IX. *The α Particles emitted by the Active Deposits of Thorium and Actinium.*

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RECEIVED NOVEMBER 6, 1911. READ NOVEMBER 24, 1911.

THE present Paper deals mainly with the application of probability to the emission of α particles from thorium and actinium active deposits. The most important result of the investigation will, however, be found in the latter part of the Paper, where it is shown that the numbers of α particles emitted by thorium C_1 and thorium C_2, when the active deposit is in equilibrium, are not equal, thus proving that these products are not directly genetically connected. The nomenclature of the various products proposed by Rutherford and Geiger ("Phil. Mag.," XXII., p. 621, 1911) will be adopted throughout. The Paper may conveniently be divided as follows :—

1. Discussion of probability laws as applied to the emission of α particles.
2. Experiments with actinium active deposit.
3. Experiments with thorium active deposit.

1. In a previous Paper* we showed that if α particles are counted on a zinc sulphide screen at a mean rate of μ per second, then the probability of occurrence of a time interval of length between t and $t+\delta t$ is $\mu e^{-\mu t}\delta t$. This law of intervals may be employed in all experiments in which α particles are counted to give some indication as to whether the results are reliable. Thus, when counting α particles of short range by the scintillation method, the scintillations are so faint that with an untrained observer they are often missed, and generally in such a way that the eye fails to recognise, not merely a single scintillation here and there, but frequently a whole group of successive scintillations. An analysis of the intervals between successive records of scintillations would reveal such errors and give some idea of the correction necessary. Again, as has been pointed out by Mme. Curie,† in counting α particles by the electrical method, the damping of the electrometer needle,

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* Phys. Soc. "Proc.," XXIII., V., p. 367, 1911.

† "Le Radium," VIII., p. 354, 1911.

and the effect of the Bronson resistance, is generally such that two α particles coming within a small interval would scarcely produce separate effects, and from an examination of the numbers given by Rutherford and Geiger* the magnitude of the throws does not always give reliable indication as to whether one or two α particles have entered the ionisation chamber. In the experiments of Rutherford and Geiger the rate at which the α particles were counted was very small (3·5 per minute), and, assuming that intervals of $\frac{1}{4}$ second would not be recognised, the correction would be 1·4 per cent. The shape of the curve of intervals given in their Paper, in which there are too few small intervals, is due to this cause.

The formula given above is derived initially from the assumption that the probability of a scintillation occurring in any small time interval, δt, is $\mu\delta t$, and it is, therefore, analogous to the derivation of the exponential law of decay in radio-active disintegration, the only difference being the exact meaning of μ in the two cases. If, then, we have in our source of α particles a product present of very short life, it will reveal itself by there being too many short intervals, and thus, by observing the intervals between successive α particles, we can hope to determine the presence of such products. Thus, by the experiments of Geiger and Marsden,† a product was discovered after thorium emanation of mean life $\frac{1}{5}$ second, while our own experiments‡ on uranium and polonium showed that no short life products were present in these substances.

2. *Experiments with Actinium Active Deposit.*

The active deposit of actinium is known to consist of three successive products—actinium B (β), C (α) and D (β). The double-screen experiments of Geiger and Marsden,§ however, indicated certain irregularities in the emission of the α particles, in that 10 per cent. of them seemed to be emitted in pairs. Irregularities indicating the presence of a product of short life (not act. A) were also obtained in the experiments of Mlle. Blanquies.‖ On the other hand, Moseley and Fajans,¶ by a special method, failed to recognise any such product. The

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* Roy. Soc. "Proc.," A, LXXXI., p. 141, 1908.
† "Phys. Zeitschr.," XII., p. 7, 1910.
‡ *Loc. cit.*
§ *Loc. cit.*
‖ "Le Radium." VI., p. 230, 1909.
¶ "Phil. Mag.," XXII., p. 629, October, 1911.

question as to the existence of such a product is very interesting, for it would indicate a lateral disintegration, such as Fajans* found in radium C and Antonoff† in uranium. We have, therefore, repeated the experiments of Geiger and Marsden to find out whether the large number of connected particles is explained by probability, which we have shown gives what at first sight appears to be an unexpectedly large number of small intervals. The apparatus used is the same as was employed in the above experiments, and for convenience is drawn in Fig. 1.

Two microscopes, M_1 and M_2, are focussed on exactly opposite areas of two zinc sulphide screens, S_2 and S_1. Aluminium foils are placed between the screens in such a way as to allow emanation from the actinium source A to diffuse between them, at the same time preventing the screen S_2, say, being visible through the microscope M_2. The emanation source A was placed in position and left there for 36 minutes, so that the active deposit between S_1 and S_2 attained half its equilibrium value. The source was then removed, and, after allowing about 25 seconds for the emanation to decay, scintillations were observed and recorded by both observers on a travelling chronograph tape until the expiration of five minutes. The source A was then replaced for five minutes, and the observations repeated until a sufficient number of scintillations had been recorded. The results of several experiments are given in Table I., the sums of all the observations being placed in the last column. The calculated values given are obtained from the formula

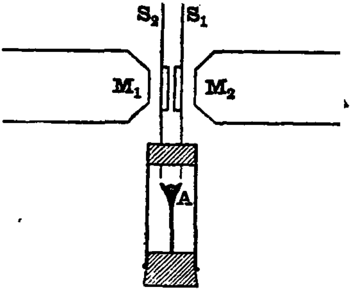

FIG. 1.

$$N\mu\int_{t_1}^{t_2} e^{-\mu t}\,dt = N(e^{-\mu t_1} - e^{-\mu t_2}).$$

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* "Phys. Zeitschr.," XII., p. 369, 1911.

† "Phil. Mag.," XXII., p. 419, 1911.

TABLE I.

Duration of interval between successive scintillations. Seconds.	Number of Intervals.					
	Experiment V.		Experiment IX.		Totals of all experiments.	
	Observed.	Calculated.	Observed.	Calculated.	Observed.	Calculated.
0·0 to 0·5	51	49·3	9	7·6	219	207·3
0·5 to 1·0	46	44·2	7	7·5	200	189·6
1·0 to 2·0	77	76·8	17	14·3	340	332·4
2·0 to 3·0	74	62·5	19	13·4	285	278·0
3·0 to 4·0	43	51·4	15	12·7	220	232·9
4·0 to 5·0	37	41·6	8	11·9	184	193·9
5·0 to 7·0	59	62·4	23	21·7	290	301·7
7·0 to 10·0	44	56·8	24	27·9	279	296·5
10·0 to 15·0	61	43·7	29	36·5	263	262·4
15·0 to 20·0	14	16·1	26	26·8	128	122·4
20·0 to 30·0	6	7·8	32	34·3	97	93·4
30 to infinity	2	1·2	46	40·4	70	64·5
$\mu =$	0·201		0·061		Average. 0·149	

The agreement between the observed and calculated values is on the whole good, though for intervals up to three seconds the observed numbers are slightly in excess.[*] If this effect is real, we have no certain explanation for it, but the excess only corresponds to about 1 per cent. of the whole number of scintillations, indicating that if there is a product present of period about one second it must be a lateral one, and consist of only about 1 per cent. of the main line of descent. It is of interest to note that, assuming a period of one second for such a hypothetical product, its range, according to the empirical formula given by Geiger and Nuttall,[†] would be only 4 mm. in excess of that of actinium C., and in such circumstances it would be very difficult to detect. Experiments are, however, in progress on this point.

We, therefore, conclude that the 10 per cent. of connected particles found in the experiments of Geiger and Marsden, referred to above, are due to probability, and that no certain evidence of the existence of a short life product can be drawn from them.

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[*] The question as to the probability difference between the observed and calculated values will be examined in a later Paper.

[†] "Phil. Mag.," XXII., p. 613, October, 1911.

3. *Experiments with Thorium Active Deposit.*

In the thorium family, after the emanation, there are known to be three α-ray products. The first, thorium A, was discovered by the double-screen experiments of Geiger and Marsden, and has a period of only 0·14 second, so that it practically accompanies the emanation. The other α-ray products have been investigated chiefly by Hahn, who was, however, unable to separate them by recoil or by chemical methods. They are only recognised by their different ranges, for which Hahn found the values 5·0 cm. and 8·6 cm. Hahn explained his results by assuming that the products are genetically derived from one another, and that the second has an extremely short period.

Geiger and Marsden, in some preliminary experiments,[*] found that the number of pairs of scintillations, using a double-screen arrangement, is smaller and of a different order to the number obtained with thorium and actinium emanations: Previous results had also shown that thorium emanation +A give twice as many α particles as the active deposit. From these results Rutherford and Geiger[†] have suggested that the changes occurring in thorium active deposit are irregular, and have proposed the following scheme of disintegration :—

$$\begin{array}{llllll} & & & & \underline{C_1+C_2} & \\ & & & & \quad\| & \\ \text{Product:} & \text{Th. Em.}\longrightarrow & \text{Th. A}\longrightarrow & \text{Th. B}\longrightarrow & \text{Th. C}\dashrightarrow & \text{Th. D} \\ \text{Radiation:} & \alpha & \alpha & \beta & \alpha(5{\cdot}0 \text{ and } 8{\cdot}6) & \beta \end{array}$$

Assuming that for each α particle given out by thorium emanation and A, thorium C_1 and C_2 give one particle between them, this scheme explains all the above observed facts. However, Miss Leslie,[‡] by comparing the numbers of α particles from radio-thorium and thorium emanation has come to the conclusion that the latter gives (+A) four α particles, in which case no irregularities of the nature of lateral disintegration need be assumed. To throw light on this interesting question we have, therefore, made double-screen experiments on thorium active deposit, and have also, by counting the number of α particles at different distances from a source of

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[*] See note Rutherford and Geiger, "Phil. Mag.," XXII., p. 621, October 1911.

[†] Rutherford and Geiger, *loc. cit.*

[‡] "Le Radium," VIII., p. 356, 1911.

thorium active deposit, determined the relative number of α particles emitted by thorium C_1 and C_2. The apparatus used in this latter experiment is shown in Fig. 2.

A plate, A, was made active by exposure to the emanation from a preparation of mesothorium,* and placed at a distance greater than 8·6 cm. from the zinc sulphide screen S sealed on to the end of a wide tube. Scintillations were counted on the screen for different pressures of the air in the tube, and thus a curve was drawn showing the number of α particles corresponding to different "ranges" from the source, the latter being obtained by multiplying the distance from S to A by the number of centimetres of pressure in the tube and dividing by 76, correction being also made for the temperature to reduce the observations to 15°C. The results of a particular experiment are shown in Fig. 3.

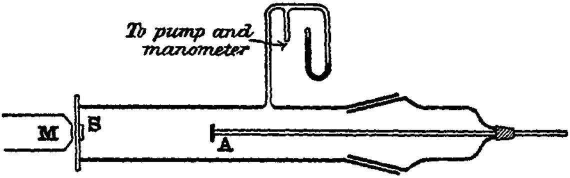

FIG. 2.

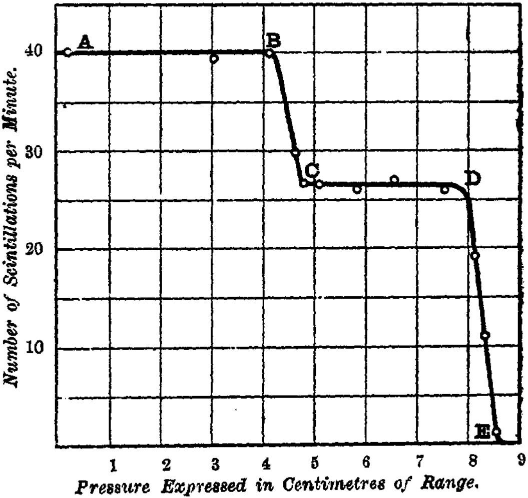

FIG. 3.

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* We are deeply indebted to Mr. F. H. Glew, who very kindly loaned us this preparation.

In the figure the part AB corresponds to the α particles from both thorium C_1 and C_2. The part BC shows that the α particles from thorium C_1 are gradually being reduced in number, C denoting the end of the range. The part CDE corresponds to the α particles from thorium C_2 only. The curve is similar to those given by Geiger* for actinium, &c. It will be noticed (1) The ranges of thorium C_1 and C_2 are 4·8 cm. and 8·6 cm. respectively; (2) the ordinates of the part of the curve CD are greater than half the ordinates corresponding to AB, thus showing that thorium C_2 gives more than half as many α particles as thorium C_1+thorium C_2. The weighted mean of 12 different experiments in which the ratio found varied between 0·60 and 0·70 is

$$\frac{\text{No. of } \alpha \text{ particles from thorium } C_2}{\text{No. from thorium } C_1 + \text{No. from } C_2} = 0{\cdot}646.$$

That is to say, thorium C_2 gives 1·83 times as many α particles as thorium C_1.

This ratio is not due to an admixture of radium C with the thorium active deposit, as might, perhaps, be expected, since mesothorium preparations contain generally a large percentage of radium. Observations of the ratio were made in several cases for 30 hours after the active plate was taken from the mesothorium, and although the active deposit decayed accurately with a period of 10·6 hours, the ratio was unaltered, showing that the decay of both thorium C_1 and thorium C_2 is governed by that of thorium B. Again, with a short exposure, the ratio was within the experimental error independent of the rise in activity of the active deposit.

We will withhold a full discussion of the deductions to be derived from the above experiments until the figures referring to the double-screen experiments have been considered. These experiments were carried out in the same way as those with actinium, except that mesothorium was used instead of the actinium preparation, while in some cases the active deposit was first deposited on an aluminium foil which was subsequently placed between two other aluminium foils between the two screens. In the experiments countings were generally made for two hours, the decay of the active deposit in this period being such that the calculated values of the various intervals are influenced to something less than 1 per cent. by

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* "Phil. Mag.," XXII., p. 201, 1911.

the fact that the source was not constant, and consequently the value of μ variable. The results are given in Table II., where the columns have the same significance as in Table I. The last two columns refer to an experiment in which the active source was thorium emanation plus about 20 per cent. of its equilibrium amount of active deposit. The calculated values in this case are obtained from the above formula, with the additional assumption that 59 per cent. (found by trial) of the scintillations are such that half of them are produced by α particles from emanation atoms, and the other half by the thorium A produced by the disintegration of these emanation atoms, the period of thorium A being taken as 0·145 second*. This experiment gives an idea of the efficiency of the screens, as the total possible number would be about 90 per cent.

TABLE II.

Duration of interval between successive scintillations.	Number of Intervals.							
	Experiment VII. Active deposit obtained by diffusion of emanation.		Experiment VIII. Active deposit on separate aluminium foil.		Totals. Active deposit.		Thorium emanation.	
Seconds.	Observed.	Calculated.	Observed.	Calculated.	Observed.	Calculated.	Observed.	Calculated.
0·0 to 0·5	91	68·5	101	109·1	353	314·2	438	435·0
0·5 to 1·0	55	60·3	98	93·5	311	278·9	178	179·0
1·0 to 2·0	92	99·0	159	149·0	438	466·2	212	203·2
2·0 to 3·0	63	76·5	114	109·5	354	362·8	96	107·5
3·0 to 4·0	59	59·0	74	80·4	250	289·4	55	56·6
4·0 to 5·0	44	45·5	48	59·4	214	228·3	27	30·2
5·0 to 7·0	57	61·7	75	75·8	323	327·6	26	24·7
7·0 to 10·0	57	49·1	59	53·6	295	290·1	12	8·3
10·0 to 15·0	26	30·3	32	28·0	226	225·3	1	1·5
15·0 to 20·0	15	8·2	4	6·0	102	93·4		
20·0 to 30·0	2	2·8	2	1·6	76	64·6		
30·0 to infinity	0	0·2	0	0·1	24	25·1		
$\mu=$	0·2606		0·3069		Average. 0·196		...	

The observed and calculated values are, on the whole, in good agreement, except that there is a small excess of intervals between 0 and 1 second, corresponding to about 2 per cent. of the whole number of scintillations. This may be partly or wholly due to probability, and this latter question will be

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* Moseley and Fajans, "Phil. Mag.," XXII., p. 629, October, 1911.

examined in a later theoretical Paper. However, the excess is only shown in the experiments in which the active deposit was obtained by diffusion of the emanation between the two screens, and is not pronounced in the experiments in which the active deposit was first obtained on a separate aluminium foil. Again, the excess of small intervals is not noticeable in the analysed observations of each separate observer. An explanation may be that the emanation diffuses through unavoidable small holes in the aluminium foils, causing the active deposit to be deposited directly on the zinc sulphide screens themselves, the pairs of scintillations being the result of either (1) a single α particle producing a scintillation on both screens, or (2) the recoil atom or δ-rays produced on the explosion of an α particle. In the former case we should expect the whole of the excess of intervals to be from 0 to 0·5 second, while in regard to the latter the experiments of one of us* have shown that the luminescent effect of β-rays is not altogether simultaneous with their impact on the screen. Diffused patches, or " sploshes " of light, were occasionally seen, and sometimes the observer might tap these as scintillations without being able to decide at the exact moment as to whether or not they are real scintillations. A special experiment, in which the whole of the active deposit was on one of the zinc sulphide screens, gave 255 scintillations on the active screen and 144 on the other, with an excess of 4 per cent. of intervals 0 to 1 second, the difference in the numbers being largely explained by the uneven surface of the crystals on the screen.

It is also improbable that the excess of small intervals mentioned above is due to the missing of scintillations by either observer. Evidence on this question was obtained as follows: The tapes were examined with regard to the number of times either observer recorded 1, 2, 3, &c., scintillations without the sequence being interrupted by the other observer recording a scintillation. The total numbers so obtained are given in Table III., and it will be seen that they are in excellent agreement with the numbers calculated from simple probability—*i.e.*, the probability of 1's$=\frac{1}{2}$, 2's$=\frac{1}{4}$, &c. These results are also interesting from the point of view of the question as to the application of probability to radio-active disintegration.

Independently of the possible small excess of small intervals the above double-screen experiments show conclusively that

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* E. Marsden, " Proc." Roy. Soc., A 83, p. 548, 1910.

if thorium C_2 comes directly after thorium C_1, its period must be greater than a few seconds. In that case, however, Hahn would have been able to separate the products by recoil experiments, while similar experiments by Geiger and one of us, in which an α-ray recoil product of a fraction of a second period would have been detected, failed to reveal any. Thus we may conclude that thorium C_2 is not produced by direct disintegration of thorium C_1, and the above experiments on the relative number of α particles from these products show that they must be on different lines of descent, otherwise the numbers would be equal. It is, however, just about within the limit of experimental error that the ratio C_2/C_1+C_2 above should be 0·667. In that case we should have thorium C_2 with twice as many α particles as thorium C_1. To explain this without bringing in the idea of lateral disintegration we should have to assume two products of range 8·6 cm., and also that thorium emanation (+A) gives 6 α particles. These assumptions seem somewhat out of the question.

TABLE III.—*Analysis of Scintillations with respect to the number of times either observer records a sequence of Scintillations uninterrupted by the other observer recording a Scintillation.*

—	—	Number of Scintillations in Sequence.								
		1.	2.	3.	4.	5.	6.	7.	8.	9 and over.
Records of actinium active deposit	Observed	533	297	125	63	23	9	6	5	1
	Calculated	531	266	133	66	33	16	8	4	4
Records of thorium active deposit	Observed	836	423	215	109	57	26	13	7	9
	Calculated	848	424	212	106	53	26	13	7	7
Totals	Observed	1,369	720	340	172	80	35	19	12	10
	Calculated	1,379	690	345	172	86	43	21	11	11

We have tried another method to throw light on the question. Thorium X was obtained on a plate of 3 cm. diameter by recoil from radio-thorium, and an attempt was made by sucking air over it to determine the ratio of the number of α particles emitted from thorium emanation to the number from thorium X. The results were, unfortunately, not decisive, for assuming one α particle per disintegrating atom of thorium X, it was calculated that the film retained more than 25 per cent. of its emanation, even when a strong current of air was passed over it continuously.

If the logarithms of the transformation constants be plotted against the logarithms of the ranges according to the method of Geiger and Nuttall,* we find that the product of 4·8 cm. range corresponds to the 60 minutes' period, previously known as that of thorium C; while the period of a product of 8·6 cm. range should be of the order 10^{-12} second.

However, as noted above, with a short exposure of a plate to thorium emanation the rise in activity with time of both the products is practically the same. If thorium C_2 has a short life and comes directly after thorium B such a plate would not rise in activity with time, but would decay along with the thorium B to half value in 10·6 hours. Thus thorium C_2 must have a parent of period about the same as that of thorium C_1 (60 mins.) It therefore appears that the lateral disintegration begins with the atoms of thorium C_1, 35 per cent. giving rise to α particles of 4·8 cm. range, and the remaining 65 per cent. giving rise to α particles of 8·6 cm. range with probably an intermediate emission of slow β particles.

The experiments of Geiger and Kovarik† are interesting in regard to the above questions. Assuming that, with the active deposit in equilibrium, thorium C_1+C_2 give two α particles per disintegrating atom of thorium D, they found results which suggest that the latter product gives two β particles per disintegrating atom. If we take our above result that thorium C_1+C_2 give only one α particle between them, we need only assume one β particle from thorium D, provided we also assume a scheme of disintegration somewhat as follows :—

$$\begin{tikzcd}
 & & & \underset{\alpha\ (4\cdot8)}{\text{Th. C}_1} \arrow[drr] & & \\
\underset{\alpha}{\text{Th. Em.}} \arrow[r] & \underset{\alpha}{\text{Th. A}} \arrow[r] & \underset{\beta}{\text{Th. B}} \arrow[ur] \arrow[dr] & & & \underset{\beta}{\text{Th. D.}} \\
 & & & \underset{?}{\text{Th. C}_1'} \arrow[r] & \underset{\alpha\ (8\cdot6)}{\text{Th C}_2} \arrow[ur] &
\end{tikzcd}$$

However, the various assumptions involved are such that this latter question cannot be taken as settled, and further consideration must be delayed until more experimental evidence is available.

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* "Phil. Mag.," XXII., p. 613, October, 1911.

† "Phil." Mag., XXII., p. 604, October, 1910.

Summary.

1. A discussion is given of the magnitude and nature of an error which sometimes affects experiments on the counting of α particles.

2. No irregularities in the emission of α particles from actinium active deposit are shown by the experiments.

3. The range of thorium C_1 has been determined and found to be 4·8 cm.

4. The experiments show that there is a lateral disintegration in thorium active deposit, thorium C_1 and thorium C_2, being on different branches in the ratio 1 : 1·83.

This research was carried out at the East London College, and we are much indebted to the Research Grant Committee of that institution and to Prof. Lees for placing the resources of the laboratory at our disposal.

ABSTRACT.

In a previous Paper ("Proc." Phys. Soc., Aug., 1911) the authors showed that if α-particles are counted on a zinc sulphide screen at a mean rate of μ per second, then the probability of occurrence of a time interval, of length between t and $t+\delta t$, is $\mu e^{-\mu t}$. This formula may be applied, to test whether two α-particles are given off simultaneously from a disintegrating atom or whether in any source of α-particles there exist two successive α-ray products, the latter being of short life.

In the previous Paper Uranium and Polonium were shown not to give such irregularities, and in the present Paper the same result has been found for Actinium and Thorium active deposits, although experiments of various investigators pointed to the probability of positive results.

The experiments further suggest a lateral disintegration in Thorium active deposit, and this is proved to be the case by results which show that the two α-ray products in Thor. Act. Dep. (Th. C_1 and C_2) do not give an equal number of α-particles when the active deposit is in equilibrium, which is required by the ordinary disintegration theory. Thus it is concluded that of the atoms Th. C, 35 per cent. give rise to α-particles of 4·8 cm. range and 65 per cent. to α-particles of 8·5 cm. range, with probably the intermediate emission of β-particles. Various cognate questions are also discussed in the Paper.