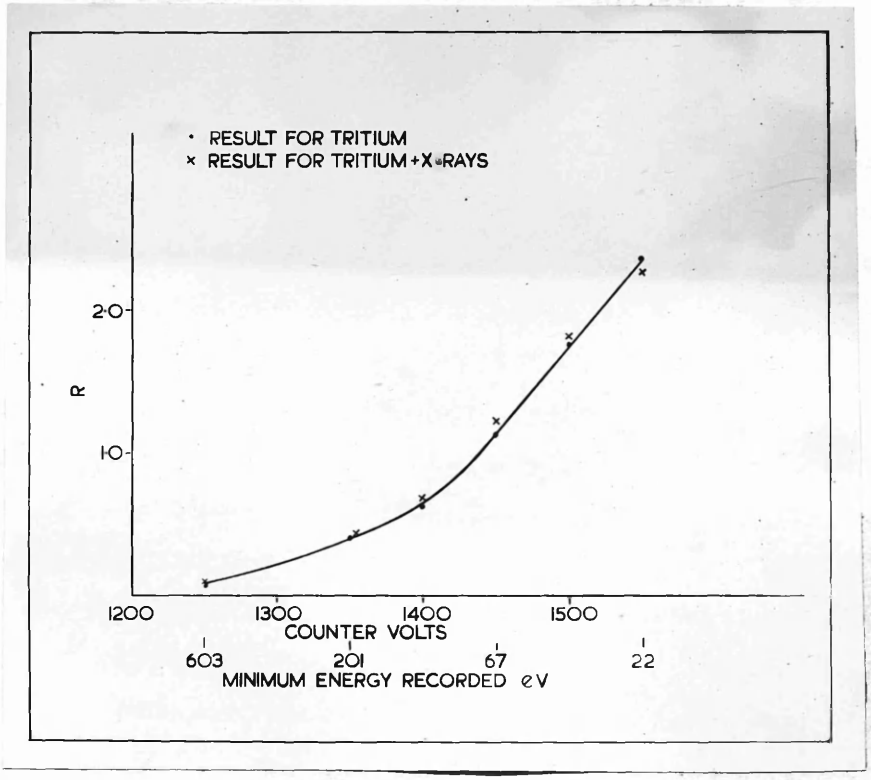


Fig.20. - Results of coincidence measurements.

giving the ratio R of the coincidence counting rate to the total counting rate in terms of the case volts was plotted (fig.20). This curve rose steeply at high gas gain. K α fluorescence X-rays of copper were fired through the central window, resulting in an approximate doubling of the counting rate in each volume. An exactly similar coincidence curve was again plotted (also fig.20) and it was found that the points on this curve overlapped the points of the previous curve to within the accuracy of the experiment. The X-rays of copper gave pulses of average size roughly the same as those produced by the tritium beta rays. The close similarity of the two sets of observations shows that very few, if any, of the coincidences, observed with tritium alone, can be ascribed to the hypothetical beta-quantum decay process. It is unfortunate that this coincidence technique fails to give definite results in the region of 400 ev. Clearly, it makes the technique quite inadequate for testing the hypothesis of much softer quanta ($\ll$ 400 ev.).

We are, thus, forced to conclude that the spectrum of H^3 as observed in a proportional counter, is not in accordance with Fermi theory at energies below about 1 KeV. We are unable to establish experimentally the reason for the discrepancy, although it seems possible that it might have its origin in the simultaneous emission of a quantum or quanta of total energy about 400 ev.

This value is considerably higher than the expected energy release ($\sim$ 60 ev.) in the de-excitation of the H^3 ion.

Since the completion of this work we have devoted much thought to the problem of finding a satisfactory explanation of the apparent disagreement with Fermi theory exhibited by the measured spectrum. A brief outline of some of our ideas is contained in Appendix III.

CHAPTER III

The beta spectrum of RaD

The main features of the disintegration of RaD have not yet been well defined, despite a number of investigations with this source. The presence of a relatively large number of gamma rays, coupled with the fact that the radiations, both particle and quantum, are of low energy, has made the task of finding the complete decay scheme a formidable undertaking. However, it is believed that the principal mode of decay lies in the emission of a very low energy beta particle, leaving the residual nucleus, RaE, excited to an energy of 46.7 KeV. Kinsey (1948) and Cranberg (1950) conclude that about 75 percent of the disintegrations can be thus explained. The beta spectrum has not been isolated, mainly because of the extreme softness of the radiations. Most observers (Richardson and Leigh-Smith 1937; see also Nuclear Data N.B.S. 499) agree that the beta rays are very soft, but apart from this little is known about the transition. There has not even been agreement about the limiting energy, estimates ranging from $\sim$ 15 to $\sim$ 40 KeV. Some observers (Kinsey and Cranberg) have mentioned the suggestion that the spectrum may be complex, having a second harder component, but this has not been verified experimentally.

As we have mentioned, the decay gives rise to a rather large number of gamma rays. These are

fairly low in intensity and have energies of 7.3, 16.1, 23.2, 31.3, 37, 42.6 and 46.7 KeV. Feather (1949), in his summary of the knowledge of the decay, discusses possible groupings of these quantum energies. There has recently been a number of investigations relating to this part of the decay and the work of Cranberg (1950), Frilley et al (1951), and Butt and Brodie (1951) has improved our knowledge in this respect. However, they have not been concerned with the object of the present investigation, which is the examination of the beta spectrum.

The existence of the 46.7 KeV gamma ray has been demonstrated by detection of both unconverted gamma radiation and natural photoelectrons from the L, M and N shells of RaE. The presence of these photoelectrons enables us to study the beta spectrum using the integrating features of the proportional tube (Wilson and Curran 1951). This technique consists of examining the beta spectrum which is "carried forward" by the time coincident photoelectrons. The fact that the photoelectron peak "carrying" the beta spectrum has a finite width is a disadvantage, but not a serious one.

Preparation of the source.

The disintegration of RaD has a half life of 22 years, giving RaE which emits high

energy beta particles with a half life of 5 days, resulting in RaF which decays by α -emission. Hence, it is clear that sources of RaD free from RaE and RaF must be used since these latter would contribute very large disturbing pulses in the counter.

Two different methods were to be adopted to obtain a relatively clean source. The investigation of the usefulness of the first method was not completed because of the success achieved in the application of the second. However, since both techniques are of interest, they shall be described here.

The first method consisted of introducing RaD, in the form of lead tetraethyl vapour, into a glass vessel. This vapour had associated with it a small quantity of RaE in the form of bismuth triethyl. It was decided to heat the mixture to a temperature above the dissociation point of bismuth triethyl (107°C), and to pass the gases remaining after this heating down a long tube with a number of glass wool plugs to help catch any bismuth escaping from the vessel. The lead tetraethyl, associated at this stage with ethylene, was then to be admitted into a vessel which contained a small platinum ball, heated by an R.F. induction furnace, and supported at about 3 mm. distance from a thin sheet of mica. The lead tetraethyl would be dissociated and RaD collected on the

mica sheet. However, as we have mentioned, this experiment was not completed.

The second method consisted of essentially the same procedure as that described by Cranberg (1950). RaD in the form of $PbCl_2$ solution in doubly distilled water was left in a tube for about a day, care being taken to ensure that the tube was not disturbed. Small samples taken from the surface of this solution appeared relatively free of RaE, and our almost complete failure to detect any radiation of energy greater than about 70 KeV verified that the source was relatively pure.

The source used for the present investigation consisted of $\sim 20\mu$ gm. of chloride mounted on a thin aluminium foil of thickness ~ 1.6 mgm./cm^2. Other sources were used in preliminary investigations and some of these were mounted on nylon ($\sim 50\mu$gm./cm^2). The counting rate during the present investigation was 2.5×10^4 c/min. when the source was held at an aperture in the cathode providing a spectrometer of 2π acceptance angle. Using this arrangement it was possible to separate the beta rays from the relatively very intense and complex system of photoelectrons, the separation being sufficiently good to permit comparison of the spectrum with Fermi theory.

Apparatus

A properly end-corrected proportional

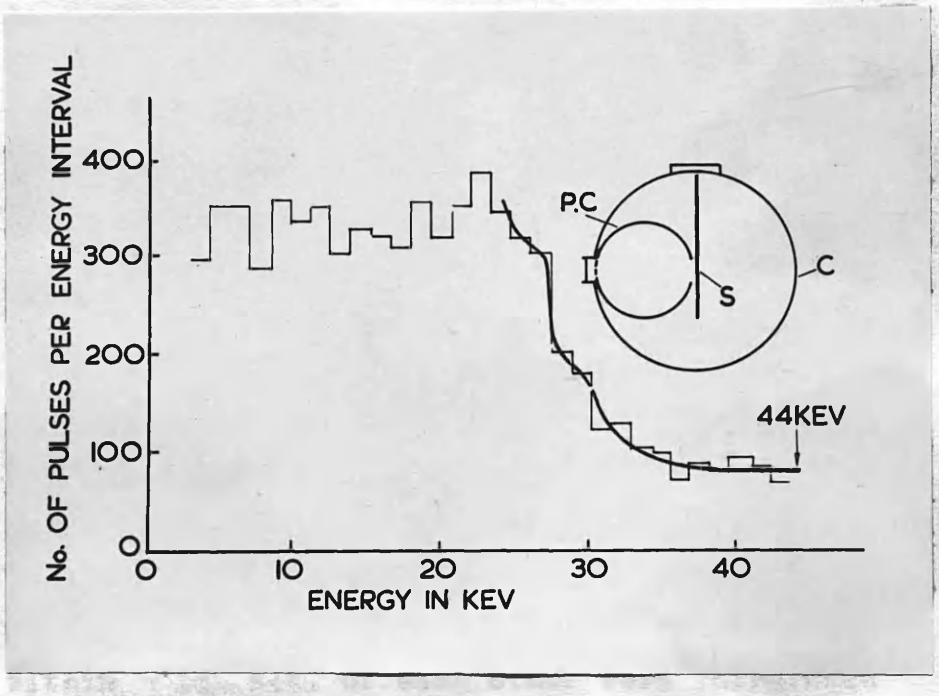


Fig.21. - Pulse spectrum shape between ~ 4 and 40 KeV using source of counting rate 2.5 x 10^4 c/min. mounted on aluminium 1.6 mgm./cm^2. The counter is shown in the insert. The source S is at an aperture in the proportional tube spectrometer P.C.

counter (Cockroft and Curran 1951), 8" in fully operating length and 1.4" in internal diameter, was used throughout this work. It had a central wire of tungsten, 0.004" in diameter, and two diametrically opposite apertures in the cathode. In one of these was constructed a window for the admission of calibrating radiation and the other was used for positioning of the source. The counter was completely enclosed in a cylindrical vessel C, fig.21, which was filled with ethylene to a pressure of 60 cm.Hg. and argon to a pressure of 16 cm.Hg. This mixture was chosen to permit as many as possible of the L X-rays of RaE, resulting from the internal conversion of the gamma radiation, to escape detection. Under these conditions the efficiency for detection of the radiation was < 5 percent. This made the spectrometer responsive chiefly to photoelectrons and beta rays, but it was, of course, sensitive to the very soft M X-radiations. This was somewhat unfortunate, because these radiations were not always detected, due mainly to the geometric arrangement. All ionizing events which occurred within $\sim 10\mu$sec. of each other were integrated by the spectrometer. Hence, if the life of the excitation level at 46.7 KeV does not exceed 10 μsec., the beta particles and the photoelectrons entering the counting volume will be detected simultaneously. In addition M X-radiations will

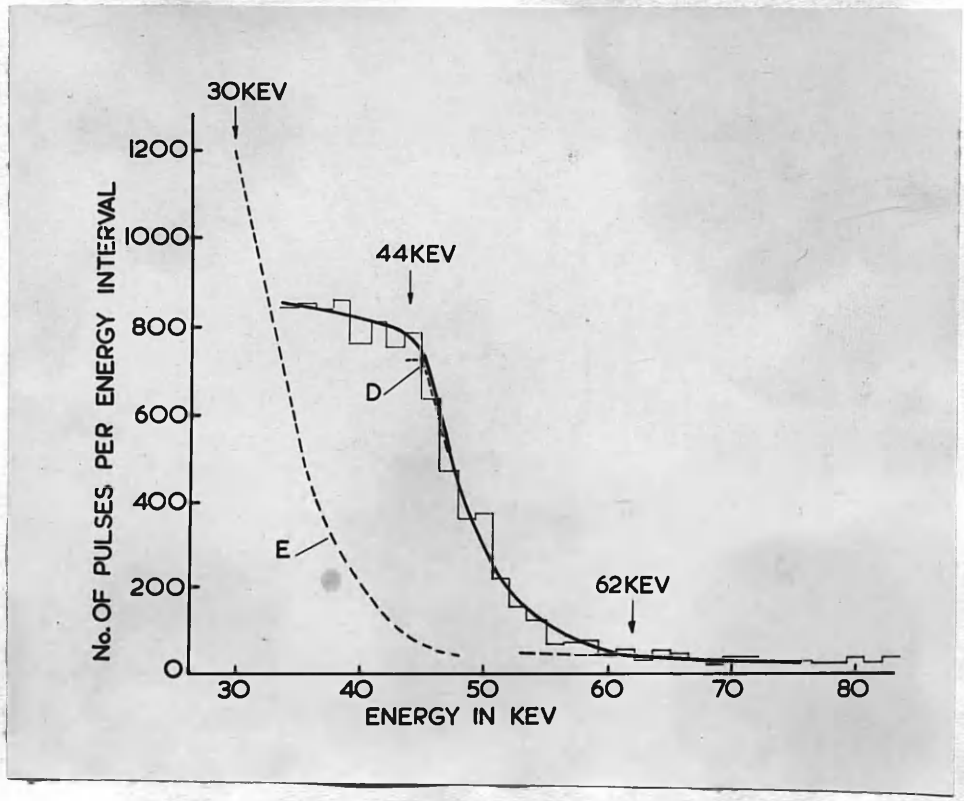


Fig.22. - Curve D shows modification of end of spectrum obtained by using curve F which represents the combination of photoelectrons, ~30 KeV in energy, with beta rays.

sometimes be detected, with the result that the beta particles can be "carried forward" either by a coincident photoelectron alone or by a photoelectron and an M X-ray simultaneously detected. The output pulses from the amplifier were analysed by means of the cathode ray tube and moving film technique.

Pulse distribution.

The total range of the pulse spectrum was covered in two overlapping parts from $\sim$ 4 to 40 KeV (fig.21) and from 33 to beyond 80 KeV. To examine this upper energy portion of the spectrum all pulses of energy less than 30 KeV were blacked out. This allowed us to obtain good statistics and at the same time avoid the darkening of the film which would otherwise result. Both parts of the spectrum were calibrated using the fluorescence Kα X-rays of copper, of energy 8.04 KeV, the amplifier gain being increased by a factor of 4 for both measurements. The region of the spectrum from 33 KeV to beyond 80 KeV is shown separately in fig.22. The overall end-point of the distribution was determined from this and was found to be 62 ± 2.5 KeV. This value was verified by critical examination of the spectrum shape between 50 and 70 KeV with improved statistical data, the measurement being made using sources mounted on both nylon film and thin aluminium. In the case

of the nylon film an extremely thin deposit of aquadag was used to make electrical contact between the source and the cathode of the counter.

Examination of fig.21 shows that, around energy values of 10 and 23 KeV, there are evident definitely significant peaks, superimposed on a nearly flat plateau which extends from 4 to 26 KeV. The remarkable shape of the distribution is in keeping with the results of independent work carried out in this Department by Bannerman and Curran (in the press), who used scintillation spectrometers. Their measurements confirm our results and suggest that the form of the distribution arises from the emission of homogeneous photoelectrons of various energies, produced by internal conversion of soft gamma rays in the L and M shells of RaE, which are frequently emitted in cascade. In our arrangement the beta particles are frequently associated with the photoelectrons and each photoelectron peak can "carry" a beta spectrum. Thus the shape of our distribution is formed by different combinations of beta rays and photoelectrons, the failure to resolve the many photoelectron groups being explained by the smoothing effect of the associated beta rays. However, this low energy part of the curve does not immediately concern us and we do not propose to carry out any detailed investigations in this region.

The photoelectron peaks, resulting from

conversion of the hardest gamma ray, energy 46.7 KeV, in the L and M shells of RaE, will both "carry" the beta spectrum. In the first case the L X-radiation will almost always escape detection, but in the case of M shell conversion, the M X-radiation will frequently be associated with the photoelectrons, and this will lead to uncertainty concerning the exact position of the peak. In addition to making allowance for this, account must be taken of the fact that the photoelectrons may lose energy in the source itself. Realising all this, we expect a distribution of the form obtained. Beyond the main plateau, which we have already discussed, two plateaus of shorter length are clearly visible. These terminate at energies of nearly 30 KeV and about 44 KeV and result from the detection of photoelectrons caused by conversion of the gamma ray of energy 46.7 KeV in the L and M shells of RaE. The smooth fall of the curve above each plateau is ascribed to the association of the beta particles and the photoelectrons. We have already mentioned that, if the life of the excitation level at 46.7 KeV does not exceed 10μsec., beta particles of the continuium and photoelectrons can be detected simultaneously, and the shape of the distribution demonstrates the usefulness of this integrating property of the proportional tube spectrometer. Four combinations of beta particle

and photoelectron are possible:-

(a) beta and photoelectron backward, neither detected;

(b) beta forward into counter, photoelectron back, beta alone detected;

(c) photoelectron forward, beta back, photoelectron alone detected;

(d) beta and photoelectron forward, both detected as a single ionizing event.

Case (b) gives rise to beta rays which become merged in the general distribution at low energies building up the main plateau already mentioned. Case (c) gives a tendency to peak at about 30 and 44 KeV, while case (d) is chiefly responsible for the tails at energies above the peaks. We expect, therefore, that a clear picture of the form of the beta spectrum associated with the level at 46.7 KeV will be obtained by careful study of the shape of the curve from around 44 KeV to 62 KeV. This part of the curve must necessarily represent the beta distribution closely.

Analysis of the beta spectrum.

We have seen that the overall end-point of the distribution of fig.22 occurs at 62 KeV, and this fact, coupled with the knowledge that the photoelectron peak due to M-shell conversion occurs at 44 KeV, gives a value of 18 KeV for the maximum energy of the beta particles. A theoretical Fermi distribution

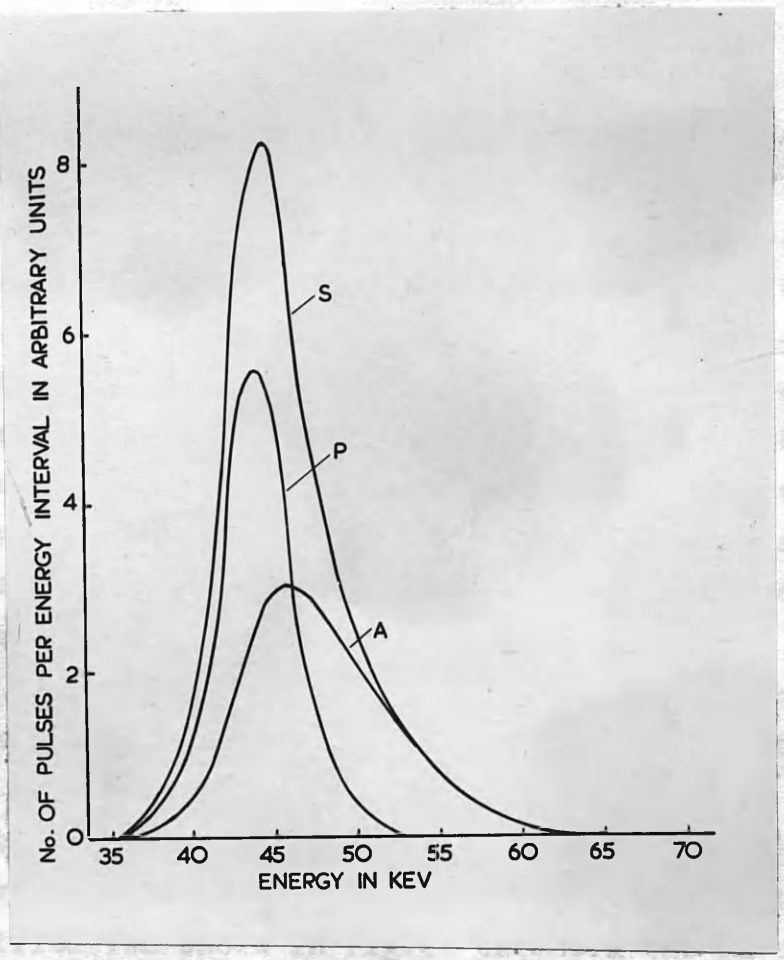


Fig.23. - Illustration of the method of deriving the curve S from curves P and A. Curve P represents photoelectrons alone, curve A photoelectrons + beta-rays and S the composite curve.

F(Z = 83, allowed) for an energy limit E_0 of 18 KeV was evaluated. Because of the complexity of the observed curve, resulting from combinations of cases (c) and (d) above, the curve F has to be modified before making comparison of experiment with theory. The method of making this modification is seen in fig.23. Thus curve P, centred at 44 KeV, represents the form that a homogeneous group of electrons of this energy would take as experimentally observed by the tube. This curve is based on observations of the distribution obtained with gamma rays of energy 46.7 KeV. It was arranged that the areas under P and F (not shown) were equal. Using P and F, and weighting properly in accordance with their shapes, curve A was drawn. This represents the distribution obtained by recording simultaneously any one electron of group P with any beta particle of group F. In our spectrometer P and A, which enclose the same areas, correspond to cases (c) and (d) above. The distribution S of fig.23 is obtained by combining these curves, and is then the derived curve which we shall use for comparison with the distribution shown in fig.22 to check the extent of the agreement of our results with Fermi theory.

In fig.22, therefore, a curve D, represented by the dashed line, is drawn to coincide with the observed distribution from 62 to (62-14), i.e. 48 KeV and below this energy differing from

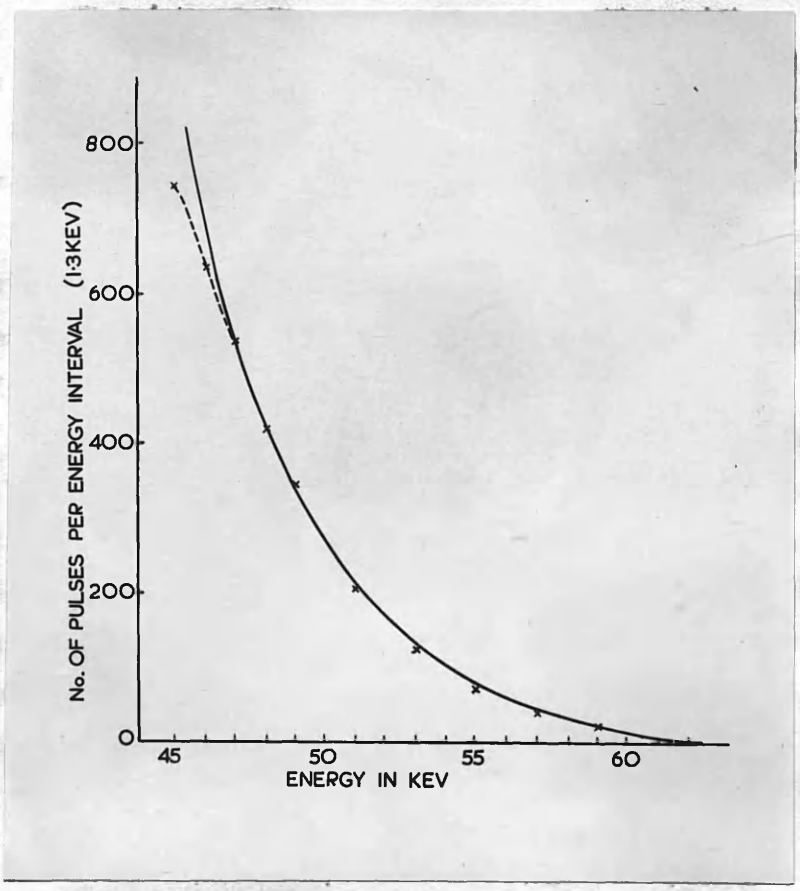


Fig.24. - Comparison of the derived curve S with the experimental results. The experimental points are shown by crosses.

the experimental curve by an amount corresponding to the tail of the curve increased in the ratio $\frac{3}{2}$, since the heights of the 30 and 44 KeV plateaus are as $\frac{3}{2}$.

A comparison of the modified experimental curve D above 44 KeV with the corresponding part of the derived curve S provides us with a final test of the agreement between our experimental results and theory. This comparison is shown in fig.24 and the close fitting of the two curves is very satisfactory. This agreement appears to confirm our belief that the region of the observed spectrum above 44 KeV corresponds to the combination of photoelectrons and beta rays of limiting energy 18 KeV.

In a certain number of the disintegrations the X-rays are associated with the photoelectrons, while in other cases they escape detection. Because of this, the photo-peak is not accurately defined and this makes the value of 18 KeV somewhat uncertain. However, we believe that the limiting energy can be stated within fairly conservative limits by

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

Conclusions.

Our results indicate that the main mode of decay of RaD consists of the emission of beta particles of limiting energy $E_0 = 18$ KeV, leaving the RaE nucleus excited to 46.7 KeV. This value of

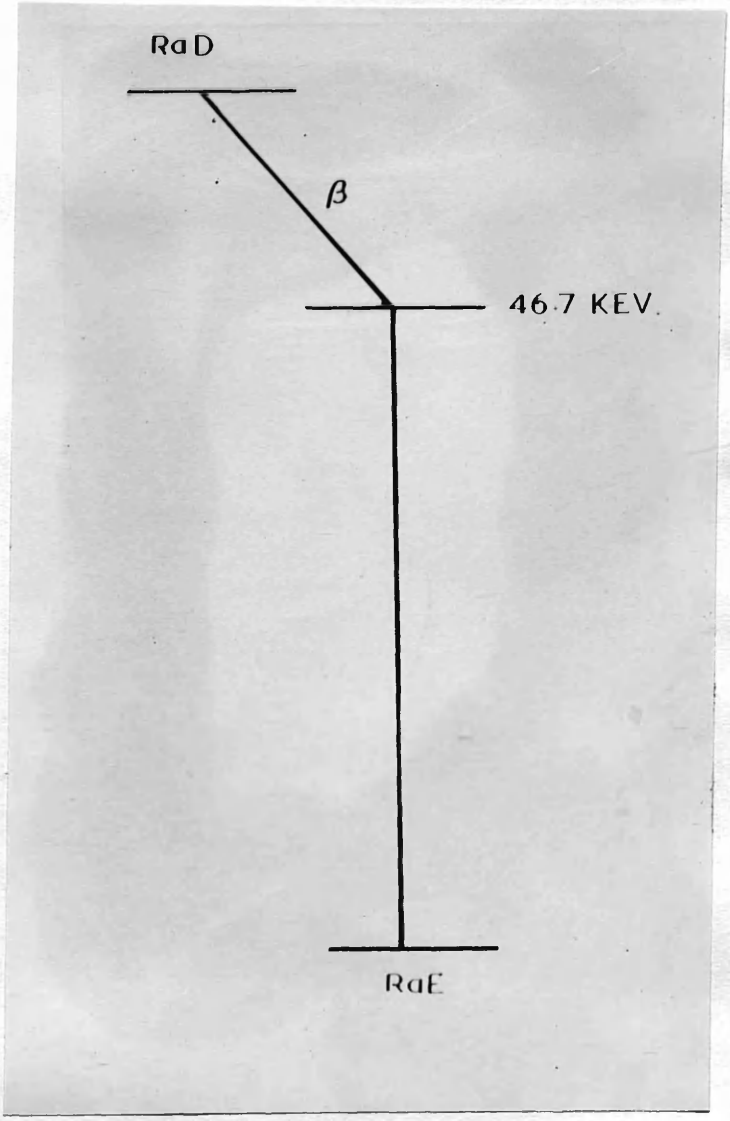


Fig.25. - The main mode of decay of RaD.

18 KeV arises, not only from the curve in fig.22, but also from the results of another investigation using a source on a thin nylon support, when extremely good statistics were employed. Using this limiting energy the value of log ft. for the transition is 5.5. This classifies the transition as allowed unfavoured (Feenberg and Trigg 1950), and not as first forbidden, which follows from the results of previous investigations (Feingold 1951).

Our results give a value of 64.5 ± 2.5 KeV for the disintegration energy of RaD. This is obtained from our knowledge of the limiting energy of the beta spectrum and the fact that the main mode of decay lies in the emission of a soft beta particle, leaving the nucleus of RaE excited to an energy of 46.7 KeV (see fig.25). This level can de-excite in a number of ways. In addition to the direct de-excitation by internal conversion in the L and M shells of RaE to the ground state of this nucleus, frequent cascade processes of internal conversion occur. This explains the rather remarkable shape of the spectrum obtained in our investigation. The integrating feature of the spectrometer results in the recording of photoelectron radiation and Auger electrons associated frequently with beta rays of the continuum. The pulses produced by these build up our spectrum and the beta particles smooth out the distribution, making it impossible to

resolve peaks in the lower energy region. The average energy of the observed spectrum is 18 KeV, but this value is defined almost entirely by the photoelectron radiation and not by the beta rays.

Because of the associated beta rays it is impracticable to conduct any measurements on the probability of direct de-excitation, due to the smoothing of the distribution already mentioned.

We have already mentioned the suggestion of some observers that a second beta ray of higher energy may be present. This hypothesis is in no way supported by our results which indicate that, if any transitions to other levels of RaE do exist, they are of extremely low intensity. This is particularly true of the ground to ground transition of energy 64.5 KeV, since the presence of such a transition would upset our analysis and prevent us achieving the success here reported.

The close agreement between the shape of the observed spectrum and the predictions of Fermi theory is noteworthy. The decay consists of the emission of a very low energy beta particle from a heavy nucleus, and our results indicate that the rather radical modifications of the process which have been suggested for such a case (Frilley, Surugue, and Tsien San-Tsiang 1946 and Ivanenko and Lebedev 1950) are not verified by experiment, at least for this particular transition.

CONCLUSION

We have discussed the advantages of the proportional tube spectrometer over more orthodox instruments used in beta ray spectroscopy. We have also noted that, at the commencement of this work, the instrument had definite limitations, particularly in the range of energies over which it could be usefully employed. The investigations reported here have helped to extend the technique. Improvements to the methods available for calibration of the energy scale, involving the employment of a few suitably chosen radioactive sources which are used to excite the fluorescence radiation of various elements, have assisted in increasing the accuracy of the energy measurements, while the use of magnetic fields to increase the energy range of the instrument is of major importance. It is no exaggeration to state that the proportional tube spectrometer can now be adapted to examine almost all beta spectra. It is true that the resolution attainable cannot compete with that of magnetic spectrometers, but the advantages of the instrument make investigation of the low energy region of beta spectra a more accurate procedure. Ideal investigation of a beta spectrum could be achieved by measuring the upper energy region with a magnetic spectrometer, and the lower energy region by use of a proportional tube in a magnetic field. In this way the good resolution of the magnetic spectrometer is utilised, while the

difficulties experienced in the measurement of low energy electrons are avoided. An example of this is the investigation of the shape of the beta spectrum of Hg^{203} carried out by Wilson and Curran (1951) giving a result which could be related to previous work with magnetic spectrometers.

While the proportional tube spectrometer can be usefully employed for the measurement of almost all beta spectra, it is of particular advantage for examination of sources which can be obtained in the form of a gas suitable for introduction into a proportional counter. It is also a powerful tool for the investigation of sources having a very low activity, since the geometric efficiency of the spectrometer can be utilised. In this respect the advantages of the instrument are considerable.

We have stated that the validity of Fermi theory must be judged by its ability to explain the experimental results. Needless to say, there has been considerable experimentation in this field, and our results have helped to swell the available data, upon which modification of the theory must be based in the event of consistent disagreement. An excellent review of the position as late as 1950 is given by Wu (1950). She emphasises the difficulties experienced in measurements on low energy electrons and concludes that the disagreements which have been found using such sources as S^{35} and Cu^{64} are due

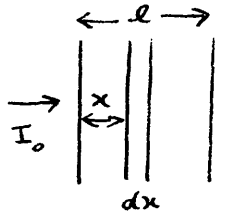
to experimental limitations. This conclusion is based on the most recent results obtained, which lead Wu to believe that the Fermi theory of beta-decay probably does predict the true distribution for electrons and positrons at very low energies (see Wu and Albert 1949 a and b). Our results from measurements using S^{35} are not in agreement with the predictions of Fermi theory. However, we do not exclude the possibility of experimental limitation in this investigation and, as we have mentioned, we hope to re-examine the spectrum shape (see Appendix II).

Because of the simplicity of the tritium nucleus, the shape of the beta spectrum should be a good test of Fermi theory. Previous work (Curran, Angus and Cockroft 1949 b) has resulted in a distribution which agreed with the theoretical predictions from the end-point of the spectrum down to an energy value of $\sim$ 1 KeV. We have extended the measurements down to very low energies ($\sim$200 ev) and our distribution appears to show departure from the theory at energy values below 1 KeV. We have been unable to establish experimentally the reason for this discrepancy, although it seems possible that it might have its origin in the simultaneous emission of a quantum or quanta of total energy about 400 ev. These quanta would be in coincidence with beta particles whose spectrum shape would be in agreement with the predictions of Fermi theory.

Ivanenko and Lebedev (1950) have suggested a new process in beta-decay, in which the beta particle is captured into an unoccupied level of the product nucleus with the emission of a photon. This would reduce the probability of escape of a very soft beta particle from a heavy atom and, consequently, give a spectrum shape in disagreement with the predictions of Fermi theory. The decay of RaD would be a good example of such a process, but our results with this nucleus are in agreement with Fermi theory. Wilson and Curran (1951) also conclude that their results using Hg^{203} do not support the suggestion of Ivanenko and Lebedev.

APPENDIX I

X-ray excitation (transmission case)



Consider a homogeneous beam of X-rays of wavelength λ_1 and intensity I_0 falling on a sheet of thickness l and density p. Let the mass absorption coefficient of the sheet be μ_1 for the radiation.

The intensity at a depth $x = I = I_0 e^{-\mu_1 p x}$

The intensity of modified X-rays generated between x and x + dx is

$$dI = -\mu_1 p I_0 e^{-\mu_1 p x} dx.$$

These modified X-rays of wavelength λ_2 have to penetrate a thickness (l-x) of the sheet which has a mass absorption coefficient μ_2 for the radiation. Hence the number of quanta of wavelength λ_2 escaping from the sheet will be

$$dI\, e^{-(l-x)\mu_2 p}.$$

Let the total number of modified X-rays which escape from the sheet be N

$$N = \int_0^l e^{-\mu_2 p(l-x)} \mu_1 p I_0 e^{-\mu_1 p x} dx$$

$$= K_1 I_0 \left[e^{-K_2} - e^{-K_1} \right] / (K_1 - K_2)$$

where $K_1 = \mu_1 p l$ and $K_2 = \mu_2 p l$.

APPENDIX II

Re-examination of the beta-spectrum of S^{35}

The method which we adopted for the examination of the beta-spectrum of S^{35} was not ideal. Although the gas used, $H_2 S^{35}$, did not appear to demonstrate any appreciable electron affinity, it penetrated behind the stainless steel lining and adhered to the walls of the tube. This did not result in a direct distortion of the spectrum, but since a subtraction method was employed, it was essential to secure within a short time interval all measurements which were to be compared as regards intensity. In addition, due to the rather large range of the particles of high energy, a "wall effect" was experienced. We believe that many disadvantages could be overcome by using $CO S^{35}$ as the counter gas, since this gas has properties which suggest that it should behave well in a proportional tube. The "wall effect" can be easily overcome by the use of magnetic fields to confine the range of the particles. In addition to these two improvements, the use of a properly end-corrected tube will avoid the need for subtraction, thus allowing better statistical data to be obtained.

We propose, therefore, in the near future, to construct a counter of internal diameter 5" and total counting length about 24", which will be properly end-corrected. This will be filled

to a total gas pressure of about 5 atmospheres containing a trace of CO S^{35}. The counter will be placed in the magnetic field with its axis perpendicular to the field direction, and high field values employed. Under these conditions it should be possible to measure the spectrum shape with high accuracy to energy values well below 10 KeV.

APPENDIX III

Some further thoughts on the decay of tritium.

We have seen that the beta spectrum of tritium, as measured in a proportional tube spectrometer, is in disagreement with the predictions of Fermi theory in the energy region below 1 KeV. We have suggested that this discrepancy may be explained by the presence of a quantum or quanta, of total energy about 400 ev., emitted in coincidence with the beta particle, but we have so far been unable either to arrange an experiment to demonstrate the presence of such quanta, or to produce a reasonable theoretical explanation of their emission. However, much thought has been devoted towards finding a solution to this latter problem, and a short outline of some of our considerations is given below.

When the beta particle is emitted the magnetic moment of the nucleus changes. The possibility of magnetic dipole radiation being emitted as a result of this was considered. The form of the distribution which would be observed using a proportional tube spectrometer was derived, assuming that the radiation was in coincidence with the beta particle. Although the exact shape of the spectrum was not obtained, the general form was similar to that observed in the experimental arrangement. However, the probability of this

process seems negligibly small when contrasted with the probability of emission of a beta particle unaccompanied by quanta. For this reason, the consideration of this process was abandoned.

A more encouraging idea lies in the possibility of a process suggested by Dr. S.C. Curran. The nucleus of tritium consists of one proton and two neutrons. When the beta particle leaves the nucleus we have two protons and one neutron. Let us consider the repulsive forces between these two protons. The energy of each nucleon is 2.8 MeV, and this corresponds to a velocity of 2.3×10^{9} cm./sec. Let us assume that the nucleon moves a distance of 10^{-13} cm. with this velocity. This enables us to obtain an estimate of the time factors involved. If it moves for a time Δt

$$\Delta t = 4 \times 10^{-23} \text{ sec.}$$

The Coulomb force between the two charges is

$$F = e^2/r^2 = (5 \times 10^{-10})^2 / (10^{-13})^2 = 2.5 \times 10^{7} \text{ dynes.}$$

The acceleration of one of the nucleons which results from this force is

$$\alpha = F/m = 2.5 \times 10^{7} / 1.8 \times 10^{-24}$$

$$= 1.3 \times 10^{31} \text{ cm./sec.}^2.$$

It will produce a change of velocity given by

$$\alpha \times \Delta t = 5 \times 10^{8} \text{ cm./sec.}$$

As this change of velocity is ~~extremely~~ rather small when compared with the mean velocity of the nucleon it may be neglected. Now this acceleration of the proton in the nucleus will lead to the emission of radiation.

The energy radiated per sec. $= \rho = 2e^2\alpha^2/3c^3 = 10^{12}$ ergs.[x]

The energy radiated in the time Δt will be

$$E = 10^{12} \times 4 \times 10^{-23} \text{ ergs.}$$

$$\doteq \underline{30 \text{ ev.}}$$

Let us now assume that the nucleon moves in such a fashion that it traverses a distance of 2×10^{-13}cm.

In this case $\Delta t = 8 \times 10^{-23}$ sec.

Let us also assume that the value for r in the case is $\frac{1}{2} \times 10^{-13}$ cm. F will increase by a factor of 4, making α increase by the same factor, and ρ, which involves α^2, will increase by a factor of 16.

In this case $E \doteq \underline{800 \text{ ev.}}$

It appears, therefore, that this hypothetical process can be responsible for the emission of a radiation of about the correct value of energy, and for this reason is well worth considering. It is true that some of our assumptions are a trifle bold, but the possibility of this process interests us.

[x] This equation is Classical.

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