

## VI. The behaviour of the phosphorus emanation in spherical condensers—V

C. Barus

**To cite this article:** C. Barus (1902) VI. The behaviour of the phosphorus emanation in spherical condensers—V , Philosophical Magazine Series 6, 3:13, 80-91, DOI: [10.1080/14786440209462739](https://doi.org/10.1080/14786440209462739)

**To link to this article:** <http://dx.doi.org/10.1080/14786440209462739>



Published online: 09 Jun 2010.



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VI. *The Behaviour of the Phosphorus Emanation in Spherical Condensers*\*.—V. By C. BARUS†.

1. **I**N the preceding papers it was assumed that the conditions could be so chosen (swift air-currents, highly active ionizer, &c.) that the decay of the ionization might be temporarily disregarded. Such an assumption is naturally precarious, and the following series of experiments are planned with particular reference to the factor ignored. Accordingly a closed spherical condenser was installed with its outer surface put to earth and its inner (concentric) surface, always very small, in contact with the charged electrometer. The intervening space was suitably ionized by a small piece of phosphorus, about as large as a pea, suspended at the centre. From such a condenser no ions can escape; it therefore offers greater theoretical simplicity than the plate-condenser, as discussed in an earlier paper, while the two discriminating variables, radius and voltage, may be changed at will. Practically, however, the experiments are very difficult, not only because of the baffling irregularity of the ionizer, but because it is not easy to lead a charge to or from the centre, without conduction.

As a whole, I think, the results may be interpreted as showing that decay due to the mutual destruction of ions is not in direct evidence, and may be considered negligible in investigations of the present order of precision. The enclosed air at a distance from the phosphorus rather behaves as though it contained more ions than reach it from the source.

2. A series of König's resonators seemed very suitable for the present purposes, since they were at hand in a large range of diameters, and fig. 1 shows the original adjustment. R is the brass resonator; B the curl of wire making the inner surface of the condenser, and supporting the piece of phosphorus P; C is an insulating glass tube 30 centims. long, through which the charge is conveyed by the wire *ab*, to be dissipated in the condenser. The tube C is grasped by the hard-rubber sheath F, fig. 2. B is thus in contact with the electrometer, and the capacity of the latter (about 60 cm.) is always large as compared with the condenser (negligible here and less than 1 cm. in the experiments below). D is a perforated copper plate closing the condenser and putting it to earth at E.

3. The early experiments made with this apparatus (Resonator, K 6;  $2R = 8.6$  cm.) seemed to indicate a linear

\* Cf. Am. Journ. Sci. [4] xi. p. 310 (1901).

† Communicated by the Author.—For previous communications, see Phil. Mag. [6] ii. pp. 477-488 and references there given.

relation of potential and time, at least within the first 7 minutes of discharge. This constant current was not appreciably modified by stopping the lower tubulure of the resonator with cotton, nor after vigorously airing it out; neither was there any marked change of current even after 30 minutes.

This interesting result was not, however, borne out by longer single periods of observation (20 minutes). Marked curvature was eventually in evidence, which did not seem referable to leakage errors. Moreover, the conduction decreased in succeeding curves, apparently indicating a diminution of the ionization due to the phosphorus. Possibly the activity of phosphorus may be quenched by its own emanation in the lapse of time, but the discrepancy is liable to be hygrometric.

4. A systematic series of experiments was now carried through with receivers ranging in diameter from  $2R=3$  to  $2R=23$  centims. The results were surprisingly irregular seeing that some pains was taken with the work, but they mapped out a graph of an exponential character to which different interpretations may be given. To repeat them here would carry me too far.

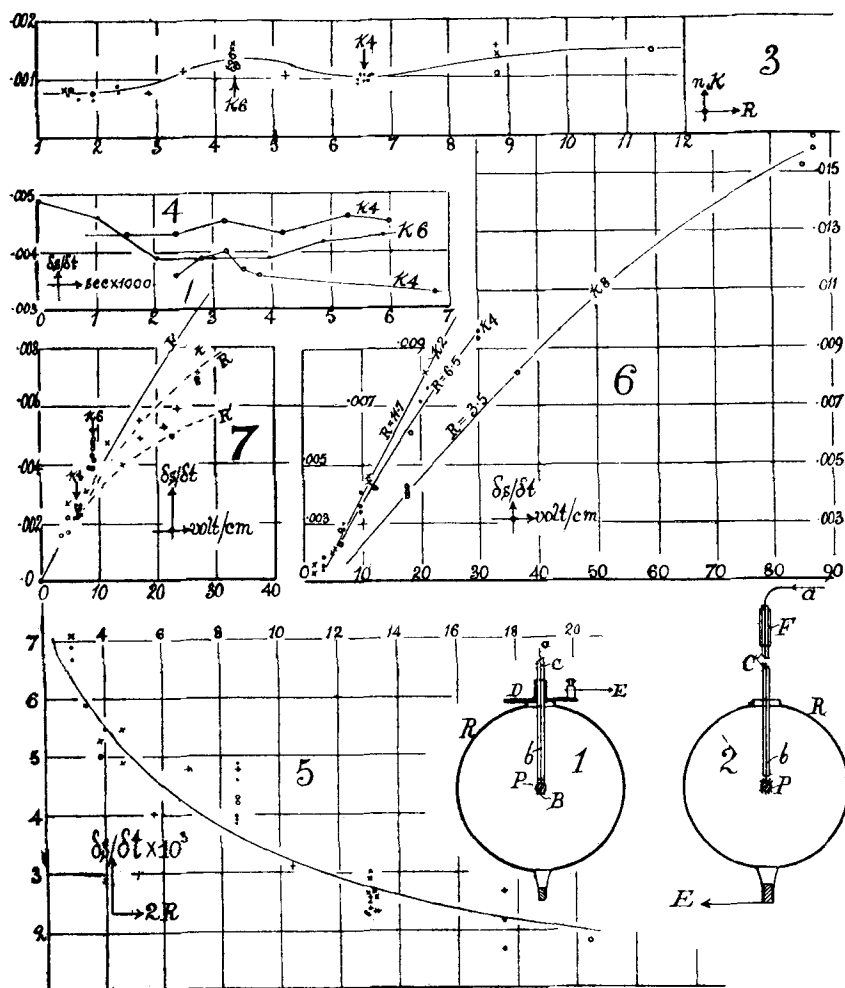
5. Before continuing the work it seemed advisable to make trials to secure more perfect insulation. No means were found for removing leakage in the apparatus perfectly (paraffin, rubber, &c. were tried in succession with no advantage); and a dry clean glass stem, C, fig. 1, held in place by a wide hard-rubber sheath F, seemed the best remedy.

Experiments were then tried to ascertain the difference between a condenser closed as above and one left open for the stem to pass through, as shown in fig. 2. Here the wire and plug attachment puts the outer surface to earth at E. The closed condenser always introduced the larger leakage. Care must be taken to avoid friction in manipulating the stem.

6. Conformably with the experience gained, the condenser in the following experiments is left open above, as seen in fig. 2, and is put to earth at E by a brass plug wedged into the neck. The glass tube, C, terminating in the ball of phosphorus P, is kept clean and dry, and extends to 30 centims. above the condenser. Unfortunately there is some escape of ions around the neck and the access of air here is a menace, but there seems to be no way out of the dilemma.

To estimate the loss of ionizing activity of the phosphorus pellicle one of the condensers (K 6 and K 4 respectively) was treated as a standard, and observations were made upon this

before and after the observations of each of the other condensers. Table I. is obtained in this way. The scheme adopted limited all observations to an interval of 5 minutes. Readings,  $s$ , at the electrometer were taken per minute, and



by dividing them into two parts the rates  $\delta s/\delta t$  were obtained linearly from data 3 minutes apart. The table also contains other constants to be discussed below. The first measurement is given in full as an example of the method, the others are abbreviated.

TABLE I.—Leakage of Spherical Condensers with a medium ionized by Phosphorus. Initial potential-difference,  $V_0=40$  volts. Electrometer deflexions,  $s$ .

No. Radius R. Field.	Time $t$ .	Deflex- ion $s$ .	$10^5 \times$ $n_1 K'$ .	No. Radius R. Field.	Time $t$ .	Deflex- ion $s$ .	$10^5 \times$ $n_1 K'$ .
K 6. 4.3 cm. 9.3 v./cm.	29m 0*	6.60	117	K 6. 4.3 cm. 9.3 v./cm.	61m—66m	...	119
	60	6.40	121				
	120	6.15	124	K 17. 1.95 cm. 20.5 v./cm.	69m—74m	...	73
	180	5.90	—				
	240	5.70	121				
	300	5.45					
K *. 1.5 cm. 27 v./cm.	37m—42m	...	79	K 6. 4.3 cm. 9.5 v./cm.	76m—81m	...	113
K 6. 4.3 cm. 9.3 v./cm.	45m—50m	...	122	K 2. 11.7 cm. 3.4 v./cm.	84m—89m	...	143
K 3. 8.8 cm. 4.5 v./cm.	53m—58m	...	102	K 6. 4.3 cm. 9.3 v./cm.	93m—98m	...	135

\* The following data are abbreviated by omitting the individual deflexions  $s$  and times  $t$ . Again  $2.3 n_2 K' = n_1 K$ .

Curiously, K6 showed a linear diminution of potential or constant current, while in other cases the currents usually fell off. Again, the values of  $\delta s/\delta t$  for K6 in the lapse of time, fig. 4, first show a decrease ( $\cdot 0049$  to  $\cdot 0039$ ), thereafter a constant variation, finally actually an increase ( $\cdot 0039$  to  $\cdot 0043$ ). Hence the factors which determine the activity of phosphorus are extremely complicated. Moreover, if the observations for the other condensers are corrected by the results for K6, by putting the current for the latter  $\cdot 0050$  throughout, the new values show worse agreement than the original values. It follows from this that the method of correction proposed is not available, and that the discrepancies to be eliminated are not regular but incidental fluctuations. Moreover, the data of this table are uniformly smaller, scarcely  $\frac{1}{2}$  or  $\frac{1}{3}$  of the values referred to in § 4. They are insufficiently definite to suggest the form of the locus. New observations, Table II., were therefore needed and were made in the same way as the results given in Table I. The condenser was slightly modified, consisting in this case of a sphere of wire gauze about 1 centim. in diameter, containing

a much larger piece of phosphorus than above. The larger condenser K 4 is now the standard, admitting of easier adjustment. The constancy of the ionizer is shown in fig. 4 (upper curve K 4), displaced in ordinate, for convenience.

TABLE II.—Leakage of Spherical Condensers with a medium ionized by Phosphorus. Initial potential-difference  $V_0=40$  volts.

No. Radius R. Field.	Time $t$ .	Deflex- ion s.	$10^5 \times$ $n_1 K'$ .	No. Radius R. Field.	Time $t$ .	Deflex- ion s.	$10^5 \times$ $n_1 K'$ .
K 4. 6.5 cm.	24m 0s	7.10	97	K 6. 4.3 cm.	61m—66m	...	138
	60	6.90					
	120	6.75		K 4. 6.5 cm.	68m—73m	...	94
	180	6.60					
	240	6.50					
	300	6.40					
K 3. 8.8 cm.	31m—36m	...	153	K 8. 3.45 cm.	78m—83m	...	110
K 4. 6.5 cm.	38m—43m	...	94	K 4. 6.5 cm.	86m—91m	...	103
K 5. 5.2 cm.	46m—51m	...	102	K 10. 2.85 cm.	93m—98m	...	72
K 4. 6.5 cm.	53m—58m	...	103	K 4. 6.5 cm.	104m—105m	...	100

In Table III. the ionizing activity of phosphorus is exhibited when the condenser, K 4, is left quite without interference. A remarkable rise and fall is as usual apparent, without discernable cause (fig. 4, lower curve). After this, the other condensers are again tested with the object of securing sufficient data to at least roughly suggest the form of the locus.

Both Tables II. and III. have been abbreviated; the results of interest admit of graphic representation, as will be explained in relation to figs. 3 and 5.

7. In view of the fact that the ionizing potency of phosphorus sometimes increases and at other times decreases, as it were incidentally and subject to arbitrary conditions\* not made out, I have in fig. 5 summarized the uncorrected values of the initial currents,  $\delta s/\delta t$ , preferably to the corrected

\* Work still in progress has shown me that the variability of the phosphorus ionizer is related to the hygrometric conditions under which the experiments are made. Temperature is of secondary importance.

values. The former, as has been stated, usually show greater regularity than the latter, upon which the fluctuations of the standard condensers have been imposed. The diameters,  $2R$ , are in centimetres.

TABLE III.—Leakage of Spherical Condensers with a medium ionized by Phosphorus. Initial potential-difference  $V_0=40$  volts.

No. Radius R. Field.	Time $t$ .	$10^5 \times$ $n_1 K'$ .	No. Radius R. Field.	Time $t$ .	$10^5 \times$ $n_1 K'$ .
K 4. 6.5 cm.	40m—45m	69	K 14. 2.35 cm.	75m—80m	74
K 4.	46m—51m	109	K 14. 2.35 cm.	91m—96m	87
K 4.	53m—58m	113	K 17. 1.95 cm.	99m—104m	64
K 4.	58m—63m	103	K 20. 1.7 cm.	107m—112	65
K 4.	64m—69m	99	K. 1.5 cm.	115m—120m	71
			K 4. 6.5 cm.	122m—127m	89

It appears at once that the data as a whole, though investigated with care, still fail to lend themselves for the nice discernment of the nature of the locus in a relation of current to the diameter of the condenser. The results, even of a single series, are not smooth. Indeed the exceptional positions of the currents for the standard condensers, those of K 6 ( $2R=8.7$  cm.) being abnormally high, while those of K 4 ( $2R=13.1$  cm.) are low, is perplexing, and has led me to suppose that some occult cause of variation has been left undiscovered. One is almost tempted to infer that each condenser behaves as an individual, a conclusion for which I am unable to discover adequate reasons. The curve, which has been put through the observations, was computed from  $(dv/dt)(R+a)=A$ , for reasons presently to be explained. The observations are in accord with it, in so far as they show an increase of current at an accelerated rate as diameter decreases.

8. *Working Hypothesis.*—The attempt must now be made to derive some theoretical conclusions from the experiments

detailed in the above paragraphs. As before, let  $n$  be the number of particles per cubic centim., so that  $n$  is the concentration or density of distribution of the phosphorus emanation. Let  $k$  be the "absorption" velocity of the ion, treated in the first instance as independent of the potential and of the concentration gradients. Let  $k'$  be the coefficient of decay, so that  $k'n^2$  is the number of ions vanishing per cubic centim. per second. Finally, let  $R$  be the external radius of the condenser and  $C$  its effective capacity including that of the electrometer.

With regard to the electrical currents, let  $V$  be the potential at a distance  $r$  from the centre of the condenser whose external face is put to earth. Let  $U$  be the aggregate velocity of the ions in the unit electric field and  $e$  the charge of each.

In all cases the observations are made when the flux is stationary, so that  $dn/dt=0$  throughout, for any shell. Moreover, as shown elsewhere, the effect of a potential gradient is but a negligible contribution to the number of ions which are absorbed by the outer surface of the condenser.

To begin with the simplest cases:—If the motion of the ion is entirely independent of  $dV/dr$  and  $n$ , the accumulation in an elementary shell at a distance  $r$  from the centre will be  $4\pi k d(r^2 n)/dr \cdot dr$ , per second; the decay per second,  $k'n^2 4\pi r^2 dr$ . Hence

$$d(r^2 n)/dr = (k'/k)n^2 r^2,$$

or if  $A$  is a constant,

$$1/n = r((k'/k) + Ar).$$

In the absence of decay,  $1/A = nr^2$  so that  $A$  is the reciprocal of the concentration,  $n_1$ , at a distance 1 from the centre. If conduction were prompted solely by the ions which reach the external shell kept at  $V=0$ , since the charge in this shell is per centim.

$$edR/(R(k'/k) + AR)$$

and its time of discharge  $dR/k$ ,

$$CdV/dt = 4\pi keR/((k'/k) + AR).$$

In the absence of decay,  $k'=0$ , and

$$dV/dt = 4\pi ken_1/C,$$

where  $n_1$  as stated holds for  $r=1$ . This case, in which  $dV/dt = ds/dt = \text{const.}$ , independent of the radius of the condenser is effectually excluded by the observations given in fig. 5. If  $k'$  is not zero,

$$dV/dt = (4\pi ke/C)(1/(k'/kR + A)),$$



so that the current increases with  $R$ , which is not admissible. Neither of these cases, moreover, would be open to computation directly, and they are thus without immediate interest.

This may be treated in a slightly different manner, however, by supposing the number,  $n$ , just found to be correctly evaluated and then introducing the ion velocity,  $U$ , instead of the absorption velocity,  $k$ , in the usual way, and they then become suggestive. If  $1/A$  is replaced by  $n_1$ , the number of ions per cubic centimetre at a distance of 1 centim. from the centre, the above concentration  $n$  at a distance  $r$  may be written,

$$n_1/n = r((k'/k)n_1(1-r) + r).$$

If decay is ignored,  $n = n_1/r^2$ , as is otherwise clear, is independent of  $k$  also.

Now if the electric conduction is determined by the number of ions which reach the external shell ( $r=R$ ),

$$-dQ/dt = -CdV/dt = 4\pi R^2 U (V/R) ne.$$

It is understood that this number is not appreciably modified by the occurrence of the field so that when decay is absent ( $k'=0$ ),  $n = n_1/R^2$ , as above deduced. Hence

$$-(dV/dt)/V = -d(\log V)/dt = 4\pi e U n_1 / CR.$$

Here the first member is equivalent to  $-d(\log s)/dt$ ,  $S$  being the deflexion of the electrometer, and is obtainable from the observations directly,  $4\pi e U / C$  is a constant,  $n_1$  expresses the waning intensity of the phosphoric source of ionization, and  $R$  is the external radius of the condenser. The equation therefore admits of being tested. The integral of the equation found for the potential gradient becomes

$$V = V_0 e^{-(4\pi e U n_1 / CR)t},$$

which is compatible with the data of tables 1-3. In these tables I have therefore inserted data for

$$n_1 K = (4\pi e U / C) \cdot n_1,$$

obtained from

$$-(dV/dt)/V \cdot R$$

for each case. To facilitate computation,  $n_1 K$  is left in common logarithms and written  $n_1 k'$ , so that

$$2.3 n_1 k' = n_1 K.$$

9. *Comparison of data.*—The values so found, *i. e.*,

$$n_1 K' = R d(\log V)/dt,$$

are shown graphically in fig. 3, as ordinates in terms of  $R$  as abscissas. The curve here is apparently sinuous, due to the abnormally high values of  $K_6$  and the abnormally low values of  $K_4$ , alluded to, both of which remain unexplained. In the absence of these there would be a rise of  $n_1K'$ , of a gradual character with increasing radius. Since in  $n_1K$ ,  $K$  is constant, this means that relatively more ions,  $n_1$ , are available at the larger radii of the condensers, corresponding to weaker fields, than for smaller radii and correspondingly stronger fields. But as there is no reason for excluding  $K_4$  and  $K_6$ , and no suggestion for the occurrence of the sinuous curve obtained,  $n_1K$  must be regarded as increasing rapidly from the values for condensers of small radii,  $r=2$  centim., but reaching a practically constant result after the radius 4 centim. has been surpassed. On the whole, therefore, the data so far as investigation has been possible agree with the remarks made in § 1, that the evidence in case of dilution is rather in favour of an increased number of ions and that an occurrence of decay is not manifest. This means more generally that whereas in the saturated emanation the ions are produced at the same rate at which they decay so that  $n$  is constant, in the diluted emanation at a distance from the centre ( $n=n_1/r^2$ ), the production is in excess of the decay and conduction relatively too great.

Another method of treating  $n_1K$ , is to refer it to strength of field. This, however, may be done more advantageously after the data of the next section, in which  $R$  is constant and  $V$  variable, have been similarly brought forward.

10. One important question as to the availability of phosphorus as an ion producer, is the intensity of its action or the number of ions produced per second in limiting cases. To make an estimate of this quality, it suffices to pass a current through the condenser for gradually increasing potential-differences, between the faces, in order to ascertain to what degree the phenomenon fails to obey Ohm's law.

Results to this end were obtained with the spherical condenser,  $K_4$ , diameter 13.0 centims., the internal surface (a copper gauze bag carrying phosphorus) being about 1 centim. in diameter. Potential-differences,  $V$ , as high as 200 volts were applied, the external surface being put to earth. The current is as usual found from equidistant observations separated by an interval of 3 minutes. A few test experiments were made by repeating the earlier measurements for 21 and 63 volts, but it was thought wisest not to attempt to correct the data for the fluctuating ionizing activity of phosphorus in the lapse of time, nor to assume curvature in

the initial ( $t=0$ , nearly) contours of the curves. It will be sufficient to report the results graphically.

The chart figure, 6, gives the relation of the current ( $\delta s/\delta t$ ), arbitrarily in scale parts, to the potential-difference  $V$  in volts per centim., or strength of the field. The graph is obviously curved so that Ohm's law is departed from, but the curvature is small, indicating a limit as yet a great way off.

These results are now to be compared with similar data for condensers larger and smaller, the diameters chosen being  $2R=6.9$  centims. for K 8 and  $2R=23.3$  centims. for K 2, respectively.

The currents here obtained are also graphically reproduced in the chart figure 6, where the abscissas are the fields in volt./cm., and ordinates are the currents in the usual arbitrary measure. The relative conductivity of the three condensers is maintained, and the saturated states (maxima of the curves) are in all cases enormously distant. The relative curvatures of the three graphs in the same field cannot be made out.

11. *Observations for constant  $V_0$  and for constant  $R$  compared.*—I shall now endeavour to compare the data of § 6 and § 10, by referring them either to the same radii or to the same electric fields. The latter method is preferable not only as yielding a greater range of data, but because the values of the currents,  $\delta s/\delta t$ , and of  $n_1K$ , have not been regarded in this light. To begin with the former, figure 7 contains the values of  $\delta s/\delta t$  varying with the fields as taken from the Tables I., II., III., in which  $V_0/R$  varies by reason of varying  $R$ . These data though vague, eventually lie within the limits marked by the curves  $R$  and  $R'$ . In the same chart I have inserted the curve  $F$ , taken from fig. 6, in which  $V_0/R$  varies by reason of varying  $V_0$ . It is the curve for K 2 for which  $R=11.7$  is largest, and the data surest. The point of importance is clear at once: as the fields grow stronger the curve  $F$  lies quite above the curves  $R$ . If therefore high fields are produced by diminishing the radius of the condenser, the currents may be upwards of 20 per cent. or 30 per cent. too small, both because of the escape of ions around the stem and of the access of air. Indeed this state of things is not unexpected, inasmuch as the chief object of the investigation with spherical condensers was the avoidance of such losses of ions as occur in plate-condensers. Returning for additional consideration to fig. 3, it appears that the definitely low data corresponding to the radii 2 and 3 centimetres, are erroneously much too low, whence it follows that the probability of a constant  $n_1K$  is enhanced.

12. *Conclusion.*—Contrary to my expectation and in spite

of the labour spent upon them, the results for spherical condensers have not enabled me to give a decisive answer to the question at issue. The difficulty encountered and which occurs here in accentuated form, is the same which has hampered me throughout the present research, namely the elusive variability of the ionizer. Moreover, as the conditions determining it exist immediately at the surface of the active phosphorus, I do not see how they are to be put under control; merely keeping the air around the phosphorus at constant temperature, &c., is not a sufficient check on the behaviour of the surface itself.

In several respects, nevertheless, definite advances have been made. It has been shown that the best results can be reached with large spherical condensers (say 20 centims., or more, in diameter), in which changes of field are produced by applying larger potential-differences while the apparatus itself is left quite without interference.

Finally, to ascertain in how far the present experiments agree as a whole with the results for plate-condensers, where a theoretically different method is involved, it suffices to compute the value of  $n_1$ , the number of particles per cubic centim. at 1 centim. from the centre of the condenser. Since  $n_1 K'$  in fig. 3 is of the order of  $\cdot 0012$ , or when referred to natural logarithms  $n_1 K = \cdot 0026$ , and since  $n_1 K = 4\pi e U/C$ , if we insert J. J. Thomson's value of  $e = 2\cdot 3/10^{19}$  and put  $U$  of the order of 1 cm./sec.,  $C = 6\cdot 7/10^{11}$  farads, it follows that  $n_1 = 6\cdot 5 \times 10^4$ . The result agrees very well with the datum  $n_0 = 5\cdot 5 \times 10^4$ , as determined from plate-condensers by the totally different method there pursued. If in both cases  $U = 1\cdot 5$  cm./sec. be assumed, the numbers will be  $n_1 = 4\cdot 3 \times 10^4$  and  $n_0 = 3\cdot 6 \times 10^4$ , respectively. In either case the saturation is that existing at the surface of the phosphorus. They thus agree with the datum for the unsaturated emanation  $n = 2 \times 10^4$ , obtained by a third method from tubular condensers. Finally, all are in accord with J. J. Thomson's value for ionized air  $n = 4 \times 10^4$ , resulting from a method theoretically different from that of my papers.

13. *Summary.*—With the present paper I have given a brief but systematic account of the bulk of my work with the phosphorus emanation, the purpose throughout being to map out roughly the phenomena in which I am interested, as a preliminary to the more rigorous study of the subject which I am now beginning. The results as a whole are eventually to be tributary to an investigation on the colours of cloudy condensation, particularly in those regions within which Lord Rayleigh's theory must, in any case, cease to be applicable.

In endeavouring to account for the data obtained it was my endeavour to follow the established theory ; but I fear that in the explanations given I have little by little made a serious departure. If I had obtained but a single coincidence the result would not have deserved record ; but after finding data of a correct order of values in all the experiments, in spite of the variations of method, I have ventured to think that more than a coincidence is in question. The theory which provisionally underlies the series of papers is substantially this :—

(1) From experiments made in the absence of an electrical field I inferred that the nucleus has its own specific velocity and that this velocity is identical with the mutual velocity of the corresponding ions in the unit electrical field (volt/centim.).

(2) The nucleus produced by phosphorus is larger than the air molecule ; and both for this reason and from the fact that it receives promiscuous bombardment of molecules simultaneously, its velocity is of the low order stated, being (say) less than  $1/300$  that of the air molecule.

(3) It is not necessary to assume that the nucleus decays or vanishes within the ionized medium ; the evidence is rather in favour of a number of nuclei somewhat larger as the dilution increases.

(4) Whenever the nucleus comes in contact with a barrier (solid or liquid) it is absorbed or broken up. If the nucleus is ionized as in the case of the phosphorus emanation, the absorption is accompanied with the discharge of an electron.

(5) The observations with plate-condensers, tubular condensers, and spherical condensers are satisfied by supposing the nuclear velocity  $k$  to be independent of the concentration or degree of saturation.

(6) The nuclear velocity is independent of the potential gradient. Instead of putting  $U = V/R$  for the velocity of the ion along the potential gradient  $V/R$ , I have considered this velocity a constant, independent of  $V/R$  ; while the number of nuclei capable of discharging the metallic terminal of a field varies as  $V/R$ .

(7) If for  $e$  the charge of a nucleus, J. J. Thomson's electron be taken ; if  $U = k$ , then the number of nuclei\* in the saturated phosphorus emanation agrees with J. J. Thomson's value for the number of ions in ionized air.

Brown University, Providence, U.S.A.

\* I have since found that the lower values found from tubular condensers are referable to the inherent non-saturation. By using an excess of freshly cut phosphorus, the number of ions could be nearly doubled, thus putting these results in accord with the correlative data for plate- and for spherical condensers.