An Electrical Method of Counting the Number of α-Particles from Radio-active Substances.

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The total number of α-particles expelled per second from 1 gramme of radium has been estimated by Rutherford* by measuring the charge carried by the α-particles expelled from a known quantity of radium in the form of a thin film. On the assumption that each α-particle carries the ionic charge $e = 3{\cdot}4 \times 10^{-10}$ electrostatic unit, it was shown that $6{\cdot}2 \times 10^{10}$ α-particles are expelled per second from 1 gramme of radium itself, and four times this number when in radio-active equilibrium with its three α-ray products, viz., the emanation, radium A and C. In order to reconcile the value of e/m found for the α-particle with that to be expected for the helium atom, it was later† pointed out that the α-particle should carry a charge equal to $2e$. On this assumption, the number of α-particles expelled per second per gramme of radium is reduced to one-half the first estimate.

The need of a method of counting the α-particles directly without any assumption of the charge carried by each has long been felt, in order to determine the magnitude of the various radio-active quantities with a minimum amount of assumption. If the number of α-particles expelled from a definite quantity of radio-active matter could be determined by a direct method, the charge carried by each particle could be at once known by measuring the total positive charge carried by the α-particles. In this way, it should be possible to throw some light on the question whether the α-particle carries a charge e or $2e$, and thus settle the most pressing problem in radio-activity, viz., whether the α-particle is an atom of helium.

In considering a possible method of counting the number of α-particles, their well-known property of producing scintillations in a preparation of phosphorescent zinc sulphide at once suggests itself. With the aid of a microscope, it is not very difficult to count the number of scintillations appearing per second on a screen of known area when exposed to a source of α-rays. The doubt, however, at once arises whether every α-particle produces a scintillation, for it is difficult to be certain that the zinc sulphide is homogeneous throughout. No confidence can be placed in such a method of

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† Rutherford, 'Phil. Mag.,' October, 1906.

counting the total number of α-particles (except as a minimum estimate), until it can be shown that the number so obtained is in agreement with that determined by some other independent method which does not involve such obvious uncertainties. The results of some observations on the number of scintillations produced by the α-particles from radium will be discussed later.

It has been recognised for several years that it should be possible by refined methods to detect a single α-particle by measuring the ionisation it produces in its path. On the assumption that an α-particle carries an ionic charge e, one of us has shown that the α-particle expelled from radium itself produces 86,000 ions in its path in air before it is stopped. Taking the charge of an α-particle as $2e$, this number is reduced to one-half. Consequently if the α-particle passes through air in a strong electric field, the total quantity of electricity transferred to the electrodes is $43{,}000e$. Taking $e = 3 \cdot 4 \times 10^{-10}$ E.S. unit, this corresponds to $1 \cdot 46 \times 10^{-5}$ E.S. unit. For the purpose of illustration, suppose that a Dolezalek electrometer of capacity 50 E.S. units, which has a sensibility of 10,000 mm. divisions per volt between the quadrants, is used for detection of the ionisation. The quantity $1 \cdot 46 \times 10^{-5}$ unit transferred to the electrometer system would cause a deflection of the needle of 0·3 mm. This is small but detectable. In a similar way, if an electroscope of capacity 2 E.S. units be employed instead of an electrometer, the movement of the leaf would correspond to a difference of potential of $2 \cdot 1 \times 10^{-3}$ volt. While there is no inherent impossibility in detecting such small quantities of electricity by either the electroscope or electrometer, yet the measurement would have to be of a refined character in order to get rid completely of all extraneous sources of disturbance. One difficulty is that the moving system in very sensitive electrometers or electroscopes has a long period of swing and consequently moves very tardily when a small difference of potential is suddenly applied. Some preliminary experiments to detect a single α-particle by its direct ionisation were made by us, using specially constructed sensitive electroscopes. As far as our experience has gone, the development of a certain and satisfactory method of counting the α-particles by their small direct electrical effect is beset with numerous difficulties.

We then had recourse to a method of automatically magnifying the electrical effect due to a single α-particle. For this purpose we employed the principle of production of fresh ions by collision. In a series of papers, Townsend* has worked out the conditions under which ions can be produced by collisions with the neutral gas molecules in a strong electric field. The

* 'Phil. Mag.,' February, 1901; June, 1902; April, 1903; September and November, 1903.

effect is best shown in gases at a pressure of several millimetres of mercury. Suppose that the current between two parallel plates immersed in a gas at low pressure is observed when the air is ionised by X-rays. The current through the gas for small voltages at first increases with the field and then reaches a saturation value, as is ordinarily observed in ionised gases at atmospheric pressure. When the field is increased beyond a certain value, however, the current rises rapidly. Townsend has shown that this effect is due to the production of fresh ions in the gas by the collision of the negative ions with the gas molecules. At a later stage, when the electric field approaches the value required to cause a spark, the positive ions also become effective as ionisers but to a much smaller degree than the negative. Under such conditions, the small current through the gas due to the external ionising agency may be easily increased several hundred times. The magnification of the current depends upon the voltage applied and becomes very large just below the sparking value.

In our experiments to detect a single α-particle, it was arranged that the α-particles could be fired through a gas at low pressure exposed to an electric field somewhat below the sparking value. In this way, the small ionisation produced by one α-particle in passing along the gas could be magnified several thousand times. The sudden current through the gas due to the entrance of an α-particle in the testing vessel was thus increased sufficiently to give an easily measurable movement of the needle of an ordinary electrometer.

Experimental Arrangement.—Before considering the various difficulties that arose in the course of the investigations, a brief description will be given of the method finally adopted. The experimental arrangement is shown in fig. 1. The detecting vessel consisted of a brass cylinder A, from 15 to

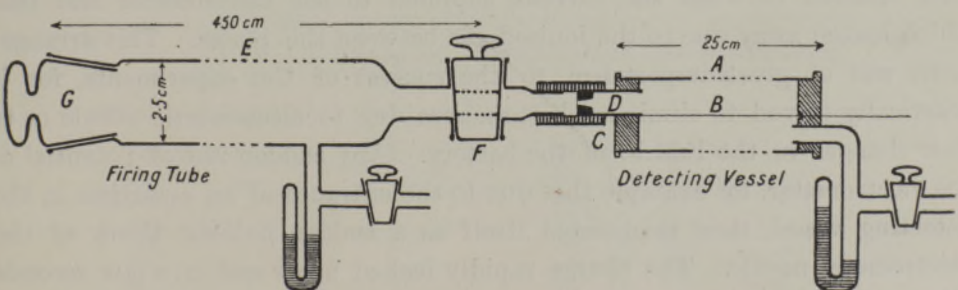

Fig. 1.

25 cm. in length, 1·7 cm. internal diameter, with a central insulated wire B passing through ebonite corks at the ends. The wire B was in most experiments of diameter 0·45 mm. The cylinder, with a pressure gauge attached,

was exhausted to a pressure of from 2 to 5 cm. of mercury. The central wire was connected with one pair of quadrants of a Dolezalek electrometer and the outside tube to the *negative** terminal of a large battery of small accumulators, the other pole of which was earthed. In the ebonite cork C was fixed a short glass tube D of internal diameter 5 mm., in the end of which was a circular opening of about 1·5 mm. diameter. This opening, through which the α-particles entered the testing vessel, was covered with a thin sheet of mica tightly waxed over the end of the tube. In most experiments the thickness of mica was equivalent, as regards stopping power of the α-particle, to about 5 mm. of air at atmospheric pressure. Over the tube D was fixed a wide rubber tube, to the other end of which was attached a long glass tube E of 450 cm. in length and 2·5 cm. diameter. A large stop-cock F with an opening 1 cm. in diameter was attached to the end of the glass tube next to the detecting vessel. The other end of the long glass tube was closed by a ground stopper G.

The general procedure of an experiment was as follows. The voltage applied to the testing vessel was adjusted so that the ionisation in the vessel due to an external source of γ-rays was increased by collision several thousand times. The radium tube which served as a source of γ-rays was then removed. Under ordinary conditions, when all external sources of ionisation were absent, there was always a small current passing through the gas. In order to avoid the steady movement of the electrometer needle due to this cause, the current was allowed to leak away through a radio-active resistance attached to the electrometer system. This consisted of two insulated parallel plates, the upper connected with the electrometer and the lower with earth. A layer of radio-active material was placed on the lower plate. As the potential of the electrometer needle rose, equilibrium was soon reached between the current supplied to the electrometer and that which leaked away due to the ionised gas between the plates. This arrangement was of great importance to the success of the experiments, for it practically served to eliminate disturbances due to electrostatic effects or to slow changes in the E.M.F. of the battery. Any sudden rise of potential ot the electrometer, for example that due to the entrance of an α-particle in the detecting vessel, then manifested itself as a sudden *ballistic* throw of the electrometer needle. The charge rapidly leaked away and in a few seconds the needle was again at rest in its old position.

The active matter, in the form of a thin film of not more than 1 square cm.

* If the tube were connected to the positive pole of the battery, the magnification by collision only became appreciable near the sparking voltage. With the negative pole, the magnification increased more gradually and was far more under control.

in area, was fixed in one end of a hollow soft iron cylinder which could be moved along the glass tube from the outside by means of an electro-magnet The glass tube was then exhausted by means of a Fleuss pump and, if required, to a still lower pressure by means of a tube of cocoanut charcoal immersed in liquid air.

When the stop-cock was closed, no α-particles could enter the vessel, and the steadiness of the electrometer needle could thus be tested at intervals during an experiment. On opening the stop-cock, a small fraction of the total number of α-particles expelled per second passed through the aperture into the detecting vessel. In practice, it was found convenient to arrange the intensity of the active matter and its distance from the opening so that from three to five α-particles entered the detecting vessel per minute. It became difficult to count a number greater than this with certainty, since the needle had not time to come to rest between successive throws.

The following example serves to illustrate the character of the observations. The source of α-rays in this case was a metal plate about 0·5 square cm. in area made active by exposure for several hours in a large quantity of radium emanation. Fifteen minutes after removal from the emanation, the α-radiation from the plate is due almost entirely to radium C. The active matter is in the form of a thin film, so that all the α-particles are expelled with the same velocity. The intensity of the radiation from radium C decreases with time, falling to half value about one hour after removal and later at a more rapid rate. In this particular case, the detecting tube was filled with carbon dioxide to a pressure of 4·2 cm. The E.M.F. applied was 1320 volts. The active plate was at a distance of 350 cm. from the aperture, which was of diameter 1·23 mm. Observations of the number and magnitude of the throws due to the α-particles were continued over an interval of 10 minutes. The results are shown in the following table :—

	Number of throws.	Magnitude of successive throws in scale divisions.
1st minute	4	11, 12, 10, 11
2nd "	3	10, 11, 8
3rd "	5	10, 9, 13, 8, 12
4th "	4	18,* 8, 12
5th "	3	10, 6, 10
6th "	4	9, 10, 12, 11
7th "	2	10, 11
8th "	3	11, 13, 8
9th "	3	8, 20*
10th "	4	8, 12, 14, 6
Average per minute = 3·5		Average throw = 10 divisions.

Each scale division was equal to 2·5 mm. The intensity of the α-radiation decreased about 15 per cent. during the time of observation.

When the stop-cock was closed so that no α-particles could enter the detecting vessel, the electrometer needle was very steady, the maximum excursion of the needle from the zero position in the course of 10 minutes being not more than three scale divisions. Only two or three excursions of such amplitude occurred in that interval. We see from the table that the average throw observed with the stop-cock open was 10 divisions.* All small excursions of magnitude less than three scale divisions are omitted. With the exception of the two numbers marked with asterisks, each of the throws given in the table is due to a single α-particle. The two large throws marked with asterisks are each due to the superposition of the separate effects due to two successive α-particles entering the detecting vessel within a few seconds of each other. This was readily seen from the peculiarity of the motion of the spot of light on the scale. As the electrometer needle was moving slowly near the end of its swing caused by the effect of one α-particle, a second impulse due to the entrance of another was communicated to it, and caused it to move again more rapidly. Such double throws occur occasionally, and are readily recognised, provided the interval between the entrance of successive α-particles is not less than one second.

It will be noted that the number of α-particles entering the opening per minute, and also the interval between successive throws varied within comparatively wide limits. Such a result is to be anticipated on the theory of probability. We may regard a constant source of α-rays as firing off α-particles equally in all directions at a nearly constant rate. The number per minute fired through a small opening some distance away is on the average constant if a large number of throws are counted. When only a small number of throws are observed over a short interval, the number is subject to considerable fluctuations, the probable percentage departure of the observed number from the correct average being greater the smaller the number of α-particles entering within a given time. This phase of the subject is of considerable interest and importance, and will be discussed in more detail later in the paper. It suffices here to say that the variation of the observed number per minute is well within the limits to be anticipated on the general laws of probability.

It is seen that the throws due to an α-particle are somewhat variable in magnitude. Such a result is to be anticipated for several reasons. In the first place, the α-particles do not all pass along the detecting tube at the same

* The magnitude of the throw due to a single α-particle is dependent upon the E.M.F. applied, and can be varied over wide limits.

distance from the axis. The magnification of the ionisation is less for those that pass closest to the central wire. In addition, as will be shown later, there is always a scattering of the α-rays by the mica screen and by the gas in the detecting vessel. This tends to spread out the pencil of rays in the detecting vessel, and consequently to introduce still greater differences in the effects due to individual α-particles.

Detection of α-Particles from Uranium, Thorium, Radium, and Actinium.

The throws of the electrometer observed with the stop-cock open have been ascribed to the α-particles fired into the detecting vessel. This can be readily proved by placing a thin screen between the source of radiation and the detecting vessel. The throws of the electrometer disappear if this screen, together with the mica plate covering the hole, is of the right thickness to stop the α-particles entirely. Under ordinary conditions, the effect due to a β-particle is very small compared to that due to an α-particle, and is not detectable. If a plate coated with the active deposit of radium is used as a source of radiation, it is found that the decay curve obtained by counting the α-particles emitted agrees closely with the ordinary α-ray decay curve.

By this electrical method, we have detected the expulsion of α-particles not only from radium and its products but also from uranium, thorium, and actinium. For example, a plate, made active by exposure to the emanation of a preparation of actinium, gave effects in the testing vessel due to an α-particle of about the same magnitude as that due to an α-particle from radium C. The decay curve obtained by counting the α-particles agreed closely with the known curve. A thin film of radium itself showed a similar effect. As the activity rose, consequent upon the production of fresh emanation and its occlusion in the radium, the number of α-particles entering the detecting vessel increased.

A special apparatus (see fig. 2) was used to detect the emission of α-particles from weak radio-active substances like uranium and thorium.

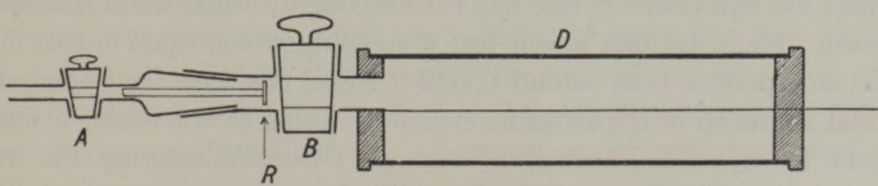

Fig. 2.

The active matter spread on a plate R (fig. 2) was placed about 5 cm. from the opening, which in this case was about 1 cm. diameter, and without any mica screen. A stop-cock of wide bore was placed between the active matter and the testing vessel D. With the stop-cock closed, the electrometer needle was very steady. On opening the stop-cock, about two throws per minute of the ordinary magnitude due to an α-particle were observed from the uranium. This was about the number to be expected from known data.

It may be of interest to record an experiment made with a preparation of thorium hydroxide. A small quantity of this (about 3 milligrammes) was wrapped in thin paper, which stopped the α-rays but allowed the emanation to pass through freely. On opening the stop-cock, the emanation diffused into the detecting vessel and immediately a large deflection of the electrometer was observed. After a few minutes an approximately steady radio-active state was reached. The electrometer needle, however, never remained steady, but made wide oscillations on either side of the mean position. Such an effect was to be anticipated, for when occasionally two or three α-particles from the emanation were fired along the cylinder within a second or two of each other, the electrometer needle was widely deflected. When the stop-cock was closed, the mean deflection due to the emanation in the testing vessel decreased with the time at the rate characteristic of the thorium emanation, but the electrometer needle continued to give excursions to and fro until the activity of the emanation had disappeared.

There is no doubt that the principle of automatic increase of the ionisation by collision can be used to extend considerably the range of measurement of minute quantities of radio-active matter.

Experimental Difficulties.

The final type of detecting cylinder which was found most satisfactory for counting purposes was of small diameter, viz., 1·7 cm., and of length not more than 25 cm. We shall now discuss the reasons that led us to adopt such a small detecting vessel. In the preliminary experiments, a cylinder of diameter 3·5 cm. and length 1 metre was used. With a pressure of air of 4 cm., the ionisation and stopping power of an α-particle passing the length of the cylinder was equivalent to that due to traversing 5·3 cm. of air at atmospheric pressure. Since the mica screen had a stopping power equal to only 5 mm. of air, an α-particle from radium C (range 7 cm.) produced the major part of its total ionisation in the detecting cylinder. Using such a vessel, it was not difficult to adjust the voltage so that an α-particle entering the vessel produced a throw of at least 100 mm. of the electrometer scale. Under such conditions, however, it was found impossible to avoid natural disturbances of the electrometer needle, when the stop-cock was closed, which were comparable in magnitude and character with those due to the entrance of an α-particle. These sudden movements of the electrometer needle were not numerous, but were sufficient to interfere with an accurate counting of the number of α-particles. These disturbances were inherent in the vessel and could not be got rid of by changing the pressure or nature of the gas or the diameter of the central wire. There is no doubt these irregular movements of the electrometer needle must be ascribed to a slight natural radio-activity of the walls of the brass tube. An α-particle projected near the end of the tube in the direction of the axis of the tube would produce a throw of the electrometer needle of about the same magnitude as that due to an α-particle fired through the opening parallel to the axis of the tube. The great majority of the α-particles emitted by the tube will only travel a short distance before being stopped by the walls, and consequently will only give rise to small individual movements of the needle. The greater part of the current observed by the electrometer with the aperture closed was due to this natural ionisation increased several thousand times by the agency of the strong electric field. The correctness of this conclusion was borne out by the observation that any change of the applied voltage, and consequently of the magnification, altered the magnitude of the natural disturbances, and the throw due to an α-particle in about the same ratio. In addition, it was observed that the number of the natural disturbances fell off rapidly with decrease of the diameter of the detecting tube. For example, the natural movements of the electrometer needle, using a long tube of 5 cm. diameter, were so numerous and so vigorous that it was impossible to use it for counting α-particles at all. With a tube, however, of 1·7 cm. diameter, the natural movements were very occasional, and of magnitude small compared with the effect due to an α-particle. Such a rapid decrease of the disturbances is to be anticipated in the light of the above explanation. If tubes are taken of the same length and of the same natural radio-activity per unit area, but of different diameters, the total number of α-particles shot out is proportional to their radii. Taking corresponding cross sections of the tubes, the fraction of the total number of α-particles emitted, which travel to the end of the tubes without striking the walls, is proportional to the cross sectional area of the tubes. Consequently the number of α-particles which pass along the tube without being stopped by the walls varies directly as the cube of the radius. We thus see that the sudden large movements of the electrometer needle due to the radio-activity of the walls should fall off very rapidly with decrease of the diameter—a result in harmony with the experimental observations.

Since the electrical capacity of the detecting vessel was smaller than the electrometer and its connections, it seemed advisable at first to use long detecting tubes in order to make the ionisation in it due to an α-particle as large as possible. From lack of accumulators at our command, it was not found feasible to work at a higher pressure than about 6 cm. of mercury, for at this pressure about 1500 volts were necessary to obtain the requisite magnification. Experiments were consequently made with a tube 135 cm. long, of diameter 1·7 cm., with a gas pressure varying from 2 to 6 cm. The natural disturbances of the electrometer needle in this vessel were very small, and it was found possible to increase the magnification such that an α-particle produced a throw of several hundred millimetre divisions on the scale. The throws due to successive α-particles were, however, very variable in magnitude. This is illustrated by the following table of observations :—

Air pressure, 3 cm.; radium C. Source of radiation; distance from aperture, 350 cm.

	Number of α-particles.	Magnitude of successive throws.
1st minute ...	4	6, 7, 10, 16
2nd „ ...	2	21, 15
3rd „ ...	1	36
4th „ ...	4	6, 25, 17, 11
5th „ ...	4	4, 28, 13, 13
6th „ ...	5	9, 16, 7, 6, 24

The great difference in the magnitude of the throws could not be ascribed to several particles entering together, for similar divergences were noted when on an average only one α-particle entered the detecting vessel per minute. Special experiments were made with sources of radiation of small area at a distance of 4 metres from the aperture and with a small aperture in the detecting vessel. Under such conditions, it was arranged that if the α-particles travelled in straight lines, they should strike the end of the detecting tube within an area of 1 square mm. The use of such a theoretically narrow pencil of rays had no effect, however, in equalising the magnitude of the throws. Finally, after a series of experiments, it was found that this effect was due to the *scattering of the α-particles in their passage through the mica screen and through the gas in the detecting vessel.* In a previous paper by one of us,* attention had been directed to the undoubted scattering of the α-particles in their passage through matter, and the magnitude of this scattering had been determined by the photographic method in special cases. We did not at first realise the importance of this effect in our experimental arrangement. Some of the α-particles, in passing through the thin sheet of mica, are deflected from their rectilinear path, and this deflection

* Rutherford, 'Phil. Mag.,' August, 1906.

is continued in their passage along the gas of the tube. The scattering was sufficiently great to cause a large fraction of the α-particles to impinge on the walls of the tube. The small throws observed were due to α-particles which only traversed a small fraction of the length of the tube before being stopped, while the largest throws were due to those that passed along the tube without striking the walls. A special series of experiments by a new method were made by one of us* to determine the magnitude of this scattering in special cases. An account of these experiments will be published in a separate paper.

As it was not feasible to decrease the scattering by reducing the thickness of the mica screen over the opening, the only way of making the throws more uniform was to diminish the length of the tube. It was for this reason that a tube only 25 cm. long was used. In this short distance, the α-particles were not deflected sufficiently to strike the walls of the tube, and the great majority travelled the whole length of the detecting vessel. Under these conditions, the throws of the electrometer due to the α-particles at once became far more uniform. An example of the throws obtained in the short vessel is given in Table I, p. 145. In the long detecting tube, there was a tendency to overlook the small throws and thus to underestimate the number of α-particles entering into the detecting vessel. The presence of this scattering also makes it necessary to exhaust the long firing tube to a low pressure. The presence of gas in this tube tends to deflect the α-particles from their rectilinear path and, if the tube is narrow near the aperture, to reduce the number entering the detecting vessel. Such a decrease of the number was at once observed, if the pressure of the gas in the long tube were raised so that its stopping power was equivalent to 2 or 3 cm. of air at atmospheric pressure.

The Number of α-Particles expelled from Radium.

A series of experiments was made to determine as accurately as possible by the electrical method the number of α-particles expelled per second from 1 gramme of radium. The arrangement of the apparatus was similar to that shown in fig. 1. A source of homogeneous α-rays was placed at a convenient distance from the detecting vessel in the firing tube, and the average number of α-particles entering the aperature per minute was determined by counting the throws of the electrometer needle.

Let Q be the average number of α-particles expelled per second from the source, consisting of a thin film of active matter. Let A be the area in

* See accompanying paper by H. Geiger, "Scattering of the α-Particles by Matter," p. 174, *infra*.

square cm. of the aperture in the detecting vessel, and r the distance in centimetres of the source of rays from the aperture. It was verified experimentally that the α-particles on an average are projected equally in all directions. Consequently, the fraction of the total number of α-particles expelled from the source which enter the detecting vessel is equal to the area of the aperture divided by the area of the surface of a sphere of radius equal to the distance of the source from the opening. The average number n of α-particles entering the opening per second is thus given by

$$n = \frac{QA}{4\pi r^2}. \tag{1}$$

This expression holds for all distributions of active matter of dimensions small compared with the distance r, provided that each element of surface of the source can fire directly into the aperture. In practice, the active matter is usually spread on the surface of a body of sufficient thickness to stop the α-particles fired into it, so that only half the total number of α-particles escape from its surface. This in no way interferes with the correctness of the above expression for the number.

After some preliminary experiments with thin films of radium itself, it was decided to employ radium C as a source of α-rays in the counting experiments. If a body is exposed for about three hours in the presence of the radium emanation, the activity imparted to it reaches a maximum value. Fifteen minutes after removal of the body from the emanation, the radiation due to radium A has practically disappeared, and the α-radiation is then due entirely to radium C. Under these conditions all the α-particles escape with the same velocity, and have a range in air of 7 cm. The use of radium C has numerous advantages. The active deposit is in the form of an extremely thin film, and the amount of active matter deposited on a body can readily be varied by altering the amount of emanation or the surface exposed to it. The chief advantage, however, lies in the ease and certainty of measurement of the quantity of active matter present in terms of the radium standard.* The penetrating γ-rays from radium in equilibrium arise entirely from its product, radium C. Consequently, by comparing the γ-ray activity of the active deposit with the radium standard, the amount of radium C present may be expressed in terms of the quantity of radium C in equilibrium with 1 gramme of radium. The chief disadvantage lies in the fact that the activity due to radium C rapidly diminishes, falling to half value in about one hour and to 14 per cent. of the maximum in two hours.

* The radium standard employed in these experiments is one that has been in use for several years. It is a part of a sample of radium which gave a heating effect of 110 gramme-calories per hour per gramme.

The shape of the body made active by exposure to the emanation must be such that each element of the active surface, when in position in the firing tube, must be in full view of the aperture of the detecting vessel. Examples of the surfaces employed are shown in fig. 3: *a* and *b* are of glass, and *c* a

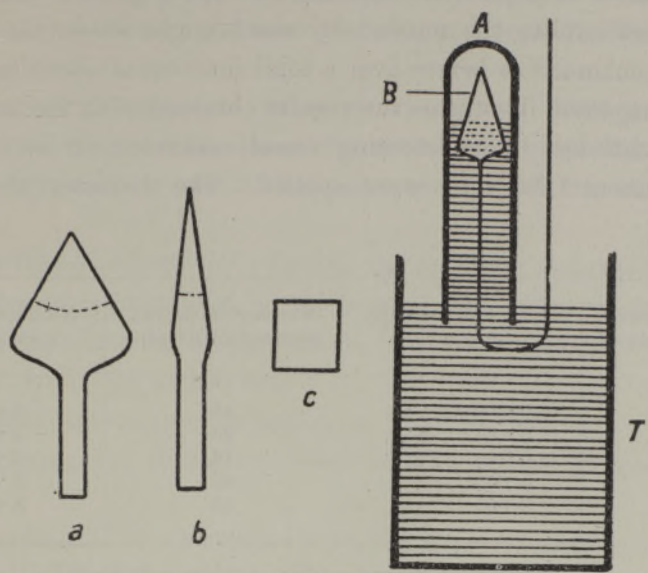

Fig. 3.

plane sheet of glass or iron, the dotted lines representing the lower limit of the active matter. The emanation, mixed with 1 or 2 c.c. of air, was collected over mercury in the end of the tube A (fig. 3). The body B, to be made active, was fixed to a glass U-tube, and introduced into the emanation space by means of the mercury trough T. After remaining in position for an interval of not less than three hours, the active body was removed and immediately tested in terms of the radium standard, using a fixed γ-ray electroscope. The active source was then placed in position in the firing tube, which was exhausted to a low pressure. In order to follow the changes of activity of the source, a second travelling γ-ray electroscope was employed in which the activity of the source was determined *in situ*. In the counting experiments the active body, as it diminished in activity, was moved nearer the detecting vessel. The electroscope was moved so as to be always directly over the active body and always at the same distance from it. At the end of the counting experiments, the active body was removed from the firing tube and its γ-ray activity again determined on the fixed electroscope. In this way a complete check was obtained on the activity measurements as well as a direct determination of the decay curve of the active body.

The general procedure of an experiment was as follows. After the γ-ray activity had been accurately measured, observations of the number of throws were made continuously for an interval of 10 minutes. The γ-ray activity was then determined again, and then another 10 minutes' count, and so on. When the number of α-particles entering the opening had fallen to between one and two per minute, the active body was brought nearer the opening, and observations continued as before over a total interval of about two hours.

The following table illustrates the results obtained with the same source as if decayed in activity. The detecting vessel contained air at a pressure of 3·75 cm. and about 1200 volts were applied. The diameter of the aperture was 1·23 mm.

Distance of active body from aperture.	Mean γ-ray activity of source.	Number of throws observed in 10 mins.	Number of α-particles expelled per gramme.
350 cm.	0·309 mgr. Ra	45	$3·06 \times 10^{10}$
350 "	0·154 "	25	$3·33 \times 10^{10}$
350 "	0·11 "	16	$2·96 \times 10^{10}$
150 "	0·055 "	49	$3·43 \times 10^{10}$
150 "	0·031 "	25	$3·11 \times 10^{10}$

Total number of throws = 160. Average = $3·18 \times 10^{10}$.

The second column gives the mean γ-ray activity of the source in terms of milligrammes of pure radium in equilibrium. The fourth column gives the total number of α-particles from radium C expelled per second in 1 gramme of radium in equilibrium. This number is calculated as follows. We have shown (equation 1) that the total number of α-particles Q emitted per second by the source is given by

$$Q = \frac{4\pi r^2}{A} \cdot n.$$

The total number Q_0 expelled for a γ-ray activity corresponding to 1 gramme of radium is given by

$$Q_0 = \frac{Q}{\rho} = \frac{4\pi r^2}{A} \cdot \frac{n}{\rho},$$

where ρ is the γ-ray activity of the source in terms of 1 gramme of radium. Since n and ρ are determined experimentally, and r and A are known, the value of Q_0 can be at once calculated. The calculated values of Q_0 for each experiment are given in the fourth column, and serve as a comparison of the agreement for the different observations.* The value of $4\pi r^2/A$ for the first

* On account of the probability variation, it is not to be expected that the numbers in the fourth column should agree very closely.

three experiments at a distance of 350 cm. is equal to $1 \cdot 25 \times 10^8$, *i.e.*, on an average, out of 125,000,000 α-particles fired from the source, only one passes through the aperture.

In the course of our experiments, we have verified, as far as possible, the correctness of the assumptions on which the deduction of the number of α-particles expelled from 1 gramme of radium depends. These points are summarised below :—

(1) For a given intensity of radiation at a given distance, the average number of throws observed in the electrometer in a given interval is independent of the pressure or nature of the gas, and also of the magnification of the ionisation.

(2) The number of α-particles entering the aperture is proportional to the activity of the source (measured by the γ-rays) and inversely proportional to the square of the distance of the source from the aperture over the range examined, viz., from 375 to 100 cm.

(3) For a given intensity of radiation at a given distance, the number of α-particles entering the detecting vessel is proportional to the area of the aperture.

(4) Using radium C as a source of rays, the α-particles are, on an average, projected equally in all directions. This has been verified by observing that, within the limit of experimental error, the calculated number of α-particles from radium C in 1 gramme of radium comes out the same whether the α-particles entering the aperture escape nearly tangentially from the active surface, as in fig. 3, *b*, nearly normally, as in fig. 3, *c*, or at an intermediate angle, as in fig. 3, *a*.

The following table gives the results for a number of separate experiments. The average value of Q_0 for each complete experiment, involving observations for different intensities of the source at different distances, is given in the last column.

Gas.	Pressure in detecting vessel.	Voltage.	Diameter of aperture.	Total number of throws counted.	Average value of Q_0.
Air	3·75 cm.	1200 volts	1·23 mm.	161	$3 \cdot 20 \times 10^{10}$
CO_2	4·8 ,,	1360 ,,	1·23 ,,	59	$3 \cdot 10 \times 10^{10}$
,,	4·8 ,,	1360 ,,	1·23 ,,	118	$3 \cdot 30 \times 10^{10}$
,,	4·1 ,,	1240 ,,	1·23 ,,	93	$3 \cdot 13 \times 10^{10}$
,,	4·2 ,,	1320 ,,	1·92 ,,	194	$3 \cdot 43 \times 10^{10}$
,,	4·2 ,,	1320 ,,	1·92 ,,	150	$3 \cdot 34 \times 10^{10}$
,,	3·2 ,,	1320 ,,	1·92 ,,	99	$3 \cdot 43 \times 10^{10}$
				Average =	$3 \cdot 28 \times 10^{10}$

Except for the last experiment, in which a tube 21 cm. long and 2·4 cm. diameter was used, the detecting tube was of length 25 cm. and internal diameter 1·7 cm.

In determining the average value of Q_0 given in the last column, which is itself an average of a large series of experiments, the weight to be assigned to each experiment of the series was taken as proportional to the number of α-particles counted. It was found that this differed only slightly from the arithmetic mean. It is seen that the mean value of the collected observations for Q_0 is $3{\cdot}28 \times 10^{10}$. This is subject to a small correction for which it is difficult to assign a definite value. In order to make the throws due to an α-particle as uniform as possible, it was arranged that the α-particle passed obliquely across the detecting tube. A small fraction of the α-particles entering the aperture would be stopped by the central wire, diameter 0·45 mm. In counting the number of α-particles, there is a tendency to overlook or put down to natural disturbances all movements which are small compared with the average value. This would be the case if an α-particle were stopped before travelling half the length of the tube. Taking into account the dimensions of the aperture, and of the copper wire and the scattering of the beam in its passage through the mica and the gas, it has been estimated that this correction cannot be more than 3 per cent. Making the correction, the value of Q_0 becomes to the nearest figure $3{\cdot}4 \times 10^{10}$.

We consequently conclude that, on an average, $3{\cdot}4 \times 10^{10}$ α-particles are expelled per second from the radium C present in 1 gramme of radium in equilibrium. From the experiments of Bragg, and the measurements by Boltwood of the ionisation due to the α-particles from each of the products of radium, it appears certain that the same number of α-particles are expelled per second from radium itself and from each of its α-ray products in equilibrium with it.

It follows that 1 gramme of radium itself and each of its ray products in equilibrium with it expels $3{\cdot}4 \times 10^{10}$ α-particles per second. The total number of α-particles emitted per second per gramme of radium in equilibrium with its three α-ray products is $13{\cdot}6 \times 10^{10}$. Taking as the simplest and most probable assumption that one atom of radium in breaking up emits one α-particle, it follows that in 1 gramme of radium $3{\cdot}4 \times 10^{10}$ atoms break up per second.

Counting of Scintillations.

It is of importance to compare the number of scintillations produced on a zinc sulphide screen with the number of α-particles counted by the electric method, in order to see whether each scintillation is due to a single α-particle.

For this purpose the special zinc sulphide screens provided by Mr. F. H. Glew were used. A thin layer of zinc sulphide is spread over a thin glass plate, and the scintillations produced on the screen are readily seen through the glass by means of a microscope. In order to make the comparison as direct as possible, the same firing tube and aperture, covered with the mica screen, were used. The brass detecting tube was removed, and a small piece of zinc sulphide screen was attached to the end of the glass tube D (fig. 1), with its active surface towards the firing tube. Radium C served as a source of α-rays, as in the electrical method.

Regener* has made a number of observations upon the number of α-particles expelled from an active preparation of polonium by the scintillation method. He has investigated the best conditions for viewing the scintillations and the relation between the focal lengths of the eye-piece and objective to obtain the maximum illumination due to each α-particle. We have found his suggestions very useful in these experiments.

In our experiments a microscope of magnification 50 was used. The small area of screen, struck by the α-particles, covered only about one-half of the field of view. The experiments were made at night in a dark room. As Regener suggests, it is advisable to illuminate the screen slightly by artificial light, in order to keep the eye focussed on the screen. The distance and intensity of the source were adjusted so that from 20 to 60 scintillations were observed per minute. It is difficult to continue counting for more than two minutes at a time, as the eye becomes fatigued. The zinc sulphide screen usually showed a few scintillations per minute with the stop-cock closed, due to natural radio-activity and other disturbances. These were counted before and after each experiment, and were subtracted from the number counted with the stop-cock open. It was usual to count 100 scintillations and to note the time with a stop-watch. The results of a series of observations for varying intensities of the radiations are given in the following table. The corrected number of scintillations observed per minute is given in Column 1. Taking $3{\cdot}4 \times 10^{10}$ as the number of α-particles expelled per second per gramme, the calculated number of scintillations to be expected from the intensity of the radiation, if each α-particle produces a scintillation, is given in Column 1. The ratio of the observed to the calculated number is given in Column 3.

* 'Verh. d. D. Phys. Ges.,' vol. 10, p. 78, 1908.

Diameter of aperture, 1·23 mm. with mica covering. Distance of active source from aperture, 200 cm.

I. Calculated number of α-particles per minute.	II. Observed number of scintillations per minute.	III. Ratio of observed to calculated number.
39	31	0·80
38	49	1·29
34	29	0·85
32	31	0·97
31	32	1·03
28	27	0·96
27	28	1·04
25	21	0·84
23	25	1·09
21	21	1·00
Total number = 294		Average = 0·99

Another series of observations was made with a fresh piece of zinc sulphide screen with an aperture 3·02 times area of the first and without mica screen.

Calculated number of α-particles per minute.	Observed number of scintillations per minute.	Ratio of observed to calculated number.
36	31	0·86
34	30	0·88
31	31	1·00
30	29	0·97
27	29	1·07
Total number = 150		Average = 0·96

Considering the probability error, the agreement between the electrical and optical methods of counting is, no doubt, closer than one would expect. The result, however, brings out clearly that within the limit of experimental error, each α-particle produces a scintillation on a properly prepared screen of zinc sulphide. The agreement of the two methods of counting the α-particles is in itself a strong evidence of the accuracy obtained in counting the α-particles expelled per gramme of radium by the electrical method. It is now clear that we have two distinct methods, one electrical and the other optical, for detecting a single α-particle, and that the employment of either method may be expected to give correct results in counting the number of α-particles.

Since there is every reason to believe that an α-particle is an atom of helium, there are now two distinct methods of detecting the expulsion of a single helium atom, one depending on its electrical effect, the other upon the luminosity produced in crystals of zinc sulphide. It is not necessary here to enter upon a discussion of the mechanism of production of a scintillation. In a previous paper, one of us* has pointed out that there is strong reason to suppose that the molecules of the phosphorescent preparation are dissociated by the α-particle, and that the luminosity observed may accompany either this dissociation, or the consequent recombination of the dissociated parts.

Probability Error.

We have previously drawn attention to the fact that the number of α-particles entering a given opening in a given time is conditioned by the laws of probability. E. v. Schweidler† first drew attention to the fact that, according to the theory of probability, the number of α-particles expelled per second from radio-active matter must be subject to fluctuations within certain limits. If z is the average number of atoms of active matter breaking up per second, the average error to be expected in the number is $\sqrt{z}$. The existence of fluctuations in radiations from active matter of the magnitude to be expected on this theory have been shown by the experiments of Kohlrausch,‡ Meyer and Regener,§ and Hans Geiger.‖

In most of the experiments in this paper, an intense source of α-radiation has been used. If, for example, the source had a γ-ray activity equal to 1 milligramme of radium, the average number of α-particles expelled per second is $3{\cdot}4 \times 10^7$. The error to be expected is thus 5830 particles, and the relative error $\sqrt{z}/z$ is 1/5830.

In such a case, we may consider the source as a whole to emit α-particles at a practically constant rate. The probability variation in the number is beyond the limit of detection by ordinary methods. The case, however, is quite different when we consider the number of α-particles entering a small opening at a distance from the source. In the experiments, the number entering the detecting vessel varied between two and six per minute, and the number of α-particles counted in a single experiment varied from 20 to 60. Assuming, for simplicity, that the general theory applies to this case, the probable variation of the observed number from the true mean is equal to

* Rutherford, 'Phil. Mag.,' July, 1905.

† Schweidler, Congrès International pour l'Étude de la Radiologie, Liége, 1905.

‡ Kohlrausch, 'Wien. Ber.,' p. 673, 1906.

§ Meyer and Regener, 'Ver. d. D. Phys. Ges.,' No. 1, 1908.

‖ Geiger, 'Phil. Mag.,' April, 1908.

$\sqrt{z}$. This amounts to four or five particles for a number 20 and between seven and eight for a number 60. It is not easy to compare accurately theory and experiment in this way, but there is no doubt that the observed variations are of the same order of magnitude as those to be anticipated from the laws of probability.

Some experiments have been made, both by the electric and scintillation methods, to determine the distribution of the α-particles in time. For this purpose, a thin film of radium was used as a source of rays. A large number of α-particles was counted, the interval between successive entrances of the α-particles in the detecting vessel being noted. A curve is then plotted, the ordinates representing the number of α-particles and the abscissæ the corresponding time intervals between the entrance of successive α-particles. A curve is obtained like that shown in fig. 4, which is similar in general shape

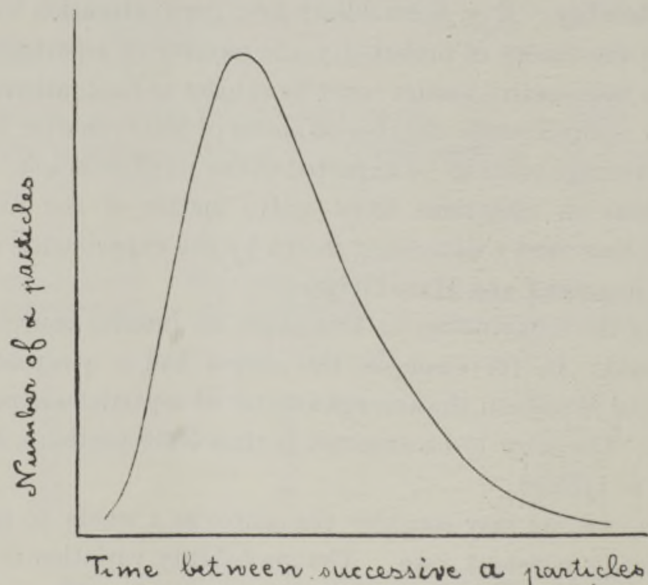

Fig. 4.

to the probability-curve of distribution in time. Further experiments are in progress to determine the distribution-curve as accurately as possible, in order to compare theory with experiment.

Summary of Results.

(1) By employing the principle of magnification of ionisation by collision, the electrical effect due to a single α-particle may be increased sufficiently to be readily observed by an ordinary electrometer.

(2) The magnitude of the electrical effect due to an α-particle depends upon the voltage employed, and can be varied within wide limits.

(3) This electric method can be employed to count the α-particles expelled from all types of active matter which emit α-rays.

(4) Using radium C as a source of α-rays, the total number of α-particles expelled per second from 1 gramme of radium have been accurately counted. For radium in equilibrium, this number is $3{\cdot}4 \times 10^{10}$ for radium itself and for each of its three α-ray products.

(5) The number of scintillations observed on a properly-prepared screen of zinc sulphide is, within the limit of experimental error, equal to the number of α-particles falling upon it, as counted by the electric method. It follows from this that each α-particle produces a scintillation.

(6) The distribution of the α-particles in time is governed by the laws of probability.

We have previously pointed out that the principle of magnification of ionisation by collision can be used to extend widely our already delicate methods of detection of radio-active matter. Calculation shows that under good conditions it should be possible by this method to detect a single β-particle, and consequently to count directly the number of β-particles expelled from radio-active substances.

Further work is in progress on this and other problems that have arisen out of these investigations.