THESIS FOR THE DEGREE OF Ph.D.

Experiments on Some &-Ray Bodies

.

An Absorption Study of Simple β -Ray Spectra representing Allowed, Second Forbidden and Third Forbidden Transitions

(With Appendices)

by

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1. INTRODUCTION

The absorption method was the earliest way in which the A-ray spectra of radioactive materials were studied, initially, using electroscopes as recording instruments, these being later replaced by ionization chambers, and ultimately by Geiger-Müller counters. The development of the cloud chamber and magnetic spectrograph tended to make it less popular for some years. especially since the latter methods provide an energy distribution of the A-particles: they both require fairly strong sources however, and analysis of data obtained with the help of the cloud chamber is a very tedious procedure. The absorption method, on the other hand, is capable of yielding valuable information with sources of strength less than 1 microcurie. The discovery of artificial radioactive materials, of which only weak preparations were available initially, made it necessary to return to the absorption method, and in recent years the improvements in counter technique and recording apparatus have revived considerable interest in it. The method is also useful when the half-value period of the source material is short, since the β -particle intensity can be measured within a few seconds of the preparation of the source; this is a more difficult task with a magnetic spectrograph when the chamber must be evacuated after insertion of the source, or the active material must be introduced through a vacuum-tight seal.

For many years, absorption determinations have been made of the maximum energies of the β -particles emitted by various substances, especially where very little γ -radiation is present.

Feather (1) and Sargent (2) have also proposed methods of obtaining the same information in the presence of appreciable γ -radiation if sufficiently accurate measurements are made; this is a relatively simple task with the aid of modern counting techniques,

Bleuler and Zünti (3) have criticised Feather's method in a recent paper and propose one depending on a comparison of a new absorption curve, with empirically calculated curves. Their method is open to criticism, however, on rather similar grounds to those raised by Feather (1) regarding the method of Widdowson and Champion (4) in which absorption limits are deduced by means of a standard polynomial, viz; such a procedure can be adopted only if the β -particle spectrum is simple, and rules out the possibility of detecting differences in spectral type even amongst such simple spectra. Further comments on Feather's method by Bleuler and Zünti are discussed later, in the section devoted to the analysis of experimental absorption curves.

The problem of deriving an energy spectrum from absorption measurements has not so far been discussed in detail in the literature. Sargent (5) has proposed a method of deriving absorption curves from calculated energy distributions backed by limited information obtained with a simple form of magnetic spectrograph. His energy distribution curve for Radium E does not fit more recent data obtained with magnetic spectrographs, and the remaining curves must therefore be accepted with reservation, although he claims that all, including that of Radium E, give absorption curves very similar to those obtained experimentally. He works back from a trial distribution to the absorption curve, stating

that the problem of finding the distribution curve from an experimental absorption curve cannot readily be solved mathematically. It is probable that the low accuracy of his data, obtained by ionisation chamber measurements, further discouraged him in this object.

The methods of Feather and Sargent give an indication of the energy distribution of the &-particles from a substance, but do not provide a distribution curve; both compare the absorption curve of the body under investigation with that of a standard β -ray emitter and show the intensities of the different energies of β -particles relative to the corresponding intensity in that part of the spectrum of the standard body. If it is required to find the absorption limit of the β -particles of the material under investigation, the former's method is to be preferred, both from the point of view of simplicity and accuracy: the latter's method is probably better if the qualitative differences in spectrum are to be displayed. Feather's method provides precisely the same information, however, with considerably less calculation, although the presentation is a little less obvious. The chief objection to Sargent's method is that it involves fitting the absorption curve to the standard curve. Until recently it has been believed that the absorption curves of a great number of radioactive materials have the same shape of β -spectrum from about half of the absorption limit to the absorption limit and it is on this basis that the absorption curves are fitted. The work described below, shows, however, that if the curves of Actinium C" and Radium E are fitted they are coincident over only the last third of the & -particle range; thus, unless accurate

measurements are made up to a range fraction of about 0.9, there is considerable danger in applying Sargent's method. Feather's method, on the other hand, does not rely on any such coincidence of parts of the absorption curve, the only assumption necessary being that of the maximum range, in the absorbing material, of the β -particles emitted by the standard substance.

In a recent paper, Das Gupta and Chaudhury (6) describe a method of deriving the energy distribution curve from the absorption curve, and compare their experimental results for Radium E with the theoretical predictions of the Fermi and the so-called Konopinski-Uhlenbeck theories (the latter theory has since been retracted by its originators). This is the first serious attempt at the solution of the problem, but their distribution curve is open to doubt. They claim that their results indicate that Radium E β -particles follow closely a K.U. form of spectrum, but their formula necessarily gives a very large deviation at low energies. There is a pronounced scatter of experimental points about the theoretical curve, and the accuracy of their absorption data is probably not sufficiently high to warrant such treatment. In the work described later, an attempt is made to derive energy distribution curves, but the distribution obtained for Radium E departs markedly at low energies from the theoretical curves and spectrographic determinations. Hughes, Eggler and Huddleston (7) describe a similar method, but compare the distributions obtained from the absorption curves with those obtained by spectrographic methods for well-known spectral shapes. Their work is applied to Be.

The deviations between the β -ray spectra of bodies undergoing

allowed and forbidden transitions are sufficient to warrant a direct comparison of representative elements since there is, as yet, very little published material on the subject, compared with other aspects of β -particle emission.

The majority of the work, so far, has been carried out by means of magnetic spectrographs (e.g. Martin and Richardson (8)), but there is no record of an absorption investigation of the problem, probably because sufficiently accurate absorption measurements have not been made to enable an energy distribution to be obtained from the absorption curves. The extension of the absorption method to this work is therefore desirable, since it allows investigation of radioelements from which sources can not yet be made sufficiently strong for spectrographic investigation; Actinium C^m is a typical material of which strong sources cannot be obtained at the present time and was chosen as a representative of an allowed transition in the work described later.

It is a well-known fact that absorption spectra measured on different apparatus cannot legitimately be compared, since differences in solid angle and scattering conditions can cause appreciable variations in spectral shape. It is essential, therefore, that precisely the same conditions must prevail for all the sources used, if a valid comparison is to be made. In the work described in this paper, strict attention has been paid to this fact and an effort has been made to reduce back-scattering by mounting the sources on perspex.

The following account describes experimental work leading up to, and a direct comparison of, the β -ray absorption spectra of three β -particle emitters, Actinium C", Radium E, and Thallium

(3.5 yr.) of similar atomic number (81, 83 and 81 respectively), known to give simple spectra associated with very little or no Y-radiation, and undergoing allowed, second forbidden and third forbidden transitions respectively. The absorption limits and maximum energies of the β -particles from Actinium C" and Thallium were determined by Feather's method, taking Radium E as the standard substance and new values are proposed for both materials. Finally, an attempt is made to derive energy distribution curves from the experimental absorption curves, for all three materials.

2. APPARATUS

All the electronic equipment used in the experimental work was designed by TRE/AERE and consisted of the following:

(a) The Stabilized high voltage supply for the Geiger-Müller

counter was a Dynatron Power Unit Type 200, providing a stable positive or negative output voltage of up to 4,000 volts in four ranges, viz. 170-500, 400-1,000, 800-2,000 and 1,600-4,000. The desired range was selected by a switch and the voltage was continuously variable within the range, by means of coarse and fine control knobs. The voltage was read directly on a meter, the full scale deflection of which was the maximum voltage of the selected range.

The regulation was such that variations of \pm 10% in the mains input voltage produced variations in output voltage of not more than \pm 0.66%.

(b) The Scaling Circuit was a Dynatron Scaling Unit Type 200 A.

consisting of a hard-valve scale-of-hundred with meon indicators, followed by a Post Office call meter. If desired, the latter could be disconnected by a switch, and the output of the scale-of-hundred fed to an output plug. The internal connections of the input circuit could be altered by a switch to provide two different functions, (i) a D.C. coupled snap circuit (for use with ionization chambers), with a resolving time of 5 microseconds or (ii) an A.C. coupled flip-flop (for use with Geiger-Müller counters) providing a paralysis time which could be preset to a range of values, nominally stated to be between 300 microseconds and 1000 microseconds for a pulse of about 5 volts amplitude, but extending

below and above these values if larger or smaller input pulses were used. The paralysis time on the G.M. (Geiger-Müller) input was designed to reduce multiple counts, but the scaler could be operated by a counter, on the I.C. (Ionization Chamber) input. (c) A Probe Unit Type 200. (later replaced by a Probe Unit Type

1014A, as described in the text). This was a single valve amplifier, and included the H.T. components associated with the Geiger-Müller counter; the gain of the amplifier was variable with a maximum of about 35 times. The Probe derived its power supplies from the Scaling Unit and fed the amplified counter pulse back to the scaler through the same 6-way screened cable; a second screened cable carried the high voltage supply from the Power Unit to the Probe and the counter was connected to the Probe by a short length (about 6 inches) of single-way screened cable. The advantage of the Probe Unit was that it allowed a greater length of cable from the counter assembly to the scaling unit, enabling the former to be placed in the most convenient position for working; without the Probe Unit, the maximum length of cable permitted was 2 feet, since greater lengths caused excessive attenuation of the counter pulses.

The Geiger-Müller Counter was a G.E.C. Type G.M.4, the body of which was copper, 1" in diameter and $2\frac{1}{6}$ " long. The wire was supported from the top of the tube and entered through a metal-to-glass seal. The end was flanged and covered by a duralumin window $6mg/cm^2$ thick.

The source holders and the rigid assembly holding the counter, source and aluminium absorbers were made from perspex, to reduce spurious effects due to scattering; Yaffe (unpublished) has





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recently found that back-scattering is increased by heavy backing materials and that, for a given material, it increases to a "saturation" value as the thickness of the backing increases. He points out also that the different backing materials used by various workers can have an appreciable effect on the results of β -ray investigations.

Throughout the experiments described in this paper, precisely the same conditions of geometry and scattering prevailed, so a valid comparison can be made for the three source materials.

The source holders (shown in Fig. 1) were machined from a solid piece of perspex, $\frac{1}{4}$ " thick and 1" square. A pin, $\frac{1}{2}$ " in diameter and $\frac{3}{32}$ " thick, was turned on the underside, and a recess, $\frac{1}{2}$ " in diameter and $\frac{1}{64}$ " in depth, with vertical sides and a smooth bottom, was turned in the top. A square holder was used in order to avoid rotation of the source and to ensure that it was always replaced in the same orientation. The pin fitted into a hole drilled in the perspex base of the counter assembly and rotation of the source was prevented by a strip of perspex cemented to the base.

The first counter assembly (Fig. 2) was built up from $\frac{1}{4}$ " perspex. The source holder fitted into a hole in a 4" square base plate; above this, supported by two vertical pieces, 1" deep, was a second plate, drilled centrally with a $\frac{1}{2}$ " diameter hole, to which two more vertical pieces, $\frac{5}{8}$ " deep and 2" long, were comented. To the top of these was sorewed a further plate, 2" x $2\frac{1}{2}$ ", drilled centrally with a 1" diameter hole. Before the top plate was sorewed on, the counter was inserted through this hole, from below, and the flange clamped by a 2" square plate of $\frac{1}{8}$ " perspex, the latter being

held by four screws passing through the top plate. The hole in the clamping plate was $1\frac{3}{16}$ in diameter, and bevelled on the inside edge to reduce scattering; the heads of the fixing screws were countersunk below the surface of the plate so that the counter could "see" no metal. The space between the base and first plate was large enough to allow easy removal and replacement of sources, and the space above the first plate received the absorbers. A lead turret of $1\frac{1}{2}$ " wall thickness was constructed round the counter.

3. PRELIMINARY INVESTIGATIONS

Preliminary routine measurements of plateau, resolving General. time and background of the counter were carried out, the plateau being found to agree very closely with that supplied by the makers. The amplification of the probe unit was set at the minimum value necessary to operate the scaler, which was initially used on the G.M. input (with paralysis). Measurements of the resolving time, using a method described by Beers (9) gave rather a high value (1,200 microseconds), but preliminary measurements were continued, using a Thallium source, in the counter assembly described above. The Thallium was chosen for the first experiments on account of its fairly long half-value period (3-5 years); the source material was in the form of Thallium Oxide, activated by neutron bombardment in the pile at A.E.R.E., Harwell. A thin layer of the material was spread uniformly in the recess in the top of one of the source holders and cemented in with a drop of a thin solution of distreme in benzene.





A set of aluminium absorbers was accurately calibrated by weighing and measuring the area of each.

The first absorption measurement is illustrated in Fig. J(a). Corrections have been applied for resolving time, background, air thickness between the source and the counter, and for the counter window; the error in each point, on the vertical scale, is 1%. A marked rise at the low energy end is evident, a rather unexpected result. The large corrections for resolving time were thought to be a possible cause for this, and accordingly a second set of readings was made with a much weaker source, to reduce the corrections; this, however, was of the same form.

A test of the plateau of the counter, using the I.C. input of the scalar showed no evidence of spurious pulses at the lower end of the plateau, so it seemed safe to use this input. The average value of 7 determinations of the resolving time of the counter was 118.9 ⁺ 19.0 microseconds and the individual values were consistent; the resolving time was accordingly taken as 119 microseconds.

Further absorption measurements using strong and weak sources still gave the rise at the low energy end of the absorption curve, discounting the possibility that resolving time corrections were the cause.

The possibility of scattering at the hole in the upper large perspex plate was reviewed, and also that of the absorption of β -particles by the solid parts of the plate coming within the solid angle subtended by the counter at the source. A new counter assembly was constructed, being, in effect, the part of the first assembly above the upper 4" square plate; it is shown in Fig. 4.









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Clamping Plate



Enlarged section showing clamping of counter

The vertical pieces supporting the counter plate were comented directly to the base plate with chloroform, alignment of counter and source being ensured by using a brass jig turned at one end to fit the counter hole in the top plate, and at the other to fit the recess into which the pin on the source fitted. After the chloroform had dried, the top plate was unscrewed and the jig removed. Two strips of persper, 2" long, $\frac{2}{16}$ " thick, and slightly under $\frac{1}{2}$ " wide, were then stuck to the base plate alongside the vertical pieces; these prevented rotation of the source, and also supported the absorbers, leaving $\frac{1}{32}$ " elearance between the absorbers and the top of the source holder. The possibility of these absorber supports causing scattering into the counter was very small.

This assembly gave an absorption curve of the expected form (Fig. 3(b)), without the rise at the beginning. In order to verify that the rise with the first assembly was not due to some effect decaying with time, a repeat determination was taken with it, after some days, resulting in precisely the same curve as before. As a further test, a $\frac{1}{6}$ " thick perspex sheet, drilled centrally with a $\frac{1}{2}$ " diameter hole, was placed in the absorber space of the second assembly; the resulting curve (Fig. 3(c)) showed a tendency to rise in the same way as those taken with the first assembly. Finally, the top plate of the first assembly was cut away to give a $1\frac{1}{2}$ " square hole instead of a $\frac{1}{2}$ " diameter round hole, the curve (Fig. 3(d)) then being of the form obtained with the second assembly.

This proved conclusively that the unusual beginning of the first absorption curve was due to the counter assembly and

illustrated clearly the spurious effects which can arise if a poor design of counter-source assembly is used.

The second assembly (Fig. 4) appeared to be free from the faults of the first and was adopted for all subsequent work. Inconsistencies in Counting Rates. A check of the resolving time of the Geiger-Hüller counter gave a value of 80 \pm 4 microseconds, which seemed rather low and was not in good agreement with the previous value of 119 \pm 19 microseconds.

The formula used in calculating the resolving time, o, was $\sigma = \frac{c_1 + c_2 - c_{12}}{2c_1 c_2} + \frac{1}{8c_{12}} \left\{ \frac{(c_1 + c_2 - c_{12}) c_{12}}{c_1 c_2} \right\}^2$ (a more convenient form for calculation is $\sigma = \sigma_1 + \sigma_2$ where $\sigma_1 = \frac{c_1 + c_2 - c_{12}}{2c_1 c_2}$ and $\sigma_2 = \frac{1}{2} C_{12} \sigma_1^2$; C_1, C_2 are the observed counting rates with each of two sources, and C12 is the observed counting rate due to both sources (9). The accuracy of the value obtained for σ_s clearly depends principally on the accuracy of $(C_1 + C_2 - C_{12})$. The best order of readings is as follows: the counting rate is taken with the first source followed by the counting rate for both sources together; the first source is then removed and the counting rate taken for the second source. In this way, there is no question of replacing the sources in precisely the same position. A repeat determination using two radium sources was made, the values of C1, C2, and C12 being found to better than 0.2%. In order to provide a check on the consistency, each of the final values of C1, C2, and C12 was made up of ten separate counts, taken for the same position of the source. The resolving time obtained from the averages of the 10 readings in each set was 38 µ sec, which seemed too low. The consistency of the three sets of 10 readings was investigated and revealed considerable differences between the

$\frac{N}{N} = \text{number of counts recorded in 2 minutes} \\ \overline{N} = \text{mean value of N} \\ \Delta = \text{deviation of N from } \overline{N} \\ \text{Standard deviation of n readings } = \sqrt{\left(\frac{\Sigma \Delta^2}{n-1}\right)} \\ \end{array}$						
C,		0	12	Cz		
N	Δ	N A		N	Δ	
50934	-1511	90340	-663	40124	+290	
51572	-873	90028	-945	40418	+584	
52734	+289	90451	-522	39849	+ 15	
52612	+167	91522	+549	40108	+274	
52586	+141	91287	+314	40045	+251	
52796	+351	91693	+720	39423	-411	
52953	+508	90715	-258	39707	-127	
52556	+111	91163	+190	39369	-465	
52735	+290	91570	+597	39630	-204	
52972	+527	90961	- 12	39664	-170	
$\vec{N} = \frac{1}{\Sigma} \left(\vec{N} \right) = \frac{1}{\Sigma} \left(\frac{\Sigma}{\Delta^2} \right) = 3$	52445 229 931956 661	$\overline{N} = \int (\overline{N}) = \int (\underline{\Sigma} \Delta^{\mathbf{Z}}) = \int (\Sigma$	90973 302 2943812 572	$\overline{\mathbf{N}} = \sqrt{(\overline{\mathbf{N}})} = \frac{\sqrt{(\overline{\mathbf{N}})^2}}{\sqrt{(\underline{\mathbf{L}}\mathbf{C}^2)}} = \frac{\sqrt{(\underline{\mathbf{L}}\mathbf{C}^2)}}{2}$	39834 200 1035249 339	

TABLE 1

×.,

standard deviation and the square root of the mean counting rate. These two values should be approximately equal, and if a sufficiently large number of readings is taken, the standard deviation should be slightly less than the square root of the mean, In all three cases, the standard deviation is considerably the greater: the sets of readings are given in Table 1. It was clear, therefore, that there was some source of error more important than the statistical fluctuation. The most obvious possibility was that of timing, which was done by stop-watch. The watch was started and the count switch depressed simultaneously at the beginning of a count, and both were operated together at the end of the count. The personal error in each of these operations was estimated to be not more than $\frac{1}{5}$ sec., which was also the accuracy of the watch. The internal operation of the switch appeared to be slower on the make than on the break, and it was thought possible that a combination of these sources of error might be influencing the results, although tending to the generous side and estimating the maximum error due to both, as $\frac{1}{2}$ sec, this represented less than 0.5% error in the times for the set of readings given in Table 1, since each was taken for 2 mins. In an attempt to verify that timing errors were the cause of the inconsistencies, two sets of readings were taken using a Thallium source in the second perspex counter assembly described earlier (Fig. 4), the first timed by hand (stop-watch) and the second by a Timing Unit Type 1003K. A brief description of this unit is given below.

The Timing Unit was designed by T.R.E./A.E.R.E. as an accessory to the Scaling Unit Type 200 and was built by McMichael Radio. It consisted of a group of telephone relays, functioning as two

scales-of-ten in series; a resettable electromechanical register. and a source of time impulses. The unit could be used either to measure the time taken to complete a predetermined number of counts or to give the number of pulses counted in a predetermined time interval. The timing impulses were obtained from a mechanical clock which gave out impulses every $\frac{1}{2}$ sec. and every $\frac{1}{2}$ min. and a relay circuit in the Timing Unit permitted the selection of 1 sec. and 1 min. pulses when required. When used to measure the time for a given number of pulses, the best accuracy was determined by the smallest time interval given by the clock, viz. $\frac{1}{2}$ sec; when used to record the number of pulses in a given time, the latter was determined to less than $\frac{1}{50}$ sec. and the accuracy of timing impulses was better than 0.5%, in addition to which the long term accuracy was good since there was a simple thermostatic control on the clock enclosure. Clearly the unit was more accurate when recording the number of pulses in a given time and was therefore used solely in this manner. Only this function is further described. In this condition the mechanical register was in parallel with the register built into the Scaling Unit, but was more convenient in use than the latter, since it was resettable; it thus registered every 100th pulse from the counter, in the work described. The relay circuit in the Timing Unit could be set to multiply any of the four timing intervals, viz. $\frac{1}{2}$ sec, 1 sec, $\frac{1}{2}$ min. and 1 min. by any of the multipliers 1, 4, 10, 40, or 100, thus giving a choice of count times of:

 $\frac{1}{2}$, 1, 2, 4, 5, 10, 20, 40, 50 and 100 seconds and $\frac{1}{2}$, 1, 2, 4, 5, 10, 20, 40, 50 and 100 minutes. At the end of the preset time, the unit switched off the count on



*

Hand T:	ming	Automatic Timing			
N	Δ	N	Δ		
28841	-105	28796	-351		
28986	+ 40	28746	-301		
29408	+462	29015	-132		
29460	+514	30022	+876		
28942	- 4	30029	+883		
28778	-168	29074	- 73		
28922	- 24	29079	- 68		
28710	-236	28966	-181		
28851	- 95	28838	-309		
28760	-186	28925	-222		
28912	- 34	291 39	- 8		
28779	-167	291 38	- 9		
= 28946	$\sqrt{(\bar{N})} = 170$	N = 29147	√(N) = 17 ⁴		

TABLE 2

the Scaling Unit, the total count for that period being read on the mechanical register of the Timing Unit, and the interpolation neon lights on the Scaling Unit. Resetting to zero, in preparation for the next count, was done entirely by switches on the Timing Unit.

Two sets of counts were taken with a Thallium source, one with hand timing as described earlier, and the other timed by the Timing Unit. Each count with both methods was of 2 minutes duration. The accuracy of the counting time was thus better than 1 part in 6,000 (0.0167%) for the automatically timed case. The two sets of readings are given in Table 2; the standard deviation was still considerably greater than the square root of the mean, this being more noticeable in the automatically timed case, proving that timing errors were not the cause of the inconsistencies. The Timing Unit was, however, used for all subsequent work.

The counter characteristic was determined and showed a rise at the start of the plateau, indicating the production of multiple counts at low overvoltages; this characteristic (b) is compared with the original one (a) in Fig. 5. The working voltage of 1,480 volts was however on the straight part of the curve and there seemed no reason to believe that the rise at the beginning was causing trouble. The effect of paralysis applied in the input circuit of the scaler, was investigated. Graphs supplied with the scaler showed that the paralysis time of the input flip-flop circuit was appreciably less when a large pulse was applied to the input and the probe amplification was accordingly adjusted to give an input pulse of 20 volts amplitude, as observed on a

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N	Δ	N	Δ	N	Δ
27470	-704	28232	+ 58	28448	+274
27414	-760	28499	+325	28619	+445
27860	-314	28182	+ 8	28469	+295
28253	+ 79	28291	+117	28350	+176
N = /(N) = Omittir are obt	28174 168 mg the firs ained: 28371	t three re	Jults, the	$E\Delta^{2} = 1,6$ $\frac{(E\Delta^{2})}{11} = 0$ Following $E\Delta^{2} = 1$	91,897 1301 values 69,228

TABLE 4

N	Δ	N	Δ	N	Δ.
26583	-1364	27952	+ 5	28459	+512
27153	-794	28203	+256	28385	+438
27248	-699	28063	+116	28286	+339
27637	-310	28054	+107	28621	+674
27634	-313	28044	+ 97	28360	+413
27966	+ 19	27972	+ 25	28432	+485

TABLE 5

T = 50 secs.		T = 1	min.	T = 1 min.	
N	Δ	N	Δ	N	4
2494	-1	2994	-4	3007	+5
2495	0	2995	-3	3003	+1
2496	+1	2996	-2	3003	+1
2496	+1	2996	-2	3002	0
2496	+1	2995	-3	3003	+1
2495	0	2998	0	3002	0
2495	0	3001	+3	2999	-3
2495	0	3001	+3	3003	+1
2495	0	3003	+5	2999	-3
2495	0	3005	+7	2998	-4

The half-hourly frequency readings at the power station were: 09.30, 50.0 cycles/sec; 10.00, 49.9 cycles/sec; 10.30, 50.0 cycles/sec.

The first set of the above readings was taken between 09.30 and 10.00 and the other two between 10.00 and 10.30.

The frequencies calculated from the readings agree perfectly with these values.

Clearly, the rise in frequency between 10.00 and 10.30, took place during the course of the second set of readings, and neglecting this set, the maximum deviation of any reading represents only 1 part in 600 calibrated cathode ray oscillograph. The preset potentiometer controlling the paralysis time was set at about the middle of its range, giving a paralysis time of a few hundredmicroseconds (according to the graphs supplied), and a characteristic was taken (Fig. 5(c)). This showed a flat plateau, without the initial rise. A consistency check (Table 3) still showed a very marked disagreement between the standard deviation and the square root of the mean. This particular set of readings included three much lower than the rest; if these were omitted, the standard deviation agreed well with the square root of the mean. A repeat of this experiment (Table 4) showed one very large individual deviation, and the standard deviation was again much too large.

There is an interesting feature about these sets of results; in many cases the largest deviations from the mean occur in the first three or four readings, a few seconds after the counter starts to record a high counting rate, though it is difficult to see why this should be so.

In order to verify that the scaler and timing unit were behaving correctly, the A.C. mains supply was counted. The 50 cycles calibration of a Cossor double-beam oscillograph was amplified in the oscillograph amplifiers to an extent sufficient to distort the sine wave into an approximately square wave and this was fed directly into the scaler; it was found that by suitable adjustment of the gain of the amplifiers, the scaler counted this without differentiation. Sets of readings were taken for 50 seconds and 1 minute, thus checking both the $\frac{1}{2}$ sec. and $\frac{1}{2}$ min. clock pulses. These are given in Table 5. The frequency readings were obtained from the power station, for the times

170.4		1999 N. 7	Δ.,	N	277
101×1		18	1		
8631	- 83	8585	-129	8670	- 44
8707		8847	+133	8778	+ 64
8764	+ 50	8676	- 38	8872	+158
8754	+ 40	8701	- 13	8855	+ 141
8669	- 45	8698	- 16	8474	-240
8421	-293	8779	+ 65	8749	+ 35
8551	-163	8586	-128	8689	- 25
8650	- 64	8695	- 19	8834	+120
8741	+ 27	8794	+ 80	8896	+182
8634	- 80	8871	+157	8858	+144
				l <u></u>	سایت در این ا
i (a)	9 741 - 1			50 ² - 1	

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TABLE 6

Ti 2 1 1

 $\{i_1,j_2,\ldots,j_{2^{d-1}}\}$

4.175

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during which the counts were taken, and very good agreement was obtained, showing that the Scaling and Timing Units were behaving correctly.

The counter high-voltage supply was very stable and showed no evidence of spurious pulses; it was therefore concluded that the Geiger-Muller counter was the source of the inconsistencies. A cylindrical, copper wall Y-ray counter was investigated, and this also gave inconsistent results, although the standard deviation did not differ from the square root of the mean to such an extent as with the G.E.C. counter. The set of results is given in Table 6. The glass insulator on the top of the G.E.C. counter was cleaned with alcohol and all high voltage connections were checked, but results were still far from consistent.

The solution adopted was to randomize the inconsistencies by building up each point on the absorption curves, from a number of short readings with each absorber, i.e. the counting rates were observed for each absorber thickness in turn, starting from zero and working up to the maximum thickness; the readings were then repeated in the reverse order and the process continued until the desired accuracy had been obtained.

Before the absorption curve for Thallium was investigated in the manner already described, the resolving time of the counting system with the paralysis applied in the scaler, was found. In order to make the critical term $(C_1 + C_2 - C_{12})$ in the determination, as large as possible, a very high counting rate was employed. The mechanical register in the scaler was cut out and were the pulses which normally operated it/fed to the output plug. This output, consisting of every 100th pulse, was fed into the

mg/om ² Al.	No.	tion	No.	Dura- tion	No.	Dura- tion	No.	Duration
1		80C.		80C.	Sec. 4	sec.		880
123-2	5197	20	5139	20	5248	20	5895	20
161.7	2405	30	2507	30	2528	30	2895	30
168-4	2644	40	2843	40	2794	40	3066	40
175-1	2712	50	2797	50	2733	50	2777	50
181-8	2551	60	2599	60	2606	60	2628	60
188-5	3341	100	3522	100	3597	100	3488	100
195-2	2715	100	2764	100	2905	100	2732	100
200+2	2650	120	2794	120	2862	120	2739	120
206.9	4166	240	2256	120	2237	120	4376	240
213-6	3269	240	3317	240	3504	240	3460	240
220-3	2514	240	2606	240	2587	240	2570	240
227.0	2611	300	2617	300	2620	300	2601	300
233.7	4012	600	4058	600	2226	300	4124	600
247-9	2220	600	4441	1200	3060	600	2174	600
286-4	3557	2400	3584	2400	3693	2400	1903	1200
324+9	2827	2400	3007	2400	2977	2400	1470	1200
363-4	2657	2400	2832	2400	2825	2400	2820	2400
401-9	2542	2400	2647	2400	2589	2400	2710	2400
440-4	2546	2400	2558	2400	2483	2400	2517	2400
487.5	2509	2400	2468	2400	24.38	2400	2432	2400
726-8	2221	2400	2754	3000	2873	3000	2313	2400
967-0	2366	2400	2665	3000	2662	3000	2732	3000
1207.3	2059	2400	2538	3000	2631	3000	2701	3000

counting system described in Appendix III. The maximum average counting rate in this determination was about $1800/\sec$. and was followed quite easily by the recording apparatus. The value obtained for the resolving time was 158.3 ± 3.1 microseconds, using an input pulse amplitude of 20 volts.

The first determination of the Thallium absorption curve was made with two sources having a ratio of strengths of approximately 13:1. The weaker source was used for the upper part of the curve and in order to reduce the counting time while maintaining the same accuracy, the stronger source was used to follow the curve to the higher absorber thicknesses. The counting rate for each absorber thickness was built up from several readings, as described earlier, but the consistency of some of these "partial" readings was very poor. A typical set for the stronger source is given in Table 7. The readings for 123.2 mg/cm², 161.7 mg/cm², 168.4 mg/cm² and 247.9 mg/cm² are particularly bad, and the validity of the absorption curve thus obtained was open to doubt.

At this stage in the work, a Probe Unit Type 1014A became available. This unit was designed by A.E.R.E. to overcome difficulties such as have been described above.

The spurious counts in a Geiger-Müller counter are caused by the release of secondary electrons from the cathode surface by positive ions which have drifted there after a discharge has occurred. These spurious counts are delayed from the true counts by the time taken for the positive ions to drift from the region of the centre wire to the cathode (i.e. a few hundred microseconds). If, however, the H.T. voltage applied to the counter is reduced below the threshold value for this period, any secondary electrons

TABLE 8

N	Δ	N	Δ	N	Δ.
11077	+22	11030	- 25	11080	+ 25
11109	+54	11050	- 5	11025	- 30
11013	-42	11008	- 47	10835	-220
11080	+25	11050	- 5	11093	+ 38
10957	-98	11255	+200	11161	+106

TABLE 9

N	Δ	N	Δ	N	Δ
13167	+ 68	11417	-1682	14331	+1232
15946	+2847	11412	-1687	13998	+ 899
12419	- 680	121 35	- 964	15885	+2786
11278	-1821	14001	+ 902	13160	+ 61
11588	-1511	13318	+ 219	12431	-668

produced at the cathode can produce no pulses. The Probe Unit Type 1014A reduced the counter voltage by about 240 volts for a preset time (which could be 200, 300, 400 or $500 \mu \sec$.) following each counter pulse; thus by choice of the suitable quenching time for the counter, spurious pulses were eliminated. In addition, the resolving time of the counter was replaced by the accurately known quenching time of the Probe Unit, reducing a further possible source of error. As in the Probe Unit Type 200, the high-voltage components associated with the counter were included in the unit, which derived its power supplies from the Scaling Unit and returned an output pulse to the latter unit; as before, the high voltage supply from the Power Unit was fed to the counter through the probe.

A consistency check using this Probe Unit instead of the Type 200, all other conditions being the same, showed a striking improvement as indicated in Table 8. The quenching time for this set, was 300μ sec. A trial with a quenching time of 200μ sec. (Table 9) showed that in this case, spurious pulses were present, so the quenching time of 300μ sec. was used for subsequent work. As might be expected, this Probe Unit also improved the plateau considerably (Fig. 5(d)). It was found necessary to reduce the sensitivity of the unit below its maximum (0.2 volts) since if a high counting rate was used, the probe tended to oscillate and could only be stopped by switching off its voltage supplies for at least 3 seconds.

Shortly after the solution of the consistency problem described above, a paper, describing similar work leading up to the production of the Probe Unit Type 1014A, was published by Putman (10).



He points out that the spurious counts increase with the overvoltage applied to the counter, most of them occurring at a time after the true pulse, roughly equal to the dead-time of the counter, and his graphs show that, for a counter of the G.E.C. pattern, there are no spurious counts later than about $300 \,\mu$ sec. after the true count. He also shows the improvement of the counter plateau, when used in conjunction with a quenching circuit similar to that used in the Probe Unit Type 1014A.

Putman treats the subject in detail, giving distribution curves of intervals between counter pulses obtained with a pulse interval analyser, apparatus which was not available to the writer. The results described above, however, are entirely in agreement with his work.

Trial Absorption Measurements. The work on the final graphs, from which the analysis was made, is given in detail later, but it is convenient at this stage to review the preliminary work done with Thallium and Radium E.

The Thallium absorption curve, investigated before the Probe Unit 1014A was used, was determined with two sources and it was arranged that the two sets of results overlapped slightly, to assist the fitting process. The readings for both sources, after all corrections had been made, were expressed in terms of the intensity for an absorber thickness in the middle of the overlapping portion, the intensity for this thickness being taken as unity. It was found, however, that, even allowing for the statistical error in each intensity reading (1%), there was a definite change of slope, illustrated in Fig. 6. The sources used were in the form of Thallium Oxide, a fairly fine brown

powder and were comented into the recess in the perspex holder with a solution of distreme in benzeme. The stronger of the two sources was quite epaque and was probably several mg/cm² in thickness. At the time, the change of slope was attributed to self absorption in this source, since it seemed unlikely that the inconsistencies could account for such a definite effect; in addition, a later determination, after the Probe Unit Type 1014A had been brought into use, showed the same phenomenon, ruling out any possibility of spurious counts being the cause.

The first trial with Radium E was made with a source deposited on nickel. A disc of nickel, $\frac{1}{2}$ " in diameter and about $\frac{1}{51}$ " thick was coated on one side with Picein wax and immersed in a solution of 5 µCof Ra. D. for 4 hours. The Ra. E. was thus deposited on the upper side only, since the wax on the other prevented collection. After removal, the disc was washed and inserted into the recess in a source holder, with the bare metal upwards, and was stuck in by a drop of chloroform which dissolved wax and perspex. comenting them securely when it evaporated. The decay of this source was followed for 22 days, the activity being found to have the correct half-value period of 5.0 days, within the limits of error. The absorption curve for this source was determined, but in order to maintain the back-scattering conditions prevailing with the Thallium sources, was discarded in favour of one taken with a source propared as follows: A nickel foil was inserted in the Ra. D. solution for 48 hours and the Ra. E., together with some of the nickel was dissolved off in nitric acid. The solution was evaporated to dryness, dissolved in water and transferred to a source holder. This solution was then evaporated under an infra-
TABLE 10

N	Δ	N	<u> </u>	N	Δ
13129	+422	12794	+ 87	12546	-161
13136	+429	12933	+226	12758	+ 51
12888	+181	12781	+ 74	12648	- 59
12850	+143	12359	-348	12492	-215
12545	-162	12513	-194	12625	- 82
12539	-168	12604	-103	12588	-119

TABLE 11

and the second second

N	4	N	Δ	N	Δ
11041	+169	10896	+ 24	10897	+ 25
11067	+195	10972	+100	10753	-119
10863	- 9	10848	- 24	10777	- 95
10834	- 38	10715	-157	10805	- 67

red lamp to avoid undue heating of the perspex. The active material was then cemented in with chloroform, relying on the dissolved top layer of perspex to act as adhesive. This source was rather large in volume, but was porcus, and its mass/sq. cm. was fairly small. The decay was followed over 25 days and found to be correct.

At this stage a new preparation of Thallium oxide of much higher specific activity than the first, was obtained from Chalk River, Canada. It was hoped that this would eliminate difficulties due to self-absorption in the stronger source. An absorption curve was taken using sources made from this material.

A consistency check made at this time, revealed that spurious counts were once more occurring (Table 10). The probe quenching time was increased to 400 µ sec. but this did not entirely remove the inconsistencies (Table 11). A measurement of the counter characteristic revealed that the plateau had shifted to a higher voltage and the slope had increased considerably: in addition, the background was rising steadily. This information indicated that the Geiger-Müller counter had reached the end of its working "life" and raised doubts as to the validity of previous results. At this time also, difficulties were encountered with background increases due to other sources in use in the room. (It was verified that this was not the cause of the background rise attributed to the end of the counter life, this being a separate effect). Accordingly, the apparatus was moved to another room, well away from all other sources, and a new counter was used for subsequent work, the Radium E and Thallium measurements being repeated.

4. THE FINAL ABSORPTION MEASUREMENTS

<u>Corrections</u>. In any accurate work with radioactive materials and Geiger-Müller counters a number of corrections must be applied. A further source of error is the statistical deviation of a random distribution, and it is convenient to include this under the same heading. The following is a brief description of the methods which must be applied.

(a) <u>Statistical Deviation</u>. The accuracy of the measurement of a random distribution, such as is obtained from a counter and a radioactive source, is determined by the number of events observed. The standard error of a total count N is given by / N, so to obtain an error of 1% in the measurement 10⁴ events must be recorded, and all determinations must therefore be of sufficient duration to give this number.

(b) Resolving Time. The recording apparatus has a finite

"resolving time" after a pulse has been recorded, during which it is "dead", and will not register another pulse. Let the observed counting rate be N counts/sec, the true counting rate be N' counts/sec, and the resolving time of the counting system be σ secs.

In 1 second, on the average, the counting system is "dead" for a time Nor secs, and the effective counting time is thus $(1 - N\sigma)$ secs; hence the true counting rate is given by: N' = $\frac{N}{1 - N\sigma}$. This is not a convenient form for calculation. Taking reciprocals we have: $\frac{1}{N'} = \frac{1}{N} - \sigma$. This expression was used throughout for correction of counting rates for resolving time, values of reciprocals being found from Barlow's Tables. The value of σ , in

the work under consideration was simply the quenching time of the Probe Unit.

(c) <u>Background</u>. Even in the absence of a source, a Geiger-Müller counter always gives a small counting rate, known as the "background". This is due to cosmic radiation and very small quantities of radioactive materials in the walls of the surrounding building. To give the effect due to the source alone, the background must in all cases be subtracted from the observed counting rate corrected for resolving time. During the course of the work described, several measurements of the background were made, and the average value calculated from the sum of the counts recorded divided by the sum of the times for which it was measured.

(d) <u>Decay of the Activity of the Source Material</u>. The activity of a radioactive material is constantly decreasing according to the relation $N_t = N_o e^{\lambda t}$ where N_o is the counting rate at zero time, N_t is the counting rate at time t, and λ is the disintegration constant of the material. It is necessary therefore, in general, to correct all readings for this decay.

If the source has a short half-value period $T \left(=\frac{\log_e 2}{\lambda}\right)$, it may decay appreciably during the measurement of a counting rate, which must extend over a finite time determined by the statistical accuracy required. However, there is a time after the start of the reading, forwhich the average value obtained for the counting rate during the period of measurement, is the true instantaneous counting rate. It is a good approximation to take the mid-point, in time, of the reading and the following shows how small an error is introduced even for counting times which



are an appreciable fraction of the half-value period.

Fig. 7 represents the decay curve for a pure radioactive material; the counting rate N is plotted, on a linear scale, against time. The curve follows the law $N_t = N_0 e^{-\lambda t}$, stated above. Suppose a reading is taken for a time $\lambda \tau$, the midpoint, in time, being at time t. The observed counting rate is decreasing during the reading, following the curve; for correction purposes relative to other readings the average counting rate is to be attributed to the time t and it is required to find the error thus incurred.

The average counting rate over the period 27 is given by:

$$\vec{N} = \frac{1}{2\tau} \int_{t-\tau}^{t+\tau} N_o e^{-\lambda t} dt$$
$$= \frac{N_o e^{-\lambda t}}{2\lambda\tau} \cdot \left(e^{\lambda \tau} - e^{-\lambda \tau} \right)$$
$$N_o e^{-\lambda t} = N_t \qquad \text{so } \vec{N} = \frac{N_t}{2\lambda\tau} \left(e^{\lambda \tau} - e^{-\lambda\tau} \right)$$

The instantaneous counting rate at time t, is simply No

Se

Now

$$\frac{\overline{N}}{N_{t}} = \frac{1}{2\lambda\tau} \left(e^{\lambda\tau} - e^{-\lambda\tau} \right)$$
$$= 1 + \frac{1}{3!} \cdot \lambda^{2}\tau^{2} + \frac{1}{5!} \cdot \lambda^{4}\tau^{4} + \dots$$

Suppose the duration of a reading is $\frac{1}{5}$ T where T is the halfvalue period of the source material: then $2\tau = \frac{T}{5}$ and $\lambda \tau = (\frac{T}{T}) \log_{e} 2 = \frac{1}{10} \log_{e} 2$. Since $\lambda \tau$ is small, compared with unity, $(\lambda \tau)^{4}$ and greater powers can be neglected.

So $\frac{N}{N_c} = 1 + \frac{1}{6} (\lambda \tau)^2 = 1.0008$ and the error involved is thus 0.08%. TABLE 12

	Que	nohing Time	$= 400 \mu$	secs.	40 8.
N	<u>∧</u> N		Δ	N	Δ
4521	-26	4534	-13	4572	+25
4622	+75	4618	+71	4619	+72
4598	+51	4456	-91	4549	+ 2
4520	-27	4524	-23	4431	-116
Ň =	4547			ΣΔ ²	42920
/(N) =	67			$\sqrt{\left(\Sigma \Delta^2\right)} =$	62

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The application of these corrections is described in the sections devoted to the three source materials since it differs slightly for each element.

Apparatus. The electronic apparatus used for all the following experiments consisted of a Power Unit Type 200, a Scaling Unit Type 200A, a Timing Unit Type 1003K, a Probe Unit Type 1014A, and a G.E.C. Geiger-Müller Counter Type G.M.4, mounted in the second perspex assembly described earlier (Fig. 4). A lead turret having a $1\frac{1}{2}$ wall, was placed round the counter and a brass box, bolted to the top of the turret, provided electrical screening for the counter wire; the braiding of the screened lead from the Probe Unit was connected to this box by means of a special brass fitting providing complete screening at the join and the cathode of the counter was earthed to the box. The first three units were mounted one above the other in a Post Office type rack for convenience in working. The Probe quenching time was set at 400 μ sec.

Consistency checks were made during the following work, the results of a typical one being given in Table 12. <u>The Absorption Curve of Thallium</u>. The initial work on Thallium, described earlier, was done with sources prepared from material activated by neutron bombardment in the pile at Harwell, but for the final work the higher specific activity material, activated in the Chalk River pile, was used. A small amount of the Thallium Oxide powder was spread uniformly over the bottom of the recess in a perspex source holder and a drop of chloroform was added to dissolve the top layer of perspex and cement in the active material. Two sources were used of approximate strengths

1.6 microcuries and 0.016 microcuries. The thickness of the stronger source was estimated at less than 1 mg/cm^2 , and that of the weaker at much less than 0.1 mg/cm². The latter consisted of an almost invisible amount of Thallium Oxide and even the former was transparent (Thallium Oxide itself, is an opaque brown powder). There was thus practically no self-absorption in the sources.

The absorption curve was determined backwards from the highest absorber thickness used. Error limits were not plotted on the curve since they were too small to be shown conveniently. The counting rate for each absorber thickness was found to approximately 1% in every case. The maximum standard error in the observed counting rate was for an absorber thickness of 967 mg/cm² and was 1.06%. The background count was measured to 1.19% and after subtraction of background, the standard error of this reading was 1.24%. The standard error was not less than 0.80% in any reading.

The stronger source was used for absorber thicknesses from the maximum, back to $161 \cdot 7 \text{ mg/cm}^2$; the weaker source was used from zero to $220 \cdot 3 \text{ mg/cm}^2$. Each reading of counting rate was corrected for probe quenching time, background and decay; the time of the first reading with each source was taken as the zero of time for all readings made with that source and each reading was corrected back to the zero, to the nearest day, the times being taken to the mid-point (in time) of the reading. Using the formula given earlier, the maximum length of any reading being 3000 secs. (i.e. $\tau = 1500$ secs.) and the half-value period being 3.5 years, it is clear that the error involved in attributing

the counting rate to the mid-point of the reading is completely negligible in this case.

The two sections of the curve were fitted together by taking the counting rate for an absorber thickness in the region where they overlapped, and assigning the value of unity to its counting rate. all other readings for both sources then being expressed as multiples or fractions of this reading. The complete absorption curve is shown in Fig. 8(a). It was found that there was a slight change of slope at the join of the two sections, as there had been with the less reliable results taken with the Harwell active material (described earlier), when the change of slope had been attributed to self absorption in the much thicker sources (the strong source in that instance was completely opaque and the weaker source was considerably thicker than the stronger source prepared from the Chalk River material). This effect can not, therefore, be attributed to self-absorption in the sources. The 400 µ sec. quenching time of the probe was accurate to better than 5%, but it is possible that, owing to the large corrections which had to be applied to the readings taken with the stronger source, in this region, a slight error in the quenching time is sufficient to cause this change in slope. The effect is undoubtedly due to some factor introduced in the electronic apparatus.

The change of slope was barely perceptible on the main absorption curve and was therefore neglected. <u>The Absorption Curve of Radium E</u>. The Radium E sources used previously had not been satisfactory, the first since it was backed with nickel and was therefore likely to give an increased

amount of back-scattering, and the second because of its bulk and the relatively large amount of nickel mixed with the Radium E; the latter was estimated to have a thickness of several mg/cm². A different technique was therefore adopted; the same solution of 5 microcuries of Radium D was used as a starting point, but before preparing a Radium E source, a small piece of silver foil was immersed in the solution for 48 hours. This collected most of the Polonium from the solution, without removing the Radium E: this foil was discarded. A small piece of copper foil was then left in the Radium D solution for 24 hours, collecting Radium E: copper also collects Polonium, but most of this had been removed by the silver foil. The copper foil was washed and dissolved in nitric acid, keeping the volume of solution fairly small and the acid concentration as low as possible. A small amount of Neodymium Oxide was also dissolved in this solution which was then neutralized with a few drops of concentrated Ammonium Hydroxide. This precipitated the Radium E and the Neodymium carrier, as the hydroxides, and left the copper in solution as a suprannonium compound. The gelatinous precipitate was centrifuged down and the clear solution decanted off. The precipitate was redissolved in nitric acid, and reprecipitated with anmonia. the clear solution being decanted off as before. This process was repeated three times to ensure freedom from copper. The precipitate was then washed by stirring it with water, centrifuging and decenting, repeating the process three times. The clean precipitate was then dissolved in a few drops of dilute hydrochloric acid, and a portion of the solution transferred to a source holder, which was then placed on a white surface under

an infra-red lamp. This evaporated the solution to dryness without undue heating of the transparent perspex source holder. The final operation was to add a few drops of chloroform to cement in the source.

The whole of the Radium E absorption curve was determined with a single source, prepared in the manner described above. The readings for small absorber thicknesses were made 16 days after the first reading at large absorber thickness was taken. The strength of the source immediately after its preparation was about 0.5 microcuries and the thickness was estimated at less than 1 mg/cm².

The half-value period of the source was checked over 15 days and found to be the expected 5 days, within the limits of experimental error.

All readings were corrected for probe quenching time, background, and source decay, the correction time being taken as the mid-point of the reading, relative to the mid-point of the first reading. Using the formula derived earlier, for the maximum time of any count (3000 secs.), the error involved in assigning the count to the mid-point of the reading was approximately 1 part in 10^6 , a negligible quantity compared with the statistical errors involved.

The greatest statistical error was 2.7% and occurred in the observed counting rate for 967 mg/cm² of absorber. The background was known to 1.45% and the standard error for the counting rate corresponding to this point on the curve was 6.3%. The points on the straight portion, beyond the β -ray and-point all have an error very slightly less than this, and all points

TABLE 13

Nt B t (N-B)	= oos = cou = bac = tim = ful B = Pro	erved coun nting rate kground co e, in days ly correct time (1 0.214 ± 0. be quenchi	correct unting , after ; = 0) of 003 could ing time	te per ted for rate first ting ra the fi ints/sec = 400	reading te, rel rst rea	quenching ative to ding	; time the
mg/cm ² of AL	Number	Duration	Nt	N't	(N'-B)	t(days)	(N'-B)
200-2	16736	240 secs.	69•73	71.73	71-52	9.013	249
206-9	15227	240 "	63-45	65-10	64-89	9-010	226
213.6	13528	240 "	56+ 37	57.67	57-46	9.006	200
220.3	12048	240 *	50-20	51-23	51.02	9.003	178
227.0	10586	240 "	44= 11	440 90	44= 69	8-999	156
233-7	11916	300 "	39.72	40-36	40-15	8-996	140
247.9	16697	600 *	27-83	28-14	27.93	8.985	97-1
254.6	14909	600 "	24.85	25-10	24-89	8-978	86-4
261 . 3	13678	600 *	22.80	23.01	22.80	8.970	79.1
268.0	11586	600 *	19.31	19.46	19-25	8-909	66-2

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 $\Phi_{l_{1}}(\mathbf{x}) =$

for lower absorber thicknesses than 331.6 mg/cm² have a standard error of about 1%. No point has a lower standard error than 0.86%. Error limits are plotted only for some points in the straight lower portion of the curve. A section of the results is shown in Table 13.

Readings taken for the maximum absorber thickness, 15 days after the preparation of the source, showed no definite increase in counting rate proving that the Y-rays from the daughter Polonium were not responsible for this part of the curve and its correction for a 5 day half-value period was therefore justified.

The curve was plotted from the end corresponding to high absorber thickness, so that the increase in counting rate due to removal of absorbers, minimized the reduction of source intensity due to decay.

The determination of the curve from the maximum absorber thickness to 84.7 mg/cm², extended over 9 days, after which the source was allowed to decay for a week and the curve was then completed. After the complete set of readings had been corrected for decay, it was found that there was a slight discontinuity at the overlapping portion, but both sections were parallel. The upper section was therefore moved downwards to fit the lower part. This discontinuity was attributed to a change in counter sensitivity.

The final curve is shown in Fig. 8(b). The actual recorded counting rate/sec. is plotted, after correction for decay. <u>The Absorption Curve of Actinium C"</u>. Owing to its short halfvalue period of 4.76 minutes, Actinium C" presented much greater problems than Thallium and Radium E. Clearly no chemical

separation is workable, since the activity decays so rapidly. The primary source available was in the form of Actinium Fluoride with Lanthamam Fluoride carrier and was rather weak. Purification was carried out at the Cavendish Laboratory, Cambridge, and at Edinburgh University. Actinium decays to Radio-Actinium which in turn decays to form Actinium X and it is possible to collect Actinium B and its products from the Actinon emitted by the Actinium X, by means of the recoil collection method; Actinium C" can then be collected from this source, again by recoil. The method used was as follows:

The powdered Actinium Fluoride (with Lanthamum carrier) was spread uniformly in a platimum dish of about 1" diameter, which was placed in the bottom of a brass pot about 1" deep and 1" in diameter, into the top of which screwed an ebonite plug. The surface on which the Actinium B was collected was a brass disc, $\frac{1}{2}$ " in diameter and $\frac{1}{16}$ " thick, at the end of a 2 B.A. screwed rod. This "button" was held in a 2 B.A. clearing hole drilled through the ebonite plug, by means of a mut on the upper side. The button was thus held tightly against the underside of the ebonite plug, leaving only the front surface exposed to the Actinon, and was maintained at a negative potential relative to the pot.

Actinon decays rapidly (T = 3.92 sec.) to Actinium A which has a shorter half-value period (2 x 10^{-3} sec.), giving Actinium B; the disintegration scheme is then as follows:





A button activated in the pot thus has the active components AcB, AcC, AcC' and AcC". The branching is greatly in favour of AcC", 99.68% of the AcC undergoing α -emission resulting in AcC", so for the purposes of the experiment under consideration, the other branch can be neglected.

Actinium C" can be collected in a pure form by recoil from the Actinium B source by placing another button close to it and maintaining it at a negative potential relative to the first.

The first procedure with the Actinium source was to check the half-value period of the Actinium B. A copper foil was wrapped round a button and activated in the pot for 24 hours with an applied potential of 120 volts. This foil was then tested for half-value period. When plotted with the counting rate on a logarithmic scale, all the points were found to lie very close to a straight line which gave a value of 36.7 minutes for the half-value period. This is very near the accepted value of 36.1 minutes for Actinium B. The results are shown in Fig. 9.

In order that a valid comparison could be made with the other two elements, it was necessary to provide the same backscattering conditions for the Actinium C" as for the Thallium and Radium E. This precluded the use of a metal button for the collection of the Actinium C" from the Actinium B button. It was decided that the best method was to use perspex, rendered conducting by a thin coating of colloidal graphite. One of the perspex source holders was modified to receive a button $\frac{1}{2}$ " in diameter and $\frac{1}{16}$ thick, with a projecting pin on the underside $\frac{1}{16}$ " long and $\frac{1}{12}$ " in diameter. The pin was a push fit in a









holder (Fig. 10) which was similar to the buttons used in the activation pot. The threaded portion of this holder was hollow to allow an ejector to be pushed through. When the button was in position in the holder, only its face was exposed. To verify that the graphited perspex would collect active material satisfactorily, the holder was inserted in the pot and the perspex button activated with Actinium B and its disintegration products. A reasonably satisfactory counting rate (about 200/sec.) was obtained with the standard counter assembly and the half-value period found to be about 36.1 mins. On removing the source, the counter background was found to have increased by a factor of about ten, but was decreasing rapidly. The perspex button was replaced in the assembly for 2 minutes and the background was checked immediately, 1 minute readings being taken every 2 minutes. After subtracting the previously known background, these results were plotted with the counting rate on a logarithmic scale and the background was found to decay with a half-value period of approximately 4.69 min. (Fig. 11(a)), sufficiently close to the value of 4.76 min. for Actinium C" to identify the contamination as being due to that material. The conclusion reached, was that the counter window was collecting the Actinium C" by recoil from the source and a further check was made with another source, covered with paper; in this case, no appreciable contamination was observed.

The perspex button in the above case was activated by mistake, with a positive collection potential and a negatively activated button was then tested in the same way. No contamination with a bare source, was observed in this case, which showed that the cause of the contamination may have been due to a retention

of positive charge by the persper, a remote possibility since handling was done throughout with rubber gloves, and the source was never definitely earthed. In the counting assembly the source was insulated by the perspex, and the counter was earthed. allowing the possibility of a potential difference between source and counter window, with the counter negative, a circumstance which would assist the collection of the AcC" on the counter. This was disproved, however, by a further trial with a button activated with a positive collection potential, when no contamination was obtained. It was therefore concluded that the first contamination was due to the recoil process removing from the perspex button some of the finely divided graphite which later "settled down", although in this case a contamination decay time nearer 36-1 min. (AcB) would have been expected. No further contamination was obtained and it was not considered worth while to pursue the subject further.

The collection of Actinium B on graphited perspex being satisfactory, an attempt was made to collect Actinium G" from a brass button activated with Actinium B. A perspex button, in its holder, was placed for 10 minutes facing an Actinium B brass button, separated from it by about 1 mm. and 120 volts negative to it; the former was then removed and counted, the time between the end of activation and the start of counting being about 1 minute. The initial counting rate of 1.6/sec. was disappointingly low, considering that the initial counting rate of the contamination was nearly double this figure; a check of halfvalue period, however, indicated that the source was definitely Actinium C". At this stage, the single 120 volt H.T. battery

used for activation was replaced by 230 wolt D.C. mains. An Actinium C" source activated with this voltage gave a greatly increased counting rate of 14/sec. which indicated that a higher collecting voltage was advantageous. A further trial was therefore made with a collecting potential of 2000 wolts across a gap of $1\frac{1}{2}$ mm, and an initial counting rate of about 290/sec. was obtained; the half-value period was checked and was found to be 4-45 minutes (Fig. 11(b)).

Results obtained from the recoil activation, both from Actinon and from the Actinium B button are interesting in many respects. The theory of the process is as follows; after emission of an α -particle, the residual atom recoils and ionises the gas in its path, its range being of the order of 0.14 mm. in air. The residual atom is initially negatively charged, having emitted a positive α -particle, but when the atom ionises the surrounding air, it loses some electrons and becomes positively charged. This process occurs for the great majority of the recoil atoms; for Radium B, for example, about 0.001% of the recoil atoms are negatively charged and nearly all the remainder carry a single positive charge. The small range of the recoil atoms can be augmented by applying an electric field, and collecting the recoil atoms on the negative electrode.

On this theory, a saturation field of the order of 100 volts/mm. would be expected and no appreciable collection should be obtained if the collecting electrode is positive. The collection of Actinium B from Actinon showed however that positive and negative electrodes collected approximately equal activities at 120 or 230 volts, the higher voltage giving an increased yield in both









cases. If a collection potential of 2000 volts was used, there was practically no resulting activity on a positive collector and a negative electrode gave a source which was not noticeably stronger than that obtained with 230 volts. The effect of increased voltage on the negative collection of Actinium C" from a button of Actinium B was however very appreciable. Starting with Actinium B sources of approximately equal strength, and employing the same activation times throughout, there was an increase in the strength of the resulting Actinium C" source by a factor of about 14 when the collection voltage was raised from 120 to 230, and by a factor of 20 when it was increased from 230 to 2000 volts. This information should be regarded as qualitative rather than quantitative, but gives an indication of the effect to be expected. The phenomenon of equal collection on positive and negative electrodes has been observed by other workers in the same laboratory, in connection with the preparation of Thorium active deposit, and a detailed investigation is to be made very shortly.

The problem of taking measurements with fairly weak sources of active materials obtained by recoil, has been attempted by several workers. One possibility is to place the Actinium B button in a tunnel of wall thickness sufficient to cut off all the β -rays, facing a collecting electrode situated at an angle, below the counter, so that the latter "sees" only the collector; Fig. 13 shows the scheme diagrammatically. This method has the disadvantage that the collector must be placed some distance from the primary button, so that the counter cannot "see" the latter, and there is also the possibility of scattering of Actinium C"

 β -particles from the end of the tunnel. The use of an "inverse" button is an improvement (Fig. 14). Again the Actinium B button is placed in a shielding tunnel, but the recoil atoms of Actinium C" are deflected on to the underside of a negatively charged button of known small thickness, by means of a positive focussing electrode. This method allows solid angle conditions to be maintained but completely alters back-scattering conditions and does not allow the curve to be continued to zero absorber thickness, although the last few points can be obtained with separate Acc" sources obtained externally. Both these methods allow a longer series of readings to be taken, since after equilibrium is reached the half-value period of the Actinium C" source is effectively the same as that of the Actinium B source (viz. 36.1 min.). A further method, used by Sargent (11) with Thorium C", for measuring the energy distribution of β -rays from atoms of different ages, was applicable to the problem under consideration. This consisted, in effect, of a moving belt (a copper strip in Sargent's experiment), driven at constant speed by an electric motor; the counter and Thorium B button were placed close to the belt, and shielded from each other. The Thorium C" was collected on the belt which was negatively charged relative to the button and was carried along under the counter; as before, when equilibrium was reached, the Thorium C" under the counter was continuously replenished, and decayed effectively, as far as the counter was concerned, with the halfvalue period of Thorium B.

Owing to the difficulty in reproducing the geometry and scattering conditions prevailing in the work on Thallium and





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Radium E, it was decided that none of the above methods could be applied satisfactorily in the present investigation and another method of maintaining the strength of the Astinium C" source was sought. Devons and Neary (12) used separate sources of Radium C" taking a "control" reading for each source and expressing the intensity of each absorber reading in terms of this reading. A similar method was therefore adopted for the measurement of the Actinium C" spectrum and the following source changing apparatus was devised. A source holder was mounted at each end of a strip which could be rotated about a horizontal axis, so that while one source was being counted, the other was being activated. The apparatus is illustrated in Fig. 15. Two persper plates 12" x 4" x 1" wure separated by two uprights 4" x 21" x 1", forming an open sided box; brass bearings to take a 1" diameter brass rod were screwed to the centres of the horizontal plates. and two holes 1" square were out in the top plate, with their centres 44" on either side of the bearing. A strip, 10" x 1" x 1", was fitted with a brass collar at its centre and carried a piece 1" x 1" x 1" at either end, on top of which were situated further pieces, 1" x 1" x 3" turned out on top, in the same way as the source holders used previously. A $\frac{1}{4}$ " diameter brass rod passed through the bearings and the collar; a light spring was inserted between the bottom plate and the narrow strip. A bakelite knob was attached to the top of the rod and the collar was secured to the rod by a grub-screw. In its upper position the strip lay flush along the underside of the upper plate, with the first square blocks, filling the holes in this plate, and with the $\frac{2}{32}$ thick squares, projecting above. These latter squares were the

sources. Over one of the holes, was fitted the counter assembly used previously, with the bottom square plate removed, and over the other was situated a brass pot, the lid of which was drilled centrally to receive one of the brass Actinium B buttons. The two sources were connected below the persper strip by a fine wire which passed up through the centre of the source blocks, the ends being flush with the bottoms of the recesses; the area of metal exposed was thus too small to give appreciable scattering. The wire was connected to a terminal on the base plate by a length of flexible stranded wire coiled round the central spindle and the bottoms of the recesses in the top of the end blocks, were rendered conducting by painting with a suspension of colloidal graphite in acctone. The terminal on the base was earthed and the Actinium B button was connected to a positive 2000 volt supply (the stabilized high voltage supply described in Appendix III was used), thus making the graphited sources negative relative to the Actinium B button. After a few minutes activation, the knob was depressed, rotated through 180° and released, thus placing the active source under the counter, and the other under the Actinium B button. In this way, a source could be changed and counted, two or three seconds after activation was completed, and another source placed in the activator at the same time. This arrangement was highly satisfactory in operation, and reproduced precisely the conditions prevailing in the experiments with Thallium and Radium E. Two strips of $\frac{1}{2}$ " x $\frac{1}{2}$ " x $\frac{1}{16}$ " angle aluminium (not shown on the diagram) were fixed on top of the upper plate, to prevent it bending under the weight of the lead turret round the counter. It was not

found necessary to provide lead shielding round the activation pot; the brass tube was sufficient to out off all β -rays from the Actinium B button and the δ -rays from Actinium active deposit are very weak (13); no appreciable increase in counter background was recorded when using this arrangement. A sufficiently high counting rate (about 600/sec.) was obtained from a 15 minute activation of one of the sources from an Actinium B button activated in the pot for 24 hours. A check of the half-value period, taking counts of 30 seconds duration, illustrated in Fig. 11(c) gave a value of 4.59 minutes.

In order to verify the constancy of collection of the Actinium C" a series of readings was taken, activating the Actinium C" source for 20 minutes and counting it immediately for 30 seconds; the results are shown in Fig. 12. The points lie close to a straight line, after equilibrium has been established, the straight portion giving a half-value period of 36.4 minutes, sufficiently close to the 36.1 minutes of Actinium B for it to be said that constancy of collection was good.

No advantage was found in using an Actinium C" source for more than 10 minutes (roughly 2 half-value periods) and the Actinium B source was changed at approximately hourly intervals, two buttons being used, one activating and the other in the counting assembly.

Owing to the short life of Actinium C", all readings were made of 1 minute duration, and spaced at minute intervals. The Timing Unit Type 1003K was particularly valuable in this connection, since when set to time a count for 1 minute, after being reset to zero, it switched on the next count, one minute after the end of the

previous one. There was thus one minute for recording the number of pulses counted, resetting the apparatus to zero and making any desired absorber changes. The accuracy of the time between readings was the same as that of the duration of the readings (viz. the error was less than $\frac{1}{50}$ sec.)

A "control" count was taken for each freshly activated Actinium C" source, with no absorber in place, and the counts taken with absorbers in place, using the same source, were expressed as a fraction of the "control" count, after they had been corrected for Probe quenching time, background and source decay, in that order. Since all the counts were of the same length and were finally expressed as a fraction of another count, the problem of the error involved in attributing the counting rate to the mid-point of the reading, did not arise.

The control count with each source was in every case taken first, since the accuracy of the ratio of each counting rate to the "control", depended quite appreciably on the accuracy of the control and any decrease in the statistical error of the other counts would have been offset by the decrease in accuracy of the "control", if the latter had been the last reading taken with each source.

At first, a control and four readings with absorbers were taken with each source, but this was later reduced to a control and two absorber readings. When the accuracy of the control count was reduced to 1% by the decay of the Actinium B button, the latter was replaced by a fresh one; in practice, this was done every 45 minutes.

Each point on the absorption curve of Actinium C" (Fig. 8(c))

is the result of taking the mean value of at least 5 readings corresponding to the same absorber thickness; between $401 \cdot 9 \text{ mg/cm}^2$ and 526 mg/cm², every point represents the mean of 9 determinations. The intensity of the control count, corresponding to the counter window and air thickness of 7.7 mg/cm² is taken as 10,000 units and all other results are expressed relative to this reading.

The accuracy of each point decreases towards the lower end of the curve, but errors are in general, too small to insert, without confusing the graph. Three typical points have the following standard errors on the vertical scale: 46-2 mg/cm², 0.56%; 161.7 mg/cm², 0.916%; 324.9 mg/cm², 2.4%. The method adopted for background correction, viz. subtraction of the mean background from the counting rate corrected only for Probe quenching time, becomes decreasingly valid as the counting rate approaches the background counting rate, although for the upper 2 of the curve, little error is introduced. However, in the part of the curve beyond the β -ray end-point, the counting rate was very little in excess of the background, even with the strongest source available and it was necessary to provide more accurate points in this region. Four points were taken, repeated 12 times and each absorber determination was immediately preceded by a control count. Counts from the previous work, which were taken 2 minutes after a control count, were also included in the calculation. After correction for Probe quenching time, the sums of the control counts and of the absorber counts were taken and the mean background for the time of the sum, subtracted from both. The summed count with absorber was then corrected for two minutes

TABLE 14

All counting times were of 1 minute duration N observed counting rate per second . N counting rate corrected for Probe quenching time -B = background counting rate time after reading taken with bare source (in t minutes) $(N-B)_{o} =$ fully corrected counting rate relative to t = 0. P fractional intensity, expressed in terms of the intensity due to a bare source A fresh source was used for every reading taken with a bare source (7.7 mg/cm² absorber, the thickness of the counter window and air (N-B) mg/cm² of Al. N' N-B No./min. t F N 7.7 0 20727 345-5 400.9 400.7 400.7 1 41-2 13161 219.4 2 0.803 240.5 240.3 321.6 41.2 9920 165-3 177.0 176-8 316-2 0.790 4 7.7 21511 358.5 418.5 418.3 0 418.3 1 34.5 14426 240.4 266.0 265.8 2 355-8 0.850 11036 34.5 183-9 198.5 198-3 0-848 355.0 4 7.7 19667 327.8 377.3 377-1 0 377.1 1 27.8 13661 0-888 227.7 250-5 250.3 2 335-0 27.8 10396 186-2 186.0 0.883 173.3 4 332.8 7.7 17832 297.2 337-3 337.1 337-1 0 1 21.1 12885 214.8 235.0 234-8 2 314-3 0.933 21.1 9938 165.6 177.4 177.2 4 317.5 0.942 7.7 17646 1 294.1 333.1 333.3 0 333-1 14-4 13230 220.5 241.8 2 241.6 323.4 0.970 166.3 178.2 178.0 318.6 0.958 9975 14.4 4

decay and expressed in terms of the summed control count. The four points thus obtained are shown with their errors on the graph, and the line representing Y-emission (bremsstrahlung) is drawn through these. The method used previously necessitated the discarding of all results smaller than the mean background. This is not a valid procedure when the counting rate is low since it implies dropping the smaller values in the statistical distribution, thus giving an artificially high value to the mean. It was not possible to retain them, however, using the initial method, since each reading was multiplied by a factor greater than unity, to correct for decay, and the negative values obtained by subtracting the mean background would therefore be made more negative. It is equally incorrect, in such circumstances, to subtract the mean background, since a low total count is probably the result of a low true count and a low background count during the counting period. The method used in determining the four points at 726-8 mg/cm², 803-8 mg/cm², 880-8 mg/cm² and 967-0 mg/cm² is perfectly valid, however, In the present investigation, it was not considered worth while to increase further the accuracy of these four points: the measurements taken, extended over about 5 hours and to decrease the statistical deviation to 5% would necessitate counting for at least 180 hours (The errors in the four points are rather greater than 30%). If a more detailed inspection of this part of the curve is required a much stronger source is desirable.

The method of correcting the results is shown in Table 14.

5. SEARCH FOR LOW ENERGY &-PARTICLES

An investigation was made of the absorption curves of the three elements at very small thicknesses of absorber. The results cannot be compared directly with the previous ones, since scattering and solid angle conditions differ rather widely in the two cases. The apparatus used was as follows:

A cylindrical brass box, about 8" in diameter and 2" in depth, carried on its upper surface near the outside wall a counter open to the inside of the box. Immediately below the counter, a ground brass plug fitted into the bottom plate, from below. Inside the cylinder, a brass disc could be rotated from outside. through a ground joint; the disc was drilled through. with a number of 1" diameter holes near its rim, and across these. aluminium absorbers were waxed. By rotation of the disc, any one of the absorbers, or a blank hole could be interposed between the counter and the source, which rested on the ground plug. The whole apparatus was vacuum-tight and was filled with an argonalcohol mixture in the proportion of 6 cm. pressure of argon and 1 cm. of alcohol. Since the counter was windowless, when the blank hole in the plate was between the source and counter, the counter gas between them was the only absorber, this being about 8 x 10⁻² mg/cm² in thickness.

The sources were contained in a recess $\frac{1}{2}$ " in diameter and $\frac{1}{64}$ " deep in the centre of a disc of perspex, $1\frac{1}{4}$ " in diameter and $\frac{7}{32}$ thick, thus giving approximately the same back-scattering conditions as in the principal experiment. The plateau of the counter was reasonably good and was about 100 wolts in length.


It has been reported by some workers, and has been the general experience at Edinburgh University, that the plateau of a counter changes during the 24 hours succeeding filling. With the counter described above, however, no appreciable alteration was recorded over the first three hours, sufficient for the completion of readings with any of the three active materials.

Thallium. A source prepared in the manner described earlier, was inserted in the box which was then pumped for ten minutes, and filled with the argon-alcohol mixture. The plateau of the counter was determined and the working voltage set at a point $\frac{1}{2}$ of its length from its lower end. The absorption curve was then determined, working from zero absorber to the maximum and then repeating in the reverse direction. The absorption curve (Fig. 16(a)) was plotted with the mean of the two results for each absorber thickness. Corrections were applied for Probe quenching time, and background, but no allowance was made for decay since the total duration of the experiment was only about three hours. Radium E. The first Radium E source, prepared by precipitation with Neodymium carrier, as before, was treated in precisely the same way as the Thallium, no correction being made for decay, other than the averaging of the two sets of results; the total duration of the experiment was 1 hour, during which the expected fall in activity was about 0.58%, so the above procedure was justified.

The absorption curve (Fig. 16(b)) showed a sharp rise at small absorber thicknesses, believed to be due to \measuredangle -particles from the daughter Polonium, the presence of which was a decided possibility, since it had not been possible to count the Radium E source in the "box-counter" until two days after its preparation.

The source was removed and recounted four days later, when the sharp rise was still found to be present, but was greater in height relative to the rest of the curve (Fig. 16(c)), proving that this part did not have a 5 day half-value period, and was almost certainly due to Polonium. A further check was made with an α -particle counter of the improved Rosenblum type (14). This counter is sensitive to α -particles only and has a negligible background (less than 10 counts per hour); in spite of the small solid angle, it gave 4 counts/sec., corresponding to approximately 0.1 microcuries of Polonium, which was of similar order to the original strength of the Radium E source, estimated at about 0.3 microcuries.

Having allowed the Radium E to grow in the Radium D solution for a week, a silver foil was immersed in the Radium D for two days to remove Polonium, followed by a copper foil for about 18 hours; the separation of the Radium E was then carried out as described earlier. The same counting procedure as before was followed, starting within two hours of the end of the chemical work; the total counting time being 70 minutes, there was no necessity to correct for decay, other than by averaging the two sets of results. There was no evidence of a sharp rise in this curve, definitely proving that the shape of the previous curve was due to Polonium α -particles. The results are shown in Fig. 16(d).

Actinium C". This material provided a more difficult task than Thallium or Radium E on account of its rapid decay. It was decided to dispense with a set of readings increasing the absorber thickness, since, with the decay working in the same

direction, it would be difficult to obtain reliable results. The graphited perspex button used in the initial experiments on Actinium C" was used, and was held in a recess in a disc of persper, similar to those used for Thallium and Radium E, but modified so that the top of the button was $\frac{1}{6L}$ below the top surface of the disc, thus reproducing the same conditions as those prevailing with the other sources. The button was activated for 10 minutes from an Actinium B source which was in equilibrium with the Actinium in the pot; the activating potential was 2000 volts and the buttons were spaced at 1 mm. The button, in its holder, was inserted in the counter, with the pump running, and counting was started less than 5 minutes after the activation of the source was stopped. As in the previous work on Actinium C", counts were made of 1 minute duration with 1 minute between each. Two sets of readings were taken, both starting with the thickest absorber: a fresh source was used for the second set. The two absorption curves were similar and one is shown in Fig. 16(e).

There is no evidence of a very low energy group of electrons in any of the three active materials, although all three show a slight upward trend of the curves, towards low energies. There is a greater probability of scattering with this arrangement than with the perspex assembly used in conjunction with the thin-window counter, and this probably accounts for the majority if not all of the upward curvature which is definitely absent in the corresponding region of the curves taken with the end-window counter.

The box-counter allows the possibility of scattering, and consequent loss of energy, of electrons from the edges of the holes

in the disc, across which the absorbers are fixed and also from a bridge across the end of the counter itself (this bridge is of brass, and is approximately $\frac{1}{16}$ " wide and $\frac{1}{8}$ " deep, extending right across the $\frac{3}{4}$ " diameter of the mouth of the counter; the end of the counter wire is anchored to a ring $\frac{1}{8}$ " in diameter in the centre, by means of a small glass rod insulator).

The disagreement between the shapes of the overlapping portions of the curves obtained with the end-window counter and the "box counter" can almost certainly be attributed to differences in scattering conditions, the former results being considered the more reliable. It is safe to say, however, that there is no evidence of any abnormal quantity of very low energy electrons emitted by Thallium, Radium E or Actinium C".

6. ANALYSIS OF THE RESULTS

The first analytical procedure, was to obtain curves for the β -activity of the three elements. This was done by extrapolation of the straight lower portions of the curves, back to zero absorber thickness. Sargent (2) extrapolates with a curve, rising more sharply at smaller absorber thicknesses, while Feather, in all this type of work, uses linear extrapolation. The latter method, while possibly not quite so true as that of Sargent, is a safer procedure, and since, in the region where Sargent's curve turns upwards, the Y-intensity represents a small fraction of the total intensity, in most cases yields a result differing negligibly from Sargent's. For these reasons, Feather's linear extrapolation was used. The intensity of the Y-ray



component, was subtracted from the total intensity of both 6- and Y-rays, at convenient intervals on the horizontal scale (10 mg/cm² for Thallium, and 20 mg/cm² for Radium E and Actinium C") deriving the total $\beta + Y$ intensities from the smooth curves of Fig. 8. The graphs of the resulting A-components were replotted on a logarithmic intensity scale (Fig. 17), with all three passing through the same point on the intensity axis, this point being taken as unity. The ranges in aluminium of the β-particles from Thallium and Actinium C" were then calculated, using the method described by Feather (1) and taking Radium E as the reference body on account of the well-authenticated value of 176 mg/cm² for the absorption limit in aluminium of its 8-particles. Bleuler and Zunti (3) criticise the choice of Radium E as reference body, on account of its large surplus of low energy electrons, and consequent initial steep fall of the absorption curve. They disagree also with the extrapolation of the absorption curves to zero absorber thickness and seem very doubtful of Feather's extrapolation method. However, Feather's method does not depend, at all, on the form of the standard absorption curve, and the accurately known value of the absorption limit for Radium E β -particles is a strong reason for choosing this material as reference body.

Following this method, therefore, the range of Radium E was divided into twenty equal intervals, and horizontal lines were drawn through the Radium E curve at its intersections with these intervals; the thicknesses of absorber corresponding to the intersections of the horizontal lines, with the other two curves were then read from the graphs. The "calculated absorption



limits" derived from each intersection, were plotted on a linear scale in Figs. 18 and 19. (The derivation of the calculated absorption limits, is as follows: consider the horizontal line corresponding to $\frac{1}{n}$ of the range of Radium E. The calculated absorption limit for the corresponding point on the curve under examination, is $\left(\frac{20}{n}\right)r_n$ where r, is the abacissa of the intersection of the horizontal line with the curve). Smooth curves were then drawn through the points thus obtained, and extrapolated to n = 20. The calculated absorption limit corresponding to this point was therefore the value for the range in aluminium of the B-particles of the element under consideration. In addition, the form of the curve, gave an indication of the number of electrons of different energies, emitted by Thallium and Actinium C". relative to Radium E. A curve trending upwards, towards the greater range-fraction end of the curve, would indicate the presence of more low energy electrons than are present in the spectrum of the reference body, since the estimated range from the early points is too low; i.e., the absorption curve is falling more steeply at the beginning. A material having a precisely similar energy distribution to that of the reference body, would yield a horizontal straight line on the calculated range curve.

This analysis gave the value of $623 \pm 4 \text{ mg/cm}^2$ for the absorption limit in aluminium, of the primary β -particles of Actinium C". This is decidedly lower than that of 0.68 gm/cm² given by Sargent (2). The range in aluminium of the β -particles of Thallium (3.5 yr.) was found to be 300 ± 3 mg/cm², slightly





lower than the previously determined value, of 0.31 gm/cm (15).

From Feather's range-energy relation R = 0.543E - 0.160, the energies corresponding to the above ranges are respectively 1.441 \pm 0.008 MeV and 0.847 \pm 0.006 MeV.

The shapes of Figs. 18 and 19 indicate that Actinium C" and Thallium both emit fewer low energy electrons than does Radium E, the relative deficiency being much greater in the case of Actinium C". The latter result was to be expected since the B-emission of Actinium C" is allowed. Points plotted for a range-fraction of less than 0.05, did not lie on the smooth curve derived for Thallium, so a more detailed investigation was made in this region. The β -ray curves were plotted on a larger scale, covering a reduction in intensity by a factor of ten; these are shown in Fig. 20. The resulting curves for the calculated absorption limit are shown in Figs. 21 and 22. The points all lie on a smooth curve for Actinium C", but there are still deviations in the case of Thallium; however, the curve extends smoothly to a lower range-fraction than does Fig. 19 and the deviations of the remaining points are attributable to inaccuracies arising from reading the absorption curves in this region.

Sargent (2) proposed a method of analysis and range derivation rather similar to that of Feather, described above. Sargent's method consists in plotting the logarithm of the β -intensity against $\frac{m}{R}$ where m is the mass/sq. on of absorbing material and R is the extrapolated range, using a standard absorption curve as in Feather's method. The absorption curve of the material for which the absorption limit is to be found,



is then fitted to this curve at the end corresponding to the greater absorber thickness; according to Sargent this is possible since many β -spectra have the same shape between the maximum energy, and half of it.

The absorption measurements described above, were displayed on this basis; using the values for the ranges, found by Feather's method, the intensities were adjusted so that all three curves had unit intensity at an absorber thickness corresponding to $\frac{2}{3}$ of the range; the results are plotted in Fig. 23. The curves for Radium E and Thallium coalesce at approximately 0.45R, but the Actinium C" curve does not merge with the other two until approximately 0.65R. The experimental absorption curve of Actinium C" is however very different in shape from the other two, having a much smaller initial slope and it would seem therefore that the curves only become coincident at about 0.5R when the spectra are similar in shape; in the case of allowed transitions, such as Actinium C", where there is a smaller number of low-energy electrons, it must be assumed that the curves do not meet, until a higher range-fraction. The differences in shape of the absorption curves are however shown very clearly by Sargent's method, indicating that, of the three materials, Radium E has most, and Actinium C" has least low energy electrons, the same result as that given by Feather's method.

Very little has been described in the literature, on further analysis of absorption curves. It should, however, be possible to derive distribution curves from them. The method described on the following page is basically similar to that

used by Das Gupta and Chaudhury (6) for Radium E but has the advantage that the initial absorption curves are more accurate. Almost identical technique was also employed by Hughes, Eggler and Huddleston (7) in dealing with Be¹⁰.

The derivation of the formula for calculating the distribution curve from the absorption curve is as follows:

Let f(R) be the distribution with respect to range of primary β -particles emitted by a source; then the fraction of primary β -particles having ranges between R and R + dR is f(R)dR.

Early experiments on the absorption of homogeneous β -particles showed that: $I(x) = 1 + a(\frac{x}{R}) + b(\frac{x}{R})^2 + c(\frac{x}{R})^3 + \dots$ where I(x) is the fraction recorded through thickness x, of absorber, R is the range of the homogeneous β -particles and a, b, c, are constants (16). To a first approximation, this may be written: $I(x) = (1 - \frac{x}{R})$.

If A(x) is the fraction recorded through a thickness x, for a source having a continuous spectrum of primary β -particles, as specified above, then:

	A(x)	$\int_{x}^{\infty} (1 - \frac{x}{R}) f(R) dR \dots$	(1)
lence	A'(x)	$-f(x) - \int_{x}^{\infty} f(R) \frac{dR}{R} + x \frac{f(x)}{x}$	
		$- \int_{x}^{\infty} f(\mathbf{R}) \frac{d\mathbf{R}}{\mathbf{R}} \qquad \dots$	(2)

where dashes denote differentiation with respect to x.

The above applies when the intensity, A(x) is plotted on a linear scale. It is customary, however, to plot A(x) on a logarithmic scale, as has been done in the work described earlier. In this case, where $L(x) = \log_{a} A(x)$;

$$L^{1}(x) = \frac{1}{A(x)} A^{1}(x)$$
 or $A^{1}(x) = A(x) L^{1}(x) \dots (5)$

Hence $A^{*}(x) = A(x) L^{*}(x) + A^{*}(x) L^{*}(x) \dots (6)$ Substituting for $A^{*}(x)$ from equation (5);

$$A^{*}(x) = A(x) [L^{*}(x) + {L^{*}(x)}^{2}] \dots (7)$$

Hence, substituting in equation (4);

$$f(x) = x.A(x).[L^{*}(x) + {L^{*}(x)}^{2}]$$
.....(8)

The values of f(x), the distribution function relative to the absorber thickness, can thus be found directly from the absorption curve, by means of equation (8).

Suppose \$(E) is the distribution function with respect to energy:

then: f'(E)dE = f(x)dx or $f'(E) = f(x)\frac{dx}{dE}$ Hence the distribution function relative to energy, can also be found from the absorption curve.

The expression for f(x) given by equation (8) involves the first and second differentials of the absorption curve; the former can be obtained with reasonable accuracy, but the latter is more difficult to measure. Das Gupta and Chaudhury did not consider their experimental data sufficiently accurate to enable the latter to be obtained directly from the absorption curve and adopted a slightly different procedure for finding the second differential. (The expression used by them was that given in equation (4), their results evidently being plotted on a linear intensity scale). In the present case, however, it was decided to derive the first and second differentials







from the absorption curves, on account of the accuracy of the latter. This procedure was found to be justified, since, except at the low energy end of the curves L''(x) was generally of the order of 10% to 20% of $\{L'(x)\}^2$ the latter term thus exerting the greater influence on the value of the distribution function f(x) in equation (8).

The practical evaluation of the differential of the curve was carried out mumerically as well as in the following manner: at the points on the smooth absorption curve of Fig. 17, corresponding to a number of absorber thicknesses, the slope of the tangent to the curve was measured, by placing a straightedge to touch the curve, and reading the values (in mg/cm²) of the intersections with convenient powers of ten, on the logarithmic scale (e.g. the intersections of the straight-edge with 1 and 10⁻² on the vertical scale). The slope was then the number of cycles of the logarithmic scale, (two in the above case) divided by the difference of the corresponding values on the horizontal scale. The slopes thus obtained, were then plotted on a linear vertical scale, against the corresponding absorber thickness in mg/cm2; the values obtained agreed well with those obtained numerically. It was necessary to treat the points with a certain amount of freedom. in order to obtain a smooth curve. The graphs are shown in Figs. 24, 25 & 26.

The second differentials of the absorption curves were derived similarly from these curves. It should be noted that the slopes of the absorption curves in Fig. 17 are always negative, so the values of $L^{*}(x)$ and $L^{*}(x)$ obtained from









TABLE 15

×10-6 2069 1129 P(E) 1089 24,96 2538 2318 1658 1396 2057 0 0.345 0-255 0-394 0.430 0-453 £97+0 0-472 624-0 194.0 湖당 **部(x) = t(x)** (XeV) 193 248 297 ŧ 388 518 122 - All 127 431 -×10-6 4250 $U_{e}^{*}(x) | U'(x) + [L'(x)]^{2} P(x)$ 6000 6335 5900 1160 2960 5115 3580 2357 ρ 308-5 308-5 308-5 308-5 308-5 306-6 309-2 313.8 320-9 306.5 ×10-6 Results for Radium E x-1x10-6 0.06 2-86 16-08 12.20 $\mathbf{f}(\mathbf{x}) = \mathbf{x}_{n} \mathbf{A}(\mathbf{x}) \left[\mathbf{L}_{0}^{n} \left(\mathbf{x} \right) + \left\{ \mathbf{L}_{0}^{n} \left(\mathbf{x} \right) \right\}^{2} \right]$ 6.01 0 0 x-1 x10-6 $A(\mathbf{x}) = [\mathbf{L}_{io}^{*}(\mathbf{x})] \left\{ [\mathbf{L}_{o}^{*}(\mathbf{x})]^{2} \mid \mathbf{L}_{io}^{*}(\mathbf{x}) \right\}$ 10.1 2.3 6.98 1.24 2.61 0 Ċ 308-5 308-5 308-5 309-5 318-5 326.0 337-0 308+5 308-5 312.5 ×10-6 ×10-3 7.63 7-63 7.63 7.63 7.63 7.68 3.75 7-84 16-1 7.64 0-0828 0-0408 0-0589 0-167 0-122 0-486 0-342 0-239 0.689 mg/om 8 8 100 180 9 8 120 \$ 8 0 H

Figs. 24, 25 & 26 must be prefixed by a negative sign. The base of the logarithms in Fig. 17 is ten and for convenience, this has been retained in Figs. 24, 25 & 26. Conversion to the base e, was made subsequently. The distribution curves, with respect to range, are shown in Figs. 27, 28 & 29, a section of the table of results being given in Table 15.

The distribution with respect to energy was calculated, with the help of data given by Marshall & Ward (17) and Curie (18) from which a range-energy curve was constructed; the values of energy and of $\frac{dR}{dE}$ corresponding to the relevant absorber thicknesses were obtained from this graph and the latter used to transform f(x) into $\varphi(E)$. The resulting energy distributions are shown in Fig. 30. A further comparison, illustrated in Fig. 31, was made as follows; the three curves of Figs. 27, 28 & 29 were plotted with the absorber thickness expressed in terms of the range of the β -particles from each body (obtained earlier), the areas under the three curves being maintained at unity. This graph thus illustrates the way in which the maxima of the three distribution curves are related to their ranges.

The energy distribution curve of Fig. 30, for Radium E, does not agree with the Fermi distribution (Fig. 32) but does fall below it at the high energy end; this is known to be the case for Radium E. It also departs markedly at the low energy end from the distribution curve (Fig. 33) obtained with a magnetic spectrograph (19 and 20).

The calculated distribution curves give the correct endpoint energies for the three materials, but the maximum of the Radium E curve (the only one for which spectrograph data is



available), falls at too high an energy; this is to be expected, since the initial absorption of homogeneous β -particles is slower than that given by the linear relation $I(x)=1-\frac{x}{R}$. Experimental curves obtained by Marshall & Ward (17) are of the form shown in Fig. 34, while the linear relation used, is given by the line AB. The line AC gives a better approximation to the curve and slightly reduces the energy corresponding to the maximum of the distribution curve, but also reduces greatly the end-point energy; the line AC is not therefore a valid approximation.

Clearly, the true state of affairs can only be approached by considering more terms in the expression, $I(x) = 1 + a(\frac{x}{R}) + b(\frac{x}{R})^2 + ...$ for the absorption of homogeneous β -particles; this, however, introduces differential coefficients of higher order than the second, and these can not be obtained with any reliability from the absorption curves.

Hughes, Eggler & Huddleston (7) have obtained very similar results with Gu^{64} , Au^{198} , W^{185} and RaE comparing the energy distributions, derived from absorption curves in a similar manner to that described above, with magnetic spectrograph data. They found that the end-point energy was correct in all cases, but the maxima of the calculated curves occurred about 100 KeV higher than expected. To overcome this difficulty, they propose the empirical method of shifting the calculated distribution by 100 KeV for an end-point energy of 600 KeV, when the maximum falls at the correct energy. The objection to this procedure is that the end-point energy is then low by the amount of the shift. The distribution obtained (Fig. 30) for Radium E agrees

fairly well with these findings since the maximum of the curve is about 200 KeV too high; this is double the value reported et al. by Hughes, but the end-point energy is also approximately double that for which they propose a shift of 100 KeV.

7. SUMMARY

Experiments have been described, in which a direct comparison is made, by an absorption method, between the β -particle spectra of Actinium C", Radium E and Thallium (3-5 yr.), representing allowed, second forbidden and third forbidden transitions, respectively.

New values of $623 \div 4 \text{ mg/cm}^2$ and $300 \div 3 \text{ mg/cm}^2$ are proposed for the ranges in aluminium of the β -particles emitted by Actinium C" and Thallium; it is shown that, of the three elements, Radium E emits most and Actinium C" least low energy electrons. It was to be expected that Actinium C" should emit least, since its β -transition is allowed, but the fact that Thallium (third forbidden), definitely emits less than Radium E (second forbidden) is new knowledge.

A search for β -particles of very low energies revealed that no abnormal number is present in the spectrum of any of the three elements.

An attempt has been made to derive energy distributions from the ebsorption curves, but the results for Radium E depart markedly from those obtained with magnetic spectrographs. It was not possible to check the other two distributions owing to the lack of information on them. The general findings, however,

fit with those of Hughes, Eggler and Huddleston (7), who recently published an account of similar work.

The formula used in the derivation of the energy distributions, includes an approximation which gives a deficiency of low energy particles, but the accuracy of the data is not sufficient to permit the use of a better approximation. The method does, however, give an indication of the relative energy distributions for the three elements considered.

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APPENDIX I

- C-12

Absorption Measurements of the Y-Rays from Ir

PARKCIP

1.

FIG.35







The absorption method of determining the energies of Y-rays can often be used when the sources available are too weak to give useful results with a magnetic spectrograph. The resolution is not high, but the average values of groups can readily be determined. There follows an account of experiments carried out on Ir^{192} .

The Iridium was in the form of $(NH_4)_2$ Ir Cl₆ and was activated by neutron bombardment in the pile at Harwell. A thin, uniform layer of the material was contained in a recess in a round brass "button" of $\frac{1}{2}$ " diameter, and was commented in with a solution of distreme in bensene. The "button" was sorewed into a hole in the centre of a $\frac{1}{4}$ " brass base-plate into which four brass pillars were also sorewed. These served to locate three lead blocks 5" x 3" $x\frac{3}{4}$ " drilled centrally with a hole 0-3" in diameter; collars were inserted between the first and second blocks from the bottom, to allow absorbers to be inserted. The counter was clamped in a suitable cradle, to a brass plate $\frac{1}{8}$ " thick, again with a central hole 0-3" in diameter, located by the pillars in the same way as the lead. The whole formed a rigid, reproducible collimating assembly and is illustrated in Fig. 35.

The electronic apparatus consisted of a Type 200 scale-ofhundred, a Type 200 Probe Unit and a Type 200 Power Unit, all of T.R.E. design.

Three counters were used in the determinations, two sets of results being taken with the first, and one each with the other two. The first, of the type shown in Fig. 36, had a copper tube, $\frac{1}{2}$ in diameter and of wall thickness 0.45 mm as cathode. It was placed, horizontally, above the hole in the lead assembly, with





its centre line as shown in Fig. 35. The other two were of the G.E.C. thin-window type. They had copper walls and duralumin endwindows through which the radiation entered, passing axially along the counter.

The resolving times of the counters were determined by a method in which two sources were used, singly and together; the combined counting rate was smaller than the sum of the counting rates using the sources singly and from this loss, and the counting rates, the resolving time was calculated (1). The input of the scaler was such that it could be used with a resolving time of 5 microseconds or could provide a paralysis time which could be preset to a value within the nominal range 300 microseconds to 1000 microseconds. It was found that in the former case, doubles were frequently produced so it was necessary to use the second input; this eliminated doubles, but the resolving time was increased.

In these measurements, it was observed, that at high counting rates, the resolving time decreased as the counting rate increased. A typical set of results is illustrated graphically in Fig. 37; here, resolving time is plotted against the counting rate due to both sources together. This effect has been reported by Muchlhause and Friedman (2), but their investigations were concerned with the resolving time of the counter itself while in the case described above, the paralysis time of the input circuit of the scaler is the governing factor. It seems, therefore, that the paralysis time of the flip-flop is affected by the counting rate.

The principal experiments were of normal type. The counting rate was observed for varying thicknesses of lead absorber, the








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results being plotted graphically with the counting rate on a logarithmic scale (Fig. 38). An asymptote representing the hardest component was then drawn to the curve at its lower end. By subtraction of this line, from the experimental curve, a similar graph (Fig. 39) was obtained, this then being treated in the same way, the final component being shown in Fig. 40. From the straight lines, the absorption coefficients and hence the energies of the component Y-rays were calculated. Since the absorption edge in the energy-absorption coefficient relation for lead, was so placed as to cause ambiguity in the energy of the softest component, it was necessary to carry out an absorption measurement with another material. Tantalum was used, but only sufficient was available to allow the plotting of the top end of the curve (Fig. 38(e)). However, the ambiguity was removed, since the Tantalum and Lead gave very similar slopes at the top end of the curve, indicating the correct value of the energy of the soft (The other possible energy indicated by the absorpcomponent. tion in Lead, had an absorption coefficient about three times as great, in Tantalum, so the initial slope of the latter curve would have been much greater, if this had been the relevant energy).

The wall thickness of the first counter was sufficient to cut off the β -rays from the source, but when the two thin-window counters were used it was desirable to ensure that the β -ray intensity should be reduced as much as possible. The collimator was altered slightly by placing the three lead blocks at the bottom, followed then by the spacing collars and the brass plate carrying the counter. An Alnico magnetron magnet was placed with its poles on either side of the lead blocks; the magnetic field



The curve shows the path of a Bray having the maximum energy of O.56MeV for Ir in the magnetic field of 657 cerstels

FIG.4I



was sufficient to ensure that no β -particle from the source could enter the counter without first striking the wall of the collimator. Fig. 41 shows the path in the collimating assembly of a β -particle having the end-point energy for Iridium. A small amount of scattered β -radiation still entered the counter as is shown by the sharp rise at the top end of the absorption curves taken with the thin-window counters (Figs. 38(c) & (d)).

The energies of the Y-rays were estimated also by a transition method, in which thin foils of a heavy metal were placed in the Y-ray beam, and the ensuing photo-electrons absorbed in aluminium. The experimental arrangement was as follows: the collars were inserted between the second and third lead blocks (numbering from the bottom) and a thin-window counter was fitted on a bridge above the top lead block, with its window $\frac{1}{2}$ " above the top plate of the collimator. A copper foil (10 mg/cm²) was placed on the second lead block, and on top of this a graphite plate (365 mg/cm²) in order to remove Iridium K & L radiations, and /3-rays respectively. To remove any electrons ejected from the collimator sides, a further plate of carbon (146 mg/cm²) was sited on top of the counter-carrying plate, and on top of this the heavy metal and aluminium absorbers were placed. There were 9 gold foils, each of area density 8.1 mg/cm², and the effect of adding these was investigated together with the effect of more than one copper foil; the result of adding gold foils is shown in Fig. 42(a). There was no significant difference caused by additional copper foils, and the best emission of secondary electrons was obtained with all nine gold foils. The absorption graph showed a discontinuity when several thin aluminium foils were replaced by a single one of





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approximately equivalent thickness. This effect was attributed to the ensuing change in geometry and an attempt was made to pack the thin foils more closely by placing a heavy square brass frame on top. A slight improvement was obtained as can be seen by comparing Figs. 43 and 44 ; the former was taken without, and the latter with the brass frame in position. These graphs confirmed the two harder components, but gave no information on the softest. A single gold foil (8.1 mg/cm²) was tried (Fig. 45) but again there was no evidence of the soft component. The graphs were plotted with the counting rate on a linear rather than a logarithmic scale, to show up, more sharply, the changes of slope due to the electrons of different energies. The energy required to remove an electron from the K. shell of gold was rather high, so further measurements were made using tin as a secondary emitter, instead of gold. As before, the effect of different thicknesses of tin, was tried, the results of this being shown in Fig. 42(b). Two thicknesses of tin were tried, 53-5 mg/cm² and 23.2 mg/cm2; as before, the two harder components were authenticated, but the soft one was not in evidence. (Figs. 46 and 47)

In all five of these graphs, there is a decrease in slope at the top, which is normal in /3-ray absorption curves; it does, however, fall in the region where the soft component would be expected, and probably masks the effect of it. It was on this account that readings of higher statistical accuracy were not carried out with this method.

The results of the four direct Y-ray absorption measurements showed quite good agreement with each other. The values of the mass-absorption coefficients in lead and the corresponding Y-ray

energies are shown in the following table:

	Counter							
	Copper		Copper		Thin		Thin	
	cylindrical		cylindrical		window 1		window 2	
	1%	E(KeV)	1%	E(KeV)	1%	E(HeV)	1%	E(KeV)
Hard component	0.125	618	0-126	615	0.118	592	0-119	592
Middle component	0.301	331	0-302	331	0.303	331	0-317	321
Soft component	2.73	146	1-62	191	2.76	145	2-77	145

An estimate was made of the relative intensities of the three groups of Y-rays. The counter efficiencies for the three energies were taken from a paper by Bradt (3) in which curves are given for counters with lead, brass and aluminium cathodes. The first counter used in the above experiments had a copper wall, so the curve for brass was taken. The thin-window counters were of copper with a duralumin window, but this was so thin (0.001 in.) that the Y-rays were more likely to produce their photo-electrons from the copper wall, and the curve for brass was therefore used in this case also.

The counting rates given by the extrapolation of the straight parts of the absorption curves to zero absorber thickness, were corrected for the counter efficiency relevant to the quantum energy of the Y-ray and the resulting counting rates were expressed in terms of the hardest component, this being chosen on account of its greater accuracy. The ratios of the intensities are given in the following table:

	Counter						
	Copper	Copper	Thin	Thin			
	cylindrical	cylindrical	window 1	window 2			
Hard component	1	1	1	1			
Middle component	2•54	2•47	2.88	2•19			
Soft component	0•32	0•27	0.17	0•22			

TABLE 16

Energy of Y-Ray KeV Levy	Energy of Y-Ray KeV Cork	Energy of Y-Ray KeV Hill & Meyerhof		
		137		
199				
202	1.15 1.17.16			
205		아이슈 아이는 것은		
206	1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1	208		
요즘 같은 말 같은 말 같은 말 같이 많은 말 같이 많다.	269			
296	294	296		
308	306	308		
316	315	317		
	401 7			
	408 7			
San San San	454 ?	a straight sta		
469	466			
Section and the	477			
488				
	586			
592	601			
608	610			
615				
	651			

Agreement is reasonably good, although the values for the soft component deviate rather widely from each other. This is to be expected since the accuracy for this component is not as high as for the other two.

Taking the averages, the determinations give three γ -rays of energies 604, 329 and 157 KeV and of relative intensities 1: 2.52: 0.25.

The energies obtained from the less accurate transitioneffect experiments confirm the two harder components, the average values after addition of the K-electron emission energies of the radiators, being 535 KeV and 323 KeV, but these experiments showed no evidence of the low energy component; any change in curvature due to this component would fall, however, in the region where the slope of the β -ray curve decreases, and would therefore be masked by this effect.

Reports by several workers indicate that there are many Y-ray lines in the spectrum of Ir^{192} as determined by magnetic spectregraphs and it is rather difficult to fit the results given above with these values. Levy (4), Cork (5) and Hill and Meyerhof (6) all give a list of several energies derived from magnetic spectrograph determinations.

Table 16 gives the average energies of the converted Y-rays as determined by these workers. The hardest component found by the absorption experiments is probably the average of the Y-rays of energies above 586 KeV; the middle component fits reasonably well with those between 294 KeV and 317 KeV while the value of 157 KeV for the soft component can be associated with the range of energies up to about 200 KeV. The absorption method should have

been capable of resolving a component having an energy of the order of 450 KeV, indicated by the results of Cork and Levy, but there was no evidence of it.

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4.17 п 91.9 ling dirmits 3.57 2 i ir $\sim 10^{-10}$. . .





FIG.48(b)



In the early days of the Geiger-Muller counter, the limitation on the resolving time of the system was imposed by the recording apparatus, often a thyratron scaling circuit. The advent of the hard-valve scaling circuit reversed the situation, and the resolving time of the counter became the governing factor. The average modern hard valve scaler has a resolving time between 5 microseconds and 10 microseconds, while a typical figure for a counter is about 100 microseconds. The electron multiplier has, however, created a demand for a much faster scaler, since its resolving time is considerably less than 0.1 microseconds. In addition, there seems to be a possibility of reducing the resolving time of a Geiger-Müller counter, if a smaller output pulse can be accepted. Muchlhause and Friedman (1) describe experiments in which they interposed a video amplifier, of 0.01 volt sensitivity and 2 Mc/sec bandwidth, between a counter and scaler, with a regulting reduction in the resolving time of certain small counters to 6 microseconds. They describe a scaleof-two, which they used in this work, having a resolving time of 1 microsecond. This is still too long to obtain the full benefit from an electron multiplier, but is a step in the right direction.

The following high-speed scales-of-two were developed for use with an electron-multiplier.

For test purposes, a simple pulse generator (Fig. 48a) was built. In this, the 1 volt r.m.s. output from a signal generator was amplified and passed to a pentode biassed so as to cut off the top and bottom of the sine wave, making it approximately square. After differentiation, the resulting waveform was amplified again in a single stage of about 5 Mc bandwidth. The output pulse shape

FIG 49 (a)



FIG.49(b)



was not good, but was reasonably satisfactory between 100 Kc/sec. and 5 Mc/sec.

Considerable development was done with this pulse generator, but, as might be expected, it was found that the trigger circuits were rather sensitive to the input pulse shape, so an improved pulse generator (Fig. 48(b)) was constructed. As before, the 1 volt r.m.s. output from a signal generator was amplified, but a stage of higher bandwidth than in the first pulse generator, was used. The output of this fed a snap circuit: this was a trigger circuit which stayed in its second state (with V, conducting), only as long as the grid of V, was held below out-off. The square wave produced by this stage, was differentiated and fed to a valve which acted as a cathode follower for positive output, and a low gain amplifier on negative output, either sign of pulse being selected by a switch. This pulse generator was a great improvement on the first one; at a pulse repetition frequency of 500 Kc/sec. the output pulse had an amplitude of up to 15 volts, and a total duration of about 0-2 microseconds; the rise time was about 0.05 microseconds.

The first scaler was based on that described by Muchlhause and Friedman (1). It was an orthodox Eccles-Jordan circuit (Fig. 49(a)) using Mullard type R.L.7 valves (Service type V.R.136), chosen because they had the highest figure of merit of those available in the laboratory at the time. Matching of components was found to increase reliability and all pairs were selected to within 5%. The minimum size of coupling condenser was about 10 pF; in the final form of the circuit, 12 pF Erie Ceramicon condensers of 5% tolerance were used.

Initially, the screen-grids of the valves were run at full H.T. potential, but it was found that the maximum speed of operation was increased considerably if this was reduced. The reason for this is not obvious, since the mutual conductance of the valve is greatly reduced. The probable solution is that the voltage swing is reduced, and recovery is thereby accelerated. It would, therefore, seem better to use smaller anode loads and keep the screen potential high.

Following the circuit of Muchlhause and Friedman, condenser input was used at first, but this has the disadvantage of introducing additional capacity into the grid circuit, and also causes some attenuation, resulting in the need for a fairly large input pulse. In the Type 200 scale-of-hundred designed by T.R.E., interstage coupling is carried out by double-diodes feeding the anodes of the trigger circuit, the coupling condensers passing the signal to the grids; the cathode of the double-diode is connected to a point on the output anode load of the previous stage. A similar method was applied to the circuit described above, but the anodes of the double-diode were connected to the trigger circuit grids and the cathode connected to earth through a resistance: the input was applied to the cathode of the doublediode. This modification, shown in Fig. 49(b) reduced the input required, from 10 volts to about 3 volts, and had the advantage of making the circuit sensitive to negative pulses only, instead of to either sign.

The performance of the scaler was tested by the pulse generators described earlier. The maximum frequency of operation was about 1.4 Mc/sec, with a positive H.T. supply of 350 volts,



FIG.50

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a negative line voltage of -300 volts, and with the screen grids at 95 volts. The output wave-form was triangular rather than square, with an amplitude of about 10 volts.

The second scaler was again of conventional design, but different values were used, the E.F.55 (C.V.173) being chosen on account of its higher figure of merit and higher anode current, allowing more rapid charging of condensers. The circuit is shown in Fig. 50. Diode input was used from the start, in this scaler, and in order to reduce the negative line voltage, the grid resistance was made smaller than the coupling resistance, both being fairly large to reduce the current drain through the path $R_A R_c R_c$. As before, components were matched to 5%.

Reduction of screen potential was not carried out to such an extent as in the first scaler, since it was decided to make full use of the high mutual conductance (g = 12) of the E.F.55, and make the anode loads as small as possible. The minimum value for these was found to be about 500 ohms and the circuit refused to operate when they were reduced to 200 ohms. In this case, as was expected, a large reduction of screen potential prevented the circuit from operating at all.

This scalar was highly successful and was in fact the best of the three, the maximum operating speed being about 5.2 Mc/sec, with an anode supply of 275 volts, a negative line of -125 volts and the screen grids at 267 volts. At 4.8 Mc/sec, the output was about 10 volts in amplitude, but the wave form tended to become triangular; at 2 Mc/sec. the wave form was good and of about 25 volts amplitude.

The original suggestion for the third scaler came from T.R.E.



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who had used a circuit of this type in the laboratory, for counting short bursts of pulses at 8 Ma/sec. Its chief feature is the use of diodes to clip the wave-forms, as shown in Fig. 51(a). The grid of the non-conducting valve is held just below cut-off by a diode, which also prevents negative overshoot. The diodes on the anodes, catch the anodes at a preset voltage, lower than the full anode swing, and cause the circuit to be fully recovered in a time which is only a fraction of the anode load anode capacity time constant, instead of several times this value. The imput is applied to the cathodes of the anode diodes.

The final form of this scaler is shown in Fig. 51(b). The minimum size of coupling condenser was found to be 40 pF, considerably higher than in the other two scalers. The maximum frequency of operation was about 1.6 Mc/sec. but the output waveform, of 15 volts amplitude, was practically square. The preset diode voltages greatly influenced this shape, and definitely assisted recovery to a high degree. The input pulse required was rather large (20 volts) and the circuit was very sensitive to pulse shape. There is little doubt that this was the reason that a higher operating speed was not attained. Given sharper pulses, a much higher frequency should be countable.

Summarising the results described above, it seems that the main factor in producing a high speed scaler is the choice of a valve with a high mutual conductance capable of supplying a high ourrent, and with fairly low internal capacities, rather than a valve of higher figure of merit (i.e. with very small capacities and a high mutual conductance) which can only supply a small current for charging the coupling condensers. It would be

interesting to compare a scale-of-two, using C.V.138 valves $(g = 9.5 \text{ mA/V}; C_{in} = 9.5 \text{ pF}; C_{out} = 4.5 \text{ pF}; i_a = 10 \text{ mA.}),$ which fall in the latter class, with one using E.L.38 output pentodes (g = 14.3 mA/V; Input capacitance 17.5 pF; outputcapacitance 6.5 pF; max. anode current 200 mA).

NOTE: Since the completion of the work described above, the fastest of the three scales-of-two has been tested at A.E.R.E. The pulse generator used, gave groups of pulses, each of width 0.05 microseconds separated by a time variable from 0 to 10 microseconds, the groups being separated by several milliseconds. The resolving time of 0.2 microseconds was confirmed, but the performance was found to be very dependent on the size of the input pulse; if an amplitude of greater than two or three volts was exceeded, the circuit failed to operate correctly. The reason for this was as follows: the recovery time constant of each pentode anode, consisting of the anode resistance, coupling condenser and stray capacities is very short (of the order of 0.02 microseconds). If an input pulse is applied, large enough to out off the valve initially passing current, then after about 0.06 microseconds, the anodes of both pentodes are at full H.T. potential, leaving both valves in the same condition. When the input pulse disappears, the valve having the longer grid base takes current, and so the circuit always returns to this condition. Thus the scale-of-two ceases to function as such, if the input pulse width is greater than the anode recovery time constant of 0.02 µ sec. and the input pulse amplitude is ten volts or greater.

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APPENDIX III

A Gircuit Incorporating a Chronoscope Recorder, for use with Particle Counters.







FIG.52

This section of the work has already been published, and a reprint is therefore included.

The counting set was originally designed and built for use in training experiments with Geiger-Müller counters, but has also been effective in conjunction with a Type 200A Scaling Unit, allowing higher counting rates to be employed; it has proved very successful in this connection.

When the Unit was used with counters, a separate, stabilized, high-voltage supply was also constructed which was based on a circuit described by Gingrich (1). Nodifications introduced were principally those required for British valves, but a more delicate voltage control was incorporated. The circuit is shown in Fig. 52, and stabilization curves in Fig. 53.

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A Circuit Incorporating a Chronoscope Recorder, for use with Particle Counters

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ABSTRACT. A circuit which makes use of a high-speed mechanical recorder, for counting experiments, is described. As designed it forms a complete low-rate counting set, but is readily adaptable for use after a scaling circuit. The difficulties which arise if two parts of the unit have different resolving times are discussed, and a solution is presented. The resolving time is approximately 1/170 sec.

INTRODUCTION

In experiments in which high counting rates are not in question, it is often more convenient to use a high-speed mechanical recorder instead of the usual scaling circuit. In addition, if a high scaling ratio is required, the number of scaling stages may be reduced by using such a recorder instead of the standard Post Office call meter. The latter is useless if used alone, since its resolving time of 1/10 sec. is such that the loss-rate on the background count of a small Geiger-Müller counter is greater than 1%. Several high-speed mechanical recorders have been described in the literature. One type uses a watch escapement controlled by a solenoid energized

by the pulse; Flammersfeld(1), Tuck(2), and Kipfer(3) have described such instruments, but all require winding. More convenient instruments are the 'Cenco' counter and those described by Lewis(4) and Neher(5). Neher's instrument is probably the fastest mechanical recorder so far developed, having a resolving time of 0.8×10^{-3} sec. The Marconi Instrument Company have recently produced a recorder for chronoscope work, which has a resolving time of the order of 1/200 sec. A circuit incorporating this instrument has been developed by the writer and is described below.

CONSTRUCTION AND OPERATION OF RECORDER

A small armature pivoted centrally carries a pallet engaging in two gear wheels which are meshed together. The recording pointer is carried on the same shaft as one of the gear wheels. Two solenoids are situated one on each side of the armature, and if these are energized alternately by the operating pulses, the gearing is such that the pointer advances one division for each pulse. The pointer shaft also carries a small wheel which is part of the resetting device. Two insulated brushes bear on this wheel which is conducting except for a small gap which breaks the circuit when the pointer reaches zero.

In operating the recorder, the first problem is the direction of a single pulse train, derived from a Geiger-Müller counter, so that pulses pass alternately into the two solenoids. The obvious choice for this is a trigger circuit, but this leads to difficulties when counting random pulses if the resolving times of the trigger circuit and the recorder are different. Consider the case in which the resolving time of the recorder



The recorder circuit

 $\begin{array}{l} R_1, 2 \ \mathrm{M}\Omega; \ R_2, \ R_{13}, \ R_{14}, \ 100,000 \ \Omega; \ R_3, \ 10,000 \ \Omega; \ R_4, \ R_8, \ 20,000 \ \Omega; \ R_5, \ 1200 \ \Omega; \ R_6, \ R_{23}, \ R_{24}, \ 500,000 \ \Omega; \ R_7, \ R_{12}, \ R_{16}, \ R_{17}, \ R_{17}, \ R_{17}, \ R_{27}, \ 250,000 \ \Omega; \ R_9, \ 30,000 \ \Omega; \ R_{10}, \ 15 \ \mathrm{M}\Omega; \ R_{11}, \ 600,000 \ \Omega; \ R_{10}, \ R_{22}, \ R_{25}, \ 1000 \ \Omega; \ R_{20}, \ 60,000 \ \Omega; \ R_{21}, \ 5000 \ \Omega; \ R_{26}, \ 75,000 \ \Omega; \ R_{28}, \ 1000 \ \Omega; \ R_{2}, \ 50,000 \ \Omega; \ R_{21}, \ 5000 \ \Omega; \ R_{26}, \ 75,000 \ \Omega; \ R_{28}, \ 1000 \ \Omega; \ R_{20}, \ 60,000 \ \Omega; \ R_{21}, \ 5000 \ \Omega; \ R_{26}, \ 75,000 \ \Omega; \ R_{28}, \ 1000 \ \Omega; \ R_{20}, \ 60,000 \ \Omega; \ R_{21}, \ 5000 \ \Omega; \ R_{26}, \ 75,000 \ \Omega; \ R_{28}, \ 1000 \ \Omega; \ R_{20}, \ 60,000 \ \Omega; \ R_{21}, \ 5000 \ \Omega; \ R_{26}, \ 75,000 \ \Omega; \ R_{28}, \ 1000 \ \Omega; \ R_{20}, \ 60,000 \ \Omega; \ R_{21}, \ 5000 \ \Omega; \ R_{26}, \ 75,000 \ \Omega; \ R_{26}, \ 85,000 \ \Omega; \ R_{26}, \ R_{$

is the longer. An incoming pulse causes the trigger circuit to take up a state A, in which one side is conducting and the armature of the recorder takes up its corresponding A state. If a second pulse now enters the circuit after a time interval less than the resolving time of the recorder but greater than that of the trigger circuit, the latter changes to its other state B, but the recorder has not recovered and therefore remains in state A, missing the pulse. The next pulse entering the system returns the trigger circuit to state A, but the recorder is already in this state and this pulse also is unrecorded. The next pulse operates the whole system correctly. Thus, having missed one pulse which has arrived too soon after the preceding pulse, the recorder also misses the next one irrespective of the time interval between it and the previous pulse.

The prevention of such double losses may be achieved in one of two ways: either the trigger circuit must have exactly the same resolving time as the recorder, or the intervals between the input pulses must never be shorter than the resolving time of the recorder. In the circuit described, the latter method was used, since it was found that the pulse shape from the trigger circuit was such that it would not operate the recorder when the resolving times of the trigger circuit and the recorder were the same. The short time intervals were eliminated by a flip-flop circuit, the time constant of which was adjusted to be the same as the resolving time of the recorder.

DESCRIPTION OF CIRCUIT

The input stage is a negative-sensitive cathode-coupled flipflop, the dead time of which is adjusted by means of R_6 to be the same as the resolving time of the recorder. Grid voltages are rather critical and are preset by means of R_8 . The diode V_2 and the condenser C_3 speed up the recovery of the circuit and make the dead time more sharply defined. The input condenser C_1 should be a mica dielectric type with a highworking voltage since one side is connected directly to the counter anode. The stage can be triggered by a negative pulse of amplitude 1 V. and delivers an output of about 12 V. If the input is less than 1 V., the dead time of the stage increases, since the condenser C_2 must discharge more fully before triggering can take place.

The second stage, which switches the pulses alternately into two channels, is the Eccles-Jordan circuit in its negativesensitive form (6,7). The coupling condensers C_7 , C_8 are relatively large to provide a pulse of sufficient length to operate the following stage; in addition, voltage adjustments are very critical if these condensers are small. Component values are not otherwise critical, but pairs R_{15} , R_{16} and R_{17} , R_{18} must be matched at least to 5 %. Voltage supplies on the original unit were derived from a single positive supply, the cathode potential being adjusted by means of R_{21} . An improvement, however, would be the provision of a separate negative supply for the suppressor grids, the cathodes then being earthed. In this stage also, in the interests of standardizing the valves, a double-triode trigger circuit, fed through a double-diode, could be used. The third stage is a power amplifier feeding the recorder. It might be thought that the position of the count switch S_2 in this stage would give rise to spurious counts, but this was not found to be so.

The recorder dial is graduated to 50, so some method of counting whole turns must be introduced. The reset terminals on the recorder are connected across a small 2000Ω highspeed relay L_3 which operates on less than 10 mA., the maximum current which the reset contacts will carry. When the recorder pointer is at zero, the relay is energized, discharging a 4μ F. paper condenser C_{15} through a Post Office call meter L_4 ; when the relay is 'off', the condenser recharges from the h.t. supply through a high resistance R_{27} . The spark suppressing combination R_{28} , C_{16} is essential to avoid sticking at the relay contacts. This method of counting whole turns may be rather complicated but it has two definite advantages-there is at no time a sudden drain of current from the h.t. supply and it prevents a continuous current from passing through the Post Office call meter, even if the recorder pointer stops at zero. A simple flashing neon pulse generator is included for testing and resetting the recorder to zero. Two pulse repetition frequencies are provided, one of about 1 per sec. and the other of about 10 per sec.

The resolving time of the unit was determined by means of a variable frequency pulse generator. This method was used since the time required for accurate determinations by means of random pulses was prohibitively long, due to the low counting rates which had to be employed. Experiments were made, however, with a Geiger-Müller counter and two sources (8), the results of which agreed with the other determination, within the limits of statistical error.

The maximum 'safe' speed of operation of the recorder, on regular pulses, is 170 per sec. Any further increase in speed depends entirely on very careful adjustments of spring contacts in the recorder. It has been possible to push the speed to 190 per sec., but the adjustment was not maintained for any length of time.

Finally, the author wishes to express his sincere thanks to Prof. N. Feather for his constant interest and encouragement throughout the course of this work.

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