(5) The value of n is probably 0.5 for monatomic gases, and 1.5 for diatomic gases, on the simple hypothesis of integral degrees of freedom. These values give very fair agreement with experiment, but there is no *a priori* reason why the number of degrees of freedom should be an integer for a polyatomic molecule.

(6) The properties of CO_2 at moderate pressures are well represented by the assumption $n=2$, provided that account is taken of the variation of the specific heat as observed by Regnault. This reconciles the hitherto discordant results for the cooling-effect and the compressibility.

(7) The properties of Steam, including the variation of the latent heat and the saturation-pressure, are well represented by the value $n=3.3$, if the limiting value of the specific heat at zero pressure is assumed to be independent of the temperature, provided that the variation with pressure is not neglected.

(8) The value of the Absolute Zero, as deduced from the pressure-coefficient of hydrogen, is probably within one or two hundredths of a degree of $273^{\circ}.10$.

IV. *Excited Radioactivity and the Method of its Transmission.* By E. RUTHERFORD, M.A., D.Sc., Macdonald Professor of Physics, McGill University, Montreal*.

- § 1. Introduction.
- § 2. Connexion between the excited activity and the emanations.
- § 3. Method of transmission of excited activity.
- § 4. Velocity of carriers of thorium-excited activity.
- § 5. Increase of excited radiation with time.
- § 6. Radium-excited activity.
- § 7. Distribution of excited activity on the anode.
- § 8. Velocity of the carriers.
- § 9. Origin of the carriers.
- § 10. Nature of the radiations.
- § 11. Evidence of chemical changes.
- § 12. Summary of results.

§ 1.—Introduction.

ONE of the most interesting properties of the radioactive substances thorium and radium, is their power of communicating or exciting † temporary radioactivity to all bodies in their neighbourhood. If a wire charged to a high negative

* Communicated by the Author. Preliminary accounts of these results were communicated to the American Physical Society, New York, December 27, 1901. and to *Phys. Zeit.* No. 10, 1902.

† (Note)—The term "*excited*," has been used throughout these investigations rather than "*induced*," which has found favour with many

potential is placed in a closed metal vessel containing a thorium or radium compound the excited radioactivity is confined almost entirely to the negative electrode. If the wire is charged positively, it remains inactive, but the excited radioactivity is produced on the walls of the vessel.

When no electric field is acting, excited radioactivity is produced on the surfaces of all bodies in the closed vessel, independently of their being good conductors or insulators.

In previous papers the author has shown that there is a direct connexion between the presence of the radioactive emanation from thorium and radium and the production of excited radioactivity. It will be shown in this paper that the production of excited radioactivity is one of the properties of the emanation from thorium and radium. This excited radiation is caused by the deposit on the surface of bodies of radioactive matter, which is transmitted by positively charged carriers travelling through air in an electric field with about the same velocity as the positive ion, produced in air by Röntgen rays.

§ 2.—*Connexion between Excited Radioactivity and Emanation.*

In a previous paper (Phil. Mag. Jan. 1900) I have shown that thorium compounds continually give off a radioactive emanation. This emanation loses its radiating power rapidly, falling to half value in the course of one minute. Dorn showed later that radium also gave off an emanation, especially when heated. This emanation decayed much more slowly than that from thorium. In some experiments where the radium emanation, mixed with air, was kept in a closed metal vessel, I have found that the activity of the emanation fell to half value after standing several days, but was quite appreciable after a month's interval.

These emanations from thorium and radium behave in all respects like radioactive gases or vapours. They diffuse rapidly through gases and through porous substances like paper, but unlike the gaseous ions which they produce in their path, pass through plugs of cotton wool, and bubble through solutions with no appreciable absorption. In a more detailed investigation (Rutherford and Soddy, Phil. Mag. Sept. 1902) it has been shown that the thorium emanation behaves like an inactive gas, and that its activity is not

Physicists. I have avoided using the latter term, as to my mind, it conveys the idea that the effect is in some way due to an action across the medium; while the experiments in this paper show conclusively that excited radioactivity is transmitted by means of a convection of positively charged carriers.

appreciably influenced by temperature or by the most drastic chemical treatment.

From the rate of diffusion of these emanations, it appears that they must possess a considerable molecular weight. An investigation of the rate of diffusion of the radium emanation into air, a preliminary account of which appeared in '*Nature*,' 1901, p. 157, and *Proc. Roy. Soc. Canada* 1901, showed that its molecular weight probably lay between 40 and 100. On account of the rapid loss of activity of the thorium emanation it has not so far been found possible to determine with certainty its rate of diffusion into air or other gases.

The emanations from thorium and radium possess very similar properties. They both readily diffuse through gases and porous substances; they both possess the power of ionizing the gas in their neighbourhood and producing excited radioactivity on bodies.

The differences between them can be readily accounted for by supposing them to be radioactive gases or vapours of different molecular weights. According to the results which have been given in previous papers, radioactivity is an accompaniment of chemical change. Taking this view, the difference in the rates of decay of the radioactivity of the emanations from thorium and radium merely indicates a difference in their rate of chemical change, and does not imply any fundamental difference in nature.

Unlike the radiations from the emanations, the excited radiation due to thorium decays much more slowly than that due to radium (see Rutherford and Miss Brooks, *Phil. Mag.* July 1902, p. 18). In this case, the chemical change proceeds more rapidly in the material responsible for the excited radioactivity from radium than in that from thorium.

Excited radioactivity is always produced on bodies when the radioactive emanations from thorium and radium are present. In order to show the very close connexion existing between the presence of these emanations and excited radioactivity, the following experimental facts may be mentioned:

- (1) Only the radioactive substances which emit emanations, viz. thorium and radium, have the power of exciting radioactivity. Uranium and polonium, not giving off any emanation, do not possess the power of exciting radioactivity.
- (2) The amount of excited radiation obtained from thorium and radium compounds is directly proportional to the amount of emanation present. For example, thoria gives out far more emanation and produces far more excited activity than thorium nitrate in the solid state. Thoria and radium chloride, partly deemanated by strong heating, lose their power of exciting activity in like ratio.

- (3) Excited radioactivity can be produced on bodies if the emanation and not the radioactive substance itself is present. It can be produced at long distances from the radioactive compound by blowing the emanation mixed with air along tubes. In the case of radium, the emanation, which has been introduced into a vessel by blowing a current of air over the active substance, produces excited activity after a month's interval, although the radioactive substance itself has not been placed in the neighbourhood. On the other hand, the power that a thorium or radium compound has of producing excited activity on a body near it is almost completely lost by blowing over the compound a current of air which removes the emanation as rapidly as it is formed.

The amount of emanation or excited activity has no direct connexion with the radioactivity of the compound in its neighbourhood, and cannot be ascribed to any action of the "straight line" radiation in the gas through which it passes. For example, deemanated thoria produces only a small fraction of the amount of excited activity of an equal weight of ordinary thoria although the amount of the straight line radiation is not much affected by the process of deemanation.

§ 3. *Method of Transmission of Excited Activity.*

The characteristic property of excited radioactivity is that it can be confined to the cathode in a strong electric field. It is probable, therefore, that the radioactivity is due to the transport, in the electric field, of positively charged carriers of some kind. Experiments were undertaken to test this and to find the rate at which these carriers moved in an electric field, in order to obtain a rough estimate of their dimensions compared with a gaseous ion.

The method employed to determine this velocity is a modification of one already used in a determination of the velocity of the negative ion, produced at the surface of a metal by ultra-violet light*. It depended on the use of an alternating electric field. By means of a revolving commutator, a direct P. D. was commuted into an alternating P. D. of known frequency. If such an alternating field is applied to two parallel plates, between which a radioactive emanation is kept uniformly distributed, equal amounts of excited activity are produced on each electrode.

If in series with an alternating P. D. $\pm E_0$ a battery is placed of E.M.F. E_1 less than E_0 , the positive carrier moves

* Rutherford, Proc. Camb. Phil. Soc. 1897.

in a stronger electric field in one half alternation than in the other. A carrier consequently moves over unequal distances during the two half alternations, since the velocity of the carrier is proportional to the strength of the electric field in which it moves. It follows from this that the excited radioactivity will be unequally distributed over the two electrodes. If the frequency of alternation is sufficiently great, only the positive carriers within a certain small distance of one plate can be conveyed to it, and the rest, in the course of several succeeding alternations, are carried to the other plate.

Suppose A and B (fig. 1) are two parallel plates to be

Fig. 1.

made radioactive. The emanation is supposed to be uniformly distributed between them. When B is negatively charged suppose the P. D. between the plates is $E_0 - E_1$, when B is positive the P. D. is $E_0 + E_1$.

Let d = distance between the plates,

T = time of a half alternation,

ρ = ratio of the excited radioactivity on the plate B to the sum of radioactivity on the plates A and B.

K = velocity of the positive carriers for unit-potential-gradient.

On the assumption that the electric field between the plates is uniform, and that the velocity of the carrier is proportional to the electric field, the velocity of the positive carrier towards B is

$$\frac{E_0 - E_1}{d} \cdot K,$$

and in the course of the next half alternation

$$\frac{E_0 + E_1}{d} \cdot K$$

towards the plate A.

The greatest distances* x_1, x_2 passed over by the positive carrier during two succeeding half alternations is thus given by

$$x_1 = \frac{E_0 - E_1}{d} K \cdot T, \text{ and } x_2 = \frac{E_0 + E_1}{d} K \cdot T.$$

Suppose the positive carriers are produced at a uniform rate of q per second for unit distance between the plates. The number of positive carriers which reach B during a half alternation may be divided into two parts :

- (1) One half of those carriers which are produced within the distance x of the plate B. This number is equal to

$$\frac{1}{2}x_1qT.$$

- (2) All the carriers which are left within the distance x_1 from B at the end of the previous half alternation. The number of these can be readily shown to be

$$\frac{1}{2}x_1 \cdot \frac{x_1}{x_2} qT.$$

Now all the rest of the carriers produced between A and B during a complete alternation will reach the other plate A in the course of succeeding alternations, provided no appreciable recombination takes place. This must obviously be the case, since the positive carriers travel further in a half alternation towards A than they return towards B during the next half alternation. The carriers thus move backwards and forwards in the changing electric field, but on the whole move towards the plate A.

The total number of positive carriers produced between the plates during a complete alternation is $2dqT$. The ratio ρ of the number which reach B to the total number produced is thus given by

$$\rho = \frac{\frac{1}{2}x_1qT + \frac{1}{2}x_1 \frac{x_1}{x_2} qT}{2dqT} = \frac{1}{4} \frac{x_1}{d} \cdot \frac{x_1 + x_2}{x_2}.$$

Substituting the value of x_1 and x_2 we obtain

$$K = \frac{2(E_0 + E_1)}{E_0(E_0 - E_1)} \cdot \frac{d^2}{T} \cdot \rho.$$

In the experiments the values of E_0, E_1, d , and T were varied, and the results obtained were in general agreement with the above equation.

* In the equations that follow it is assumed that x_1 is less than the distance between the plates. If $x_1 > d$ the equations have to be modified.

§ 4. *Velocity of Carriers of Thorium Excited Activity.*

For experiments on thorium emanation, a thick layer of thoria was placed in a shallow copper vessel inside an ebonite box 11 cms. square and 3 cms. deep, which was tightly waxed down to a metal base. The thoria was completely covered with two layers of filter-paper, which cut off most of the direct radiation, but readily allowed the emanation to pass through. The apparatus was rendered air-tight by a metal lid, dipping into a mercury trough round the top of the ebonite box. At the beginning of an experiment a square sheet of aluminium foil was placed over the paper covering the thoria, a zinc plate on top of the ebonite box, and the lid placed in position. This was done as quickly as possible, and the alternating electric field was then applied.

The emanation rapidly diffused through the paper and thin aluminium foil, and distributed itself between the plates in the electric field. After an interval, varying in the experiments from 20 to 90 minutes, the aluminium and zinc plates were removed and their radioactivity tested in the usual way with the Dolezalek electrometer. The ratio of the excited radioactivity on the two exposed plates was thus determined. This ratio was found to be independent of the time the plates were left before testing, as the radioactivity on each plate decays at the same rate.

The amount of thoria used in these experiments varied from 25 to 100 grammes. The amount of excited activity in a given time varied with the amount of thoria, but the ratio of the activity in the two plates was unaltered. For a given voltage and time of alternation the value of ρ was slightly greater when the lower plate was negative. This is due to the unequal distribution of the emanation between the plates; for on account of the time taken in diffusion, the emanation is more concentrated near the surface of the thoria. The mean of the values of ρ with top plate negative and lower plate negative was taken as the true value.

In the early experiments a two-part commutator driven by a motor was used. In the later experiments for a more rapid rate of alternation a four-part commutator was employed.

With this arrangement of apparatus a large number of experiments were made in order to test the truth of the general theory. Comparisons of the velocity of the carrier have been made over a wide range of period of alternation and of voltage, and for different distances between the plates. The results obtained were in general agreement with the theory put forward. When the voltage was kept constant, the value

of ρ was found to decrease with increase in the number of alternations per second. With a constant speed of alternation the value of ρ increased with the voltage.

When the value of ρ is small, the velocities of the carriers, deduced from the equation, were found to be all too high, and also inconsistent among themselves. There are several disturbing factors which have a great influence on the value of ρ when ρ is small. These factors are :—

(1) Recombination and diffusion of the carriers. Unless the electric field is strong, the carriers recombine and diffuse to the electrodes. With a weak electric field the excited radioactivity is distributed on both the positive and negative electrodes.

(2) Inequality of the electric field. In this simple theory we have assumed that the potential-gradient between the plates is uniform. This is far from being the case. The experiments of Child and Zeleny have shown that there is always a sudden drop of potential near the electrodes. The electric field near the electrodes is consequently stronger than the average. For this reason, when the carriers which reach the plate B are only abstracted from within a short distance of the plate, the value of ρ leads to too high a value of the velocity.

(3) Initial velocity of the carrier. From some considerations which will be developed later (see § 9), it seems probable that the positive charge on the carrier is due to the expulsion of a negatively-charged particle of some kind from the neutral molecule. The positive carrier may thus have enough initial velocity imparted to it to carry it some distance against the electric field. This will result in a distribution of some excited activity on the anode, even with a strong field. It is difficult to obtain direct experimental evidence on this point, but there seems little doubt that such an effect is present.

In order to obtain consistent results, it was found necessary to have a considerable difference between the strength of the electric fields during the succeeding half alternations. If the difference is small, the carriers take so long to reach the plate A that recombination and diffusion of the ions become important factors in determining the distribution of excited activity. For the reasons we have explained above, it was necessary to use fairly high voltages and correspondingly rapid speed of alternation in the experiments.

The following tables are examples of some of the results obtained for different voltages and distances between the plates. Temperature 18° C. Air fairly dry.

Plates 1.30 cm. apart.

$E_0 + E_1$.	$E_0 - E_1$.	Alternations per sec.	ρ .	K.
75	50	57	.17	1.7
152	101	57	.27	1.25
225	150	57	.38	1.17
300	200	57	.44	1.24

The value of K is given in cms. per sec. for a potential-gradient of 1 volt per cm.

For the last example, since the carrier travelled over a distance greater than 1.30 cm. during each half alternation, a modified form of the equation was necessary to calculate the velocity.

The value 1.6 cm. per second for 50 volts is too high for the reasons explained above.

Plates 2 cms. apart.

$E_0 + E_1$.	$E_0 - E_1$.	Alternations per sec.	ρ .	K.
273	207	44	.37	1.47
300	200	53	.286	1.45

An average value of the velocity from a large number of alternations for different distances, voltages, and speed of alternations was about 1.3 cm. per sec. for atmospheric pressure and temperature.

This velocity is not very different from the velocity of the positive ion produced by Röntgen or Becquerel rays. The most accurate determination of this velocity by Zeleny* gave a value of 1.37 cm. per sec. for dry air.

§ 5. *Increase of Excited Radiations with Time.*

In the course of these experiments a remarkable effect was observed. It was found that a plate which has been exposed a short time in the presence of thoria emanation, after being removed, gradually increased in radioactive power for several hours. The amount of increase varied with the time of exposure to the emanation, but in short exposures it increased to three or four times its initial value. For exposures of several hours the effect is not so marked, and is difficult to detect after a day's exposure.

The following tables illustrate the results obtained.

* Phil. Trans. Roy. Soc. (1900).

- (1) Platinum wire, charged -110 volts, exposed 15 minutes in a cylinder containing thoria. First observation five minutes after removal of wire from emanation cylinder.

Time in Minutes.	Movement of Electrometer in scale-divs. per sec.
0	1.9
7.5	2.8
24	4.0
43	4.6
58	5.2
78	5.9
99	6.5

In this case the activity had increased over three times in 99 minutes and had not reached its maximum value.

- (2) Aluminium foil as cathode in parallel plate apparatus of fig. 1. Time of exposure 41 minutes. First observation 6 minutes after removal.

Time.	Radioactivity.
0	1
21 minutes.	1.6
31 "	1.8
57 "	2.0
70 "	2.2
91 "	2.5
120 "	2.9
160 "	2.9
180 "	2.9
22 hours.	1.0
49 "	0.21

In this case, for the purpose of comparison, the initial value of electrometer current is taken as unity. The activity increases to nearly three times its initial value after an interval of two hours, and then slowly decreases at the normal rate, *i. e.*, it falls to half value in about 11 hours.

Similar results were obtained if the plate was made active without the action of the electric field. The increase of activity with time is independent of the nature of the electrode, or of the concentration of the radioactive matter upon it. It was not found possible to influence the rate of increase of activity with time or the final maximum by heating the wire to about a red heat.

With increase of time of exposure of the electrode in the thorium emanation, the ratio of increase of activity after

removal decreases. For a long interval of exposure the activity begins to decrease at once after removal. This result is to be expected, for the activity of each portion of the radioactive matter deposited increases with time for two or three hours, and then diminishes. Consequently, after the electrode has been exposed for about ten hours or more, the increase of activity of the matter deposited in the last few hours does not compensate for the decrease of activity of the radioactive matter as a whole. This increase of activity with time explains an irregularity in the curve of increase of excited activity from thorium with time of exposure. It was pointed out in a previous paper (Phil. Mag. February 1900, p. 178) that, on the hypothesis of a uniform rate of deposit of radioactive matter, the activity of which decreased in a G. P. with the time, the curve of rise of excited activity with time of exposure is the same as the curve of rise of an electric current in a circuit of constant self-inductance. It was experimentally observed, however, that the rate of increase for the first few hours was much smaller than would be expected on this hypothesis. In the light of the present results, the explanation of this effect is simple. The matter deposited during the first few hours does not reach its maximum activity for several hours, and the initial effect is consequently much smaller than would be expected on the simple theory.

§ 6. *Radium Excited Radioactivity.*

Experiments were made to determine the velocity of the carriers responsible for the excited radioactivity of radium in the same way as for thorium. The radium in my possession gave out too little emanation at ordinary temperatures in the solid state to enable me to use it in the apparatus in place of thoria. The amount of emanation from radium can, however, be increased several thousand times by heating the radium compound below a red heat. A more convenient method of obtaining a large amount of emanation is to dissolve a small quantity of radium chloride in water. Radium in solution gives off several hundred times more emanation than in the solid state. If the solution is kept in a closed vessel the emanation continually collects in the upper part of the vessel. It can be transferred at any time to another vessel by bubbling a slow current of air through the solution. The procedure adopted in introducing the emanation into the apparatus was as follows :—

A large amount of emanation was introduced into a metal cylinder of about 3 litres capacity. The plates to be tested were then placed in position in the apparatus of fig. 1,

and the alternating E.M.F. applied. By means of side tubes, the apparatus was put into connexion with the emanation vessel and a small fraction of the emanation introduced into it by sending a slow current of air into the cylinder. The tubes leading into the apparatus were then closed, and the alternating E.M.F. continued for an interval of 15 to 30 minutes. Before stopping the commutator, the emanation was blown out of the apparatus by a slow current of air. The plates were then removed, and the amount of activity on them was compared by means of the electrometer. On account of the initial rapid decay of the radiations, a difficulty arose in comparing the amount of radiation on the two plates. As shown in a previous paper*, the excited radiation from radium, for short exposures, decreases rapidly for the first 5 minutes after removal, but about 15 minutes after removal reaches a value which is maintained fairly constant for an interval of about 10 minutes. It then decays to zero, falling to half value in about 30 minutes. The comparison of the activity on the two plates was made during this constant interval.

When experiments were made under the same conditions as those for thorium, somewhat higher values of the velocity of the carriers were obtained, and the numbers, for different frequencies and voltages, differed considerably among themselves. These discrepancies were found to be due to the fact that even in a strong electric field from 5 to 10 per cent. of the total excited activity was distributed on the anode. In this respect the activity excited by radium differs from that of thorium. Consequently, the value of ρ would be greater for the radium than for the thorium experiment, under the same conditions, even if the carrier of excited radioactivity travelled at the same rate in both cases.

§ 7. *Distribution of Excited Activity on the Anode.*

In order to throw more light on the cause of this distribution of excited activity on the anode, some experiments were made with the apparatus shown in fig. 2 *a*.

The emanation vessel A consisted of a brass cylinder 25.5 cms. long and 8.30 cms. diameter. A long central brass rod B C D E, diameter .518 cm., passed through an ebonite cork at one end of the tube. The outside cylinder was connected to one pole of a large battery, the other pole of which was earthed. The central rod was connected to earth. The emanation was introduced into the vessel by sending a slow current of air through a radium chloride solution contained

* E. Rutherford and Miss Brooks, *Phil. Mag.* July 1902.

in the Drechsel bottle F. The air passed through a tube containing cotton-wool, and through a drying-tube T of calcium chloride. The central rod was made of three removable parts BC, CD, DE, screwed together. After exposure

for a known time in the presence of the emanation the rod was removed, and the activity on the portion CD, length 15 cms., determined by the electrometer in the cylindrical testing vessel L, shown in fig. 2 b. By this means the excited

activity was determined on that portion of the rod where the electric field was sensibly uniform.

In most of the experiments the emanation was introduced into the vessel A a day or two before observations were taken. This ensured a uniform distribution of the emanation throughout the cylinder by the process of diffusion. If observations were required soon after the introduction of the emanation, the emanation was uniformly mixed with the air by means of a stirrer not shown in the figure.

Some experiments were made with this apparatus on the amount of excited activity on the central rod when positively charged for different voltages. For the purpose of comparison, the results are expressed in terms of the percentage amount on the same electrode exposed for the same time when negatively charged with 300 volts between the electrodes. The rod was exposed in the presence of the emanation for 15 minutes, then removed and a fresh rod introduced. In

the course of three or four hours' work, the amount of excited activity, obtained for a given time of exposure, diminished about 20 per cent. This was partly due to the decay of the radiating power of the emanation during the interval, and partly to a slight escape of the emanation in removing and replacing the central rod. By determining the ratio of excited activity at the beginning and end of the experiments, a correction was readily made for this diminution.

Table of distribution on anode, diameter 8·3 mms.

Voltage.	Percentage.
—300	100
+ 300	6
+ 150	6
+ 50	9
+ 20	10
0	14

It will be seen from the table that the amount on electrode with + 300 volts P.D. is 6 per cent. of amount on electrode with —300 volts P. D. The percentage increases with diminution of voltage, rising to 14 per cent. for zero voltage, when the distribution is due to diffusion alone of the carriers to the central electrode. In order to see how much of this amount on the electrode was due to transmission by the electric field and how much to diffusion, the experiments were repeated with the central rod of ·8 mm. diameter instead of 8·3 mms.

—300	100
+300	4
+ 50	7

Now, with a central rod of only about 1/10 of the surface area, it is obvious that the effect due to diffusion must be very much reduced. We may thus conclude from these experiments that a proportion of the excited radioactivity from radium (about 5 per cent.) travels to the positive electrode in an electric field, and the carrier must in consequence have a negative charge.

A special experiment was made to determine the amount of radium excited activity on the anode with an apparatus consisting of parallel plates. For this purpose the emanation vessel of fig. 1 was used, with the plates 1·3 cm. apart. With 300 volts between the plates about 10 per cent. of the total activity was confined to the anode. From the previous experiments we have seen that about 5 per cent. reaches the

anode in consequence of transmission by the electric field. The remaining 5 per cent. must thus reach the electrode by other agencies. With such a strong electric field, the effect due to pure diffusion must be very small. It thus seems likely that some of the radioactive carriers have sufficient initial velocity to carry them to the electrode against the electric field.

§ 8. Velocity of the Carrier.

Some experiments were made on the velocity of the carrier of excited radioactivity, using the concentric cylinders shown in fig. 2*a*. If a and b are the radii of the internal rod and the cylinder, the electric field X at a distance r from the centre, for a P. D. of v volts between the cylinders, is given by

$$X = \frac{V}{r \log_e \frac{b}{a}}.$$

Using the same notations and assumptions as in the case of parallel plates, it can be shown that the velocity K of the carrier of excited radioactivity is given by

$$K = \frac{E_0 + E_1}{E_0(E_0 - E_1)} \frac{(b^2 - a^2) \log_e \frac{b}{a}}{T} \cdot \rho.$$

The following table shows some of the results obtained for different voltages and periods of alternation :—

$E_0 - E_1$.	$E_0 + E_1$.	Alternations per sec.	ρ .	Corrected value of ρ .	K .
205	308	5.7	.35	.32	1.0
205	308	11.2	.24	.20	1.2
205	308	17.2	.19	.13	1.2
205	308	34	.16	?	
385	580	18.3	.30	.27	1.5
385	580	47	.16	?	

The values of the velocity of the carrier determined for the *uncorrected* values of ρ in the above table vary much among themselves. It will be seen, however, that the value of ρ increases with the time of alternation and the voltage, as we should expect from the elementary theory. It was not found possible to reduce the observed value of ρ below about .16, whether the voltage was diminished or the frequency of alternation increased. This is to be expected, for we have

previously shown that the amount of excited activity in the central rod, when diffusion alone is acting, is $\cdot 14$ of the total; for in cases where the carrier is only able to travel over a small fraction of the distance between the electrodes during a half alternation, only a small amount of the excited radioactivity on the central rod is due to the deposit of positively charged carriers by the electric field. The greater proportion is due to the diffusion of carriers to the electrode and to the carriers which are deposited when the central rod is the anode. The amount of this latter is, as we have shown, about five per cent. of the total. It is difficult to make more than a rough estimate of the amount of excited activity on the electrode in the various cases. For these reasons the corrected values of ρ in the table for the frequencies of 34 and 47 per second are probably not more than about one-third of the observed value $\cdot 16$.

In the above table a rough correction is made for some of the values of ρ and the resulting velocity calculated.

It will be seen that the values of the velocity of the carrier lie between 1.0 and 1.5 cm. per sec. for a potential-gradient of 1 volt per cm. This is about the same range of values as that obtained for the carrier of thorium excited radioactivity.

From the nature of the results it is not possible to definitely decide whether the carriers of thorium and radium excited activity travel at exactly the same speed. The results, however, indicate that the carriers in the two cases are not very different in speed, and that consequently they do not differ much in size.

We may conclude from these experiments that the greater part of the excited radioactivity from both thorium and radium compounds is due to the deposit of positively charged carriers, produced from the emanations on the cathode, and that these carriers travel at about the same rate as the positive ion produced in the air by Röntgen rays.

When no electric field is acting excited radioactivity is transferred by the diffusion of these carriers to the surface of all bodies immersed in the emanation.

§ 9. *Origin of the Carriers.*

Before discussing the question of the method of production of these positive carriers which cause excited activity, a brief *résumé* is necessary of the physical properties of the emanations from thorium and radium. In the first place the emanations behave in all respects like radioactive gases of high molecular weight. They do not carry with them any

charge of electricity, and are consequently unaffected by the presence of an electric field.

In my first paper on the thorium emanation (*loc. cit.*) it was pointed out that the particles constituting the emanation certainly did not move with a velocity greater than $\cdot 00001$ cm. per sec. for a gradient of 1 volt. per cm. The conclusion was drawn that the emanation itself was initially uncharged. A similar result is true for the emanation from radium; for the emanation still persists in a closed vessel after several weeks' exposure in a strong electric field. For these reasons the suggestion made by Becquerel*, that the emanations are composed of positive ions directly emitted from radioactive bodies, is untenable; for if such were the case, the ions would at once be swept to the electrodes by the electric field, and would very rapidly disappear from the gas.

These emanations possess the property of ionizing the gas and of producing from themselves positively charged carriers which cause excited activity in bodies on which they are deposited. This property lasts only a few minutes in the case of the thorium emanation, and for several weeks for the radium emanation.

Two hypotheses may be put forward to account for the origin of these charged carriers:—

(1) The radioactive matter constituting the emanation condenses on the positive ions, produced in the gas by the radiation, and is thus transferred to the cathode.

(2) The particles of the emanation possess the property of expelling from themselves a negatively charged body of some kind. The particle would thus be left with a positive charge, and would be carried to the cathode by the electric field.

It is not easy to decide definitely between these two hypotheses, but the evidence as a whole is strongly in favour of the second.

In regard to (1) it might be supposed that the emanation condensed more readily on the positive than on the negative ion; on the principle that water and alcohol-vapour condense more readily on the negative than on the positive ion. If this were the case, it would be expected that the emanation would be removed more rapidly if the number of ions were increased in the gas through which the emanation was distributed. There is no evidence that such an effect exists. I have tried the experiment of passing the emanation through a space strongly ionized by radium rays; but the amount of excited activity in a given time on the cathode, placed in this

* *Comptes Rendus*, Dec. 9, 1901.

space, was not appreciably altered. I have also tried experiments to see if the radiation from the emanation was affected by exposure in a strong electric field, but with negative results. In order to test this, the thorium was placed in the bottom of a small lead box, and covered with two layers of paper to cut off the direct radiation. The top of the box was tightly covered with a very thin layer of mica. This prevented the escape of the emanation, but allowed the radiation from the emanation to pass through and ionize the gas above the vessel. The amount of this ionization outside the vessel was unchanged if the emanation was exposed to a strong electric field by charging insulated conductors placed inside the vessel.

If the emanation is removed to the cathode in an electric field by condensation on the ion, it is to be expected that it would continue to radiate at the same rate on the electrode as in the gas from which it is removed. On this view the radiation from the cathode should rapidly decrease for the first few minutes after removal from the emanation. Some special experiments were tried to settle this point, but no decrease was observed, although even a minute effect could have been readily detected.

There is thus considerable indirect evidence against the condensation hypothesis; and it has consequently been discarded in favour of (2), which offers a satisfactory explanation not only of the production of positive carriers, but also of the origin of the radiation given out by the emanation itself. On this view the emanation consists of matter in an unstable state, which undergoes further chemical change. The change consists in the expulsion of a negative particle from the neutral molecule. The residual portion of the molecule retains a positive charge, and is carried at once to the cathode in an electric field. This matter again undergoes chemical change, giving rise to the phenomena of excited radioactivity. The experimental data in favour of this view are best considered in the next section (§ 10) on the nature of the radiations.

It has been shown that the carriers of excited activity for both thorium and radium travel at about the same rate as the ions produced in the air by Röntgen rays. From data of the Kinetic Theory of Gases it has been shown that the ion in air is probably large compared with the molecule of oxygen or hydrogen. This has been explained by supposing that the ion, immediately after its production, becomes the centre of a cluster of molecules which move with it. On this view that part of the emanation molecule which retains a positive charge immediately becomes the nucleus of an aggregation of molecules

of the surrounding gas. The size of the cluster is probably about the same for the positive ion, since the size is mainly determined by the electric charge which is the same for both. The velocities in an electric field are thus the same for the carriers of excited activity and for the gaseous ion. Since the size of the cluster is large compared with the original nucleus, the velocities of the carriers of thorium and radium excited activity would be about the same, even if the original nuclei were of different masses.

§ 10. *Nature of the Radiations.*

In considering the question of the size of the body expelled from the molecule of the emanation, and of the nature of the radiation from the emanation, it is necessary to take into account the nature of the emanations emitted from all the known radioactive bodies; for there is no reason to suppose that the processes which are taking place in the molecule of the emanation are essentially different in character from those occurring in the other radioactive bodies. It is known that uranium, thorium, and radium emit two types of radiation. One type is not appreciably deviable by a magnetic or an electric field, and is very easily absorbed in matter. These will be called the α rays. The others are deviable and more penetrating in character, and will be called the β rays. In addition I have shown that thorium and radium emit some rays nondeviable in character, but of very great penetrating power. All of the radioactive substances including polonium as well as "excited" bodies and the emanations give out these α rays. Their power of ionizing the gas is very much greater than for the other types of rays emitted; and it is probable that the greater proportion of the energy radiated into the gas is in the form of α rays. The α rays from different radioactive substances, including the emanations of "excited" bodies, do not vary very much in penetrating power. The "excited" radiations for thorium and radium are the most penetrating in character and that of uranium the least.

It has been difficult to offer a satisfactory explanation of the nature of these rays. I have previously shown as untenable the view that they are secondary rays due to the emission of β rays. I have been recently led, by a mass of indirect evidence, to the view that the α rays are in reality charged bodies projected with great velocity. The ionizing effect of the rays is due to the collision of the projected body with the molecules of the gas, in the same way that the cathode rays ionize the gas in their path. Such a projected particle probably produces many thousand ions in its path before its velocity is

reduced to the point below which it can no longer ionize the gas. Strutt has put forward the view that the α rays were positively charged bodies since the β rays emitted from the same body carried a negative charge. This view has also been advanced in a recent paper by Sir W. Crookes.

The evidence in favour of the projection nature of the α rays is so far all indirect in character, and is briefly summarized below:—

(1) The absorption of the α rays in matter (like the β rays which we know are projected particles) is approximately proportional to the density of the material. It has been shown that the absorption of uranium, thorium, and radium rays is roughly proportional to the density for air and for aluminium.

(2) The absorption of the α rays by a given thickness of matter *increases* rapidly with the thickness traversed.

I have found that this is a general property of the α radiations not only for the radioactive elements proper, but for the radiations from the emanation and excited bodies. This is to be expected if the rays consist of projected particles, but is difficult to explain if the radiations are æther-waves similar to Röntgen rays.

(3) In the case of the emanations we have direct evidence that a negatively charged particle * is projected. The radiation from the emanation is due to these projected particles which ionize the gas in their path. This satisfactorily explains the experimental observation that the amount of excited activity is directly proportional to the amount of radiation from the emanation. It also serves to explain the fact especially noticeable in the experiments on the radium emanation, that some of the carriers of excited activity have sufficient initial velocity to move against the electric field. This velocity is due to the recoil consequent upon the projection of the charged body.

If these rays are due to projected charged particles they should possess the properties of the α rays of deflectability by a magnetic and electric field.

No deviation of the α rays has so far been detected in a strong magnetic field, but the experiments have not yet been

* I was at first inclined to suppose that the particle expelled from the emanation was a negative electron, since it is known that both thorium and radium compounds and bodies excited by them, emit some deviable rays. I have, however, made a close examination of the radiation from the emanation by the electrical method, but was unable to detect the presence of any penetrating deviable rays. If such deviable rays are present, they certainly exist in far less proportion compared with the α rays than in the other radioactive substances.

pushed to the necessary limit. The results, however, indicate that if the rays are deflectable, the deviation is minute compared with the β rays. This is to be expected if the mass of the expelled particle is large compared with the electron. If, for example, the projected body had a mass 10 times that of the hydrogen atom, it would require a magnetic field about 10,000 times as strong to produce the same deviation as for the electron moving with the same velocity. There is evidence that large carriers moving with a high velocity are produced in vacuum-tubes. W. Wien* has shown that the "*Canal Strahlen*" of Goldstein are positively charged particles moving with high velocity. These rays are deviated by a magnetic and electric field. When the vacuum-tube is filled with hydrogen the ratio of the charge to the mass, $\frac{e}{m}$, of these carriers is about 10^4 , showing that the carriers have the same mass as the hydrogen atom. In an atmosphere of oxygen the size of these carriers is considerably greater than the hydrogen atom.

It is possible that the electric charge on the expelled particle may be different for different radioactive bodies under different conditions. For the emanations of thorium and radium the expelled particles are for the most part negative. It has been shown that some of the radium carriers of excited activity have a negative charge, showing that the expelled body is positive. In addition, Dorn has shown that in a radium solution the excited radioactivity is produced on the anode, and not on the cathode. This shows that the carriers of excited activity in solution have a negative charge, so that the expelled body is positive in sign.

§ 11. *Evidences of Chemical Change.*

In previous papers by Mr. Soddy and myself, the view has been put forward that radioactivity is an accompaniment of continuous chemical change. Taking, for example, thorium, which has been worked out more thoroughly than the other radioactive bodies, it has been shown that a chemical substance Th.X is produced at a constant rate by the thorium compound. This Th.X undergoes further chemical change, one of the products of which is the emanation. This emanation itself is not stable, but expels from itself a negatively charged body. The positively charged portion of the emanation is carried to the electrodes, and this again undergoes further chemical change, giving rise to the phenomenon of excited radioactivity.

* Drude's *Annal.* No. 6, p. 244 (1902).

There is thus evidence of four distinct changes in each of which the matter produced has distinct chemical properties. For example, Th.X is soluble in ammonia, while thorium and products of the later changes are not. The emanation is not soluble in hydrochloric or sulphuric acid, unlike the matter responsible for excited radioactivity. There is strong evidence also that the chemical changes in the matter responsible for excited radioactivity are complex in character. It has been shown that the excited radiation in a body increases after removal when the body is exposed for a short time in the presence of the emanation. This effect is analogous to the increase of radioactivity of Th.X for the first day after separation, which has been shown to be due to the excited activity produced in the matter constituting the Th.X.

In order to account for the increase in radiating power after removal, one must suppose that the matter which is deposited from the thorium emanation gradually undergoes a chemical change. The transformed matter undergoes a secondary change, the time-rate of which is slower than the primary, but which gives rise to greater radioactivity. From data of § 5 half the matter has undergone change about 1 hour after deposit, while in the secondary change the corresponding time is about 11 hours.

Summary of Results.

(1) Excited radioactivity produced by thorium and radium compounds is due to the deposit of radioactive matter, which is derived from the emanation given out by these bodies.

(2) Excited radioactivity is transmitted by positively charged carriers, produced from the emanation, which travel in an electric field with about the same velocity as the positive ions produced in air by Röntgen rays. This velocity (about 1.3 cm. per sec. for 1 volt per cm.) is about the same for the carriers of thorium and radium excited activity.

(3) These positively charged carriers are due to the expulsion of a negatively charged body from the molecule of the emanation.

(4) Evidence is adduced for the view that the easily absorbed and apparently nondeviable rays of radioactive substance are due to the expulsion of charged bodies at a high velocity. The rays are thus analogous to the *Canal Strahlen* of Goldstein, which Wien has shown to be positively charged bodies projected at a great speed.

In the case of the emanations the expelled particles are for the most part negative in sign.

(5) In the case of radium about 5 per cent. of the carriers

of excited activity are distributed on the anode in a strong electric field.

(6) The excited radiations from thorium due to a short exposure in the presence of the emanation increase in the course of several hours after removal, to three or four times their initial value.

(7) The emanations and the matter which gives rise to excited activity are the result of a succession of chemical changes occurring in radioactive matter. In thorium there is evidence of at least four distinct chemical changes.

Macdonald Physics Building,
McGill University, Montreal.
July 29, 1902.

V. *Effect of Temperature on the Hysteresis Loss in Iron.*
By R. L. WILLS, B.A., A.R.C.Sc.I., 1851 Exhibition
Scholar, St. John's College, Cambridge*.

IT has been known for some years that for soft iron the magnetic hysteresis, for a given range of magnetic force, diminishes considerably as the temperature is raised. Kunz † found that the curve showing the variation of hysteresis with temperature for a specimen of soft iron was practically a straight line, while the corresponding curves obtained by Morris ‡ for a specimen of iron show that the fall of hysteresis is much more rapid as the critical temperature is approached than during the earlier stages of heating.

The present paper gives the results of experiments undertaken at the suggestion of Professor J. J. Thomson, on the effect of temperature upon the hysteresis in iron and a tungsten alloy for which the effect of temperature on the magnetic permeability has already been given §.

Method.

The method employed is that devised by Mr. G. F. C. Searle ||, the energy dissipated in hysteresis being readily determined by observing the "throw" produced in the movable coil of a sensitive electro-dynamometer. Most of the apparatus used was kindly furnished me by Mr. Searle.

The movable coil of the electro-dynamometer is connected in series with the secondary coil wound round the specimen,

* Communicated by Prof. J. J. Thomson.

† *Electrotechnische Zeitschrift*, 1894, p. 196.

‡ *Phil. Mag.* vol. xlv. pp. 213-254 (1897).

§ R. L. Wills, *Phil. Mag.* vol. i. pp. 1-21 (1900).

|| G. F. C. Searle, *Proc. Camb. Soc.* vol. ix. pp. 2-6 (1895).