ON THE DISTRIBUTION IN TIME OF THE SCINTILLATIONS PRODUCED BY THE α-PARTICLES FROM A WEAK SOURCE

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Abstract

The course of radioactive transformation, which in all known cases takes place in strict conformity with an exponential law of decay, is most satisfactorily explained on the supposition that individual atoms disintegrate in complete independence one of another, so that the disintegrations throughout an extended source constitute a perfectly random series of events. Many observations favor this conclusion, but experiments have recently been described showing that at high concentrations (Kutzner) and also at very low concentrations (preliminary work of Pokrowski) departures may occur. An experiment is here described in which the record of more than 10,000 scintillations produced by the α-particles from a weak source of polonium has been obtained and analysed. A large solid angle was effective, as in Pokrowski's experiments. The various corrections to be applied to the immediate data are discussed: in the present case they were of no importance. There was no evidence to show that the Marsden-Barratt distribution formula was not completely valid under the conditions obtaining.

An investigation was also made of the effect of intense γ radiation on the rate of disintegration in a weak source (Pokrowski), with entirely negative results.

Introduction

A SERIES of exactly similar point-events separated from one another in time but limited to a prescribed small region of space is defined as a "random" sequence when the probability of occurrence of one such event is the same for equal small intervals of time irrespective of the temporal locations of those intervals. For series comprising point-events separated in space as well as in time, or of line-events having a common focus whilst being separated in time, a further condition must be satisfied if the events are to be considered as taking place "completely at random." In the former case, in any given interval of time, the probability of occurrence of a single event must be the same for equal small elements of volume, irrespective of the spatial locations of such volume elements—and, in the latter, equal probablities must characterise equal small elements of solid angle about the focus no matter what the directions of the axes of the elementary cones considered.

Radioactive disintegration in an extended source of active material may be regarded as a phenomenon involving line-events originating in foci themselves distributed in space. Now, macroscopically examined, all such cases have been found to exemplify a common transformation law and the customary interpretation of this fact has been based on the assumption that,

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in as far as concerns its time relationships, the ultimate atomic process occurs entirely at random. If this be assumed then the spatial distribution of disintegrating atoms in an extended source must also be a random one—and the observed distribution of directions of particle (or quantum) emission likewise random, for here, whilst preferential directions with respect to natural atomic axes are not excluded, in any practical case the random distribution of atomic axes in space would completely mask such regularities if in fact they were present.

Whilst this is the logical conclusion to be drawn from the usual interpretation of exponential decay, where the analysis is carried down to the behavior of individual atoms, it has been variously maintained that the phenomena observed on the macroscopic scale require no further "explanation" than that the time-distribution of disintegrations in a radioactive source, considered as unit, shall be a random one. From this point of view there remains the possibility that the complete randomness which we have considered may be found lacking under a more penetrating analysis. However unsatisfactory such a contention may appear, from the physical standpoint—and whether or not we hold the view that it is even permissible when the whole range of macroscopic experiments is considered[1]—the fact remains that a number of careful experiments has been made and that some of them point to departures from a time-randomness under certain conditions of observation.

For the type of statistical analysis here involved two methods are available, based upon the Bateman[2] and the Marsden-Barratt[3] formulae respectively. In either case a graphical means may be employed in the final comparison of experiment with theory, and, when this is done, conclusions appear more obviously in the latter case, since theoretically a linear function is predicted. But the Bateman type of analysis possesses the compensating advantage that a numerical criterion is afforded of goodness of fit by the dispersion coefficient which may be calculated from the experimental data themselves. Both methods have been used in practice. Kutzner[4] has made a very thorough analysis of the data afforded by three series of observations automatically recorded under as nearly as possible identical conditions of operation of an electrical point counter. The sole difference between the series (which had reference to 7761, 6907 and 5923 particles, respectively) lay in the surface concentration of polonium in the radioactive source employed. Here there was a factor of 25 times as between conditions in the first and third series, the actual concentrations being roughly 2×10^{-4}, 8×10^{-4} and 5×10^{-3} equivalent milligrams per cm^2, respectively. With the weakest source no appreciable deviation from perfect time-randomness

[1] In the writer's opinion the only evidence which the macroscopic experiments fail to supply is evidence for the random distribution in space of the disintegrating atoms in a given source. The exponential law of decay may be deduced alike from measurements made on the radiation emitted within a small or a large solid angle about such a source.

[2] Bateman, Phil. Mag. **20,** 704 (1910).

[3] Marsden and Barratt, Proc. Phys. Soc. Lond. **23,** 367 (1911).

[4] Kutzner, Zeits. f. Physik **44,** 655 (1927).

was found, but, with the other two, deviations quite outside the limits of probable error were set in evidence. In each case dispersion was subnormal (Bateman analysis), there being a marked lack of the longer time intervals between observed disintegrations (Marsden-Barratt analysis). The extent of the discrepancy increased with the surface concentration in the source. Whilst leaving the full interpretation until fresh data had been obtained using large solid angles (only about 1/400 of the total emission was originally employed), Kutzner concluded that his results were in agreement with those of Curie,[5] who found complete accord between experiment and theory with polonium sources of (apparently) very small concentration. The experimental material in this latter case consisted of 9974 measured time intervals belonging to four distinct series of measurements.

Recently Pokrowski[6] has published an account of preliminary observations with very weak sources and large solid angles. The concentration employed was about 10^{-7} equivalent milligrams per cm^2. It is not stated what material was used. Data having reference to 1633 time intervals, analysed by the method of Marsden and Barratt, showed an excess both of the very short and the very long intervals and no real agreement with theory over any appreciable range. In the discussion it was pointed out that new phenomena at small concentrations might be expected if at the higher concentrations mutual action between neighboring nuclei was possible. On account of the fundamental nature of such a suggestion, and because a suitable source of polonium of small concentration was readily available, it was thought worthwhile to make a series of observations parallel to those of Pokrowski, using the scintillation method of registration as he had done. Moreover, the writer did not feel compelled to make the present investigation exhaustive—covering a wide range of surface concentrations—most especially because it is explicitly stated by Pokrowski that his report is merely preliminary in scope. Rather is it intended to put forward this restricted series of results, only, as independent evidence, for what it is worth.

Experimental Method and Results

As a source of α-particles for scintillation experiments polonium is in many respects most suitable: it is not accompanied by appreciable β or γ radiation, and its period is sufficiently long for the decay generally to be negligible. However, when experiments extending over a number of weeks are in question, the latter condition no longer holds[7] and the desired constancy can be retained only by employing sources of radium D, E and F in equilibrium. Now the writer possessed some pieces of gold leaf exposed to radon in May 1929.[8] In January 1930 calculation showed that their α-

[5] Curie, J. Physique **1**, 12 (1920).

[6] Pokrowski, Zeits. f. Physik **58**, 706 (1929).

[7] The type of "uncorrected" distribution of time intervals which would be obtained in the Marsden-Barratt analysis of data referring to a source of varying activity is one in which very short and very long intervals are each in excess of the number calculated theoretically for a constant source of the same average activity.

[8] See Feather, Proc. Camb. Phil. Soc. **25**, 522 (1929).

particle activity was still increasing—but sufficiently slowly for this material to be considered a constant source of α-particles for the present type of experiment over a period of two or three weeks.[9] It was decided, therefore, to employ it in the work to be described. By placing the gold leaf directly in contact with the zinc sulphide screen a solid angle of approximately 2π radians could simply be attained. Preliminary tests showed, however, that the activity of the source was 5 or 10 times too great for convenient counting. A reduction was made as follows: one surface of a flat piece of glass was very lightly ground with fine emery. A small piece of the activated gold leaf was placed upon the ground surface and, by means of the smooth rounded end of a glass rod, used as a small pestle, was gradually ground to an extremely fine deposit of gold dust which adhered to the roughened surface. The zinc sulphide screen, supported on a microscope cover glass, was then fastened in position in contact with the ground surface. On the free upper surface of the cover glass was fastened a piece of thin aluminum foil, pierced with a hole of approximately 1 mm^2 area, which served to limit the portion of screen under observation. The microscope used was built up of a Watson's Holoscopic objective, of numerical aperture 0.45 and focal length 16 mm, and a low-power eyepiece, giving a field of view of about 8 mm^2 area. The source system was supported so that the hole in the aluminum foil was central in the field of view and, during periods of observation, a dim red light produced a faint illumination of the foil, leaving the central portion of the field relatively dark.[10] The mean interval between scintillations was somewhat greater than 3 sec.[11]

The record itself was made with the aid of a drum chronograph with magnetically controlled pen, the drum being driven at a uniform rate by the combination of falling weight and centrifugal governor. The uniformity of rate was tested from time to time and found altogether satisfactory. Throughout the presentation of the results, for sake of simplicity, time intervals have been replaced by linear intervals; the conversion factor from the one to the other is 0.921 cm/sec.

Observations were spread over thirteen consecutive days; on each day two or three "sessions"— 30 "sessions" in all. During each session "periods of observation" alternated with periods of rest, each, on the average, about a minute and a half in length: in all, 334 "periods of observation." During each period about 30 scintillations were recorded; altogether 10134 "intervals" between scintillations were measured. In the treatment of the records periods were measured to the nearest millimeter and the number of intervals in each period was counted. From this material the mean interval, ω, was determined. Intervals were measured in millimeters and entered in a statistic according to their length. In the first analysis grouping was in millimeter ranges, the elementary group containing those intervals of which

[9] A more complete statement on this question is reserved for the discussion.

[10] Cf. Chariton and Lea, Proc. Roy. Soc. **122A**, 304 (1929).

[11] From the figures given by Pokrowski it would appear that his field of view had an area of about 70 mm^2 and that the mean interval in his experiments was roughly 0.8 sec.

the length lay between x and $x+1$ mm. Later these groups were combined in sets of three and values of n_x obtained, where n_x now represents the number of intervals of length between $x-1.5$ and $x+1.5$ mm. Values of N_ξ followed immediately from this series of numbers; here N_ξ is the number of intervals of length greater than ξ mm. Now the formula of Marsden and Barratt may be written

$$N_\xi = N_0 e^{-\xi/\omega} \quad (1)$$

or

$$\log_{10} N_\xi = \log_{10} N_0 - (0.4343/\omega)\xi. \quad (2)$$

In our case $N_0 = 10134$ and $\omega = 31.28$ mm. Thus the theoretical distribution leads to the straight line

$$\log_{10} N_\xi = 4.0058 - 0.01388\xi. \quad (3)$$

This line is plotted together with the experimental points in Fig. 1. In Fig. 2 values of $\log_{10} n_x$ are plotted (A).

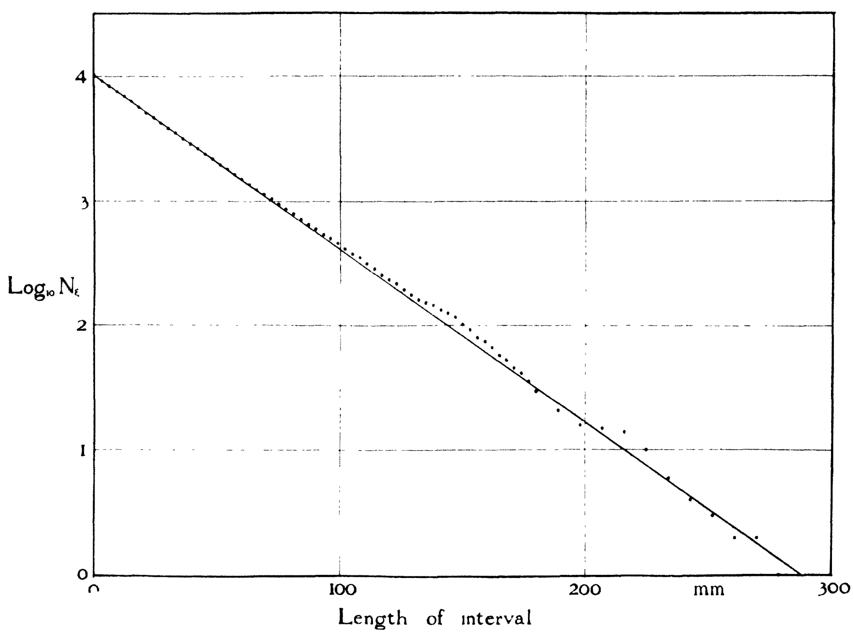

Fig. 1.

To a first approximation

$$n_x = -3(dN_\xi/d\xi)_x. \quad (4)$$

If this relation were strictly correct it is obvious from (1) that the graph of $\log_{10} n_x$ would be a straight line of the same slope as (3). A simple calculation shows that in fact a small correcting term ϵ is involved, depending on the ratio of the "plotting interval" (in this case 3 mm) to the mean length ω (here 31.28 mm), in such a way that

$$n_x = -3(1+\epsilon)(dN_\xi/d\xi)_x. \quad (5)$$

The condition of parallelism therefore remains, but the line as a whole is shifted slightly. Introducing numerical values we have

$$\log_{10} n_x = 2.9875 - 0.01388x. \tag{6}$$

This line is included in Fig. 2 for comparison with the experimental results. The straight line B, Fig. 2, corresponds to a plotting interval of 30 mm. Its equation, deduced in like manner to the above, is

$$\log_{10} \nu_x = 4.0039 - 0.01388x. \tag{7}$$

The corresponding points have been obtained by grouping the values of n_x in sets of ten—and the results are presented in this form, also, chiefly in order that they may be more nearly comparable with those of Kutzner, already referred to.

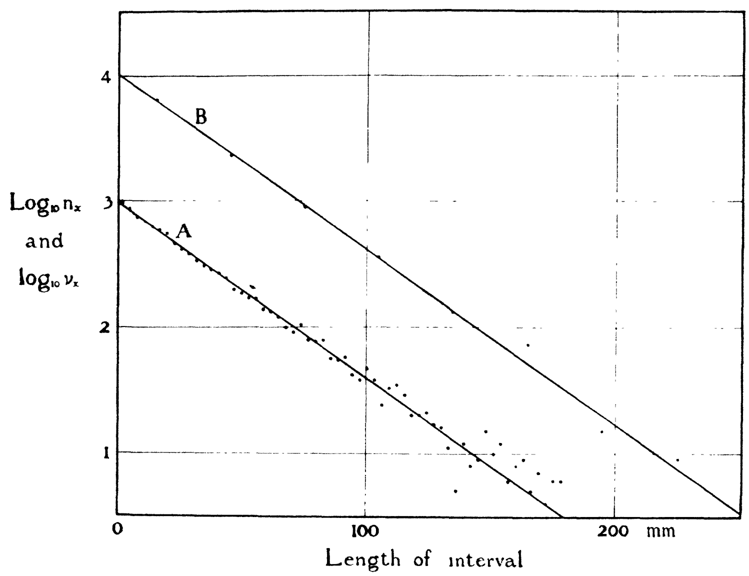

Fig. 2.

Discussion

Any consideration of the results here set forth must be twofold in character: there is the comparison of experiment with theory and, in addition, that of the present experimental results with those of previous investigators. But, before either comparison is made, it is necessary to consider what corrections, if any, should be applied to the crude observational material which the experiment provides—for the points plotted in Figs. 1 and 2 have been deduced without any corrections at all from the measurements of length on the chronograph record. First there is the varying activity of the source, which is entirely apart from any question of imperfection in the methods of registration. Depending upon the extent of this variation the theoretical logarithmic plot will depart more or less from the ideal straight line. At the time of the observations the strength of the source was increasing daily by about 0.205 percent. Columns 1 and 2 of the accompanying table show how observations were spread over the thirty sessions on the thirteen days during which the experiment was in progress. They indicate that the mean interval ω, deduced from the full material, corresponds most probably to the true mean interval on the eighth day. Column 4 gives the true mean interval corresponding to each successive day calculated on this basis and column 3 the observed mean values obtained from the daily observations. Column 5 will be referred to at a later stage in the discussion. With the material which the table provides we may proceed to calculate the theoretical form of $\log_{10} N_\xi$ in the present experiments, which in the ideal case of the constant source was represented by Eq. (3). For each day's results the theoretical distribution is calculated from Eq. (1) using the

TABLE I.

1 Day	2 Number of intervals	3 Mean interval mm	4 True mean interval mm	5 Probable error mm	1 Day	2 Number of intervals	3 Mean interval mm	4 True mean interval mm	5 Probable error mm
1	381	31.7	31.73	1.6	8	373	30.2	31.28	1.6
						268	31.9		1.9
2	274	33.1	31.66	2.0		410	28.8		1.4*
	298	30.1		1.7					
					9	389	31.7	31.22	1.6
3	332	30.8	31.60	1.7		392	31.5		1.6
	191	32.5		2.4		428	29.4		1.4*
	261	32.6		2.0					
					10	452	27.5	31.15	1.3*
4	282	33.2	31.54	2.0		438	30.9		1.5
	212	31.2		2.1		407	31.1		1.5
5	104	27.9	31.47	2.7*	11	431	32.4	31.09	1.6
	137	39.5		3.4*		405	33.0		1.6*
	159	36.6		2.9*		421	31.1		1.5
6	352	30.4	31.41	1.6	12	469	29.2	31.02	1.3*
						439	29.7		1.4
7	347	33.8	31.34	1.8*					
	309	35.1		2.0*	13	431	30.4	30.96	1.5
	342	31.6		1.7					

appropriate value of ω. The individual distributions are then combined. When this somewhat laborious procedure was carried through it became evident that the greatest departure from the straight line of Eq. (3) was so slight as not to be appreciable on a diagram drawn to the scale of Fig. 1.

The second correction concerns the failure of the method of registration when scintillations succeed one another in rapid succession. It is not that scintillations are missed, but the actual time intervals are distorted on the record obtained. The first point in Fig. 2 (A) has reference to intervals of apparent length less than 3 mm (i.e. intervals shorter than 0.32 sec.): 988 measured intervals were distributed as follows; 0–1 mm, 0; 1–2 mm, 404; 2–3 mm, 584. Assuming, to a first approximation, that the number 988 is the true number of intervals in the three sub-groups in question we should expect a distribution giving 0–1 mm, 340; 1–2 mm, 329; 2–3 mm, 319 intervals, respectively. Thus errors of registration have resulted in an apparent increase of length in the shortest intervals, and this must necessarily have been counterbalanced by an apparent decrease amongst the remainder. The figures quoted do not allow us to make any definite statement about the errors actually committed. At the one extreme is the supposition that in no case has the error been greater than 1 mm. On this basis we should assume that 340 intervals actually between 0 and 1 mm were recorded as between 1 and 2 mm in length, whilst 265 actually between 1 and 2 mm were recorded as between 2 and 3 mm long. At the other extreme is the supposition that all the intervals in the second and third groups in reality were correctly registered, the error lying entirely with the intervals of the first group. Then 75 members of that group could have been wrongly assigned to the second group and the remaining 265 members to the third. From the first point of view, amongst the total number of 10134 measured intervals (including the 988 shortest intervals with their newly assigned lengths) the correction of 605 1 mm errors of underestimation is necessary to rectify the crude distribution: from the second point of view the correction of 75 1 mm and 265 2 mm errors of underestimation is required. On account of the small fractions of the total which these numbers represent the two methods of correction lead to almost identical results. The former transfers 18.9 intervals from $n_{1.5}$ (originally 988), the second 19.4 intervals; these are effectively distributed amongst the various values n_x in gradually decreasing shares as x increases; thus $n_{4.5}$ is increased by 2.9, $n_{7.5}$ by 2.3 intervals, and so on. In Fig. 2(A) the change in position of the first point only is appreciable, as the figure indicates—and obviously to carry this correction to a further approximation is unnecessary.

From the above considerations, therefore, we may conclude that the experimental points of Figs. 1 and 2 and the straight lines accompanying them are entirely comparable: agreement between experiment and theory is to be measured by the closeness of fit of the points and the corresponding straight lines.

It must be admitted at the outset that the fit is good, though hardly as good as in the experiments of Curie.[5] On the other hand, by comparison with the results of Kutzner,[4] it will be seen that no such systematic deviations as he observed are in evidence here. It was not to be expected that they should be, since the concentration of active material in the source was smaller than the least concentration which Kutzner employed. Fig. 2 (B)—closely comparable with Figs. 2 (b), 3 (b) and 4 (b) in his paper—shows excellent agreement for the first five points and a non-systematic deviation later. Kutzner's figures likewise have five points closely on the theoretical straight line but, for his two stronger sources at least, the sixth and seventh points lie progressively below the line. Finally, making comparison with Pokrowski's results,[6] it is evident that no deviation of the order of magnitude of that reported by him is here present, though such deviation as there is in the earlier portion of Fig. 1 is certainly in the same sense as that which he observed. But the order of magnitude is entirely different. It is true that in the present experiments the surface activity of the source was roughly twenty times as great as in the experiments of Pokrowski, but it must be emphasised that this is a macroscopic comparison. When the phenomenon of aggregation is taken into account it is obvious, for instance, that the size of the mean aggregate does not increase as rapidly as does the mean surface density of deposit during a single activation process, since new centers of aggregation presumably appear continuously throughout the process. Now it is unquestionably the size of the aggregates which is of importance for theories of the mutual action of neighboring atoms. Moreover it is almost impossible to make any *a priori* statement concerning aggregation when the mode of activation is different in the two cases to be compared.

It was pointed out earlier in the discussion that variations in the activity of the source, if sufficiently great, would lead to deviations from the theoretical distribution of the type of which Fig. 1 offers very slight indication and Pokrowski's results very marked evidence indeed. It has already been demonstrated that such changes cannot account for a non-linear relation in the present experiments, but it must be borne in mind that the same effect would be shown with a constant source and a varying efficiency of registration. It is very probable that this explanation is likewise inadequate. Column 5 of Table I gives the probable error in the thirty observed values of the mean interval and asterisks mark those cases in which the latter value differs from the true value (column 4) by more than the probable error. It will be seen that there are ten such cases as against twenty cases of closer agreement than that which the limits of probable error represent. Thus we are dealing with a set of observations more than normally uniform, and it is therefore unlikely that variations in registration efficiency are here in point.

Finally, in order to make sure that simple errors in length measurement were not vitiating the results, the total period of observation was deduced in two ways; first by adding together the measured lengths of the 334 periods, secondly by adding together the measured lengths of the 10134 intervals. In the latter addition the supposition was made that each interval classed as between x and $x+1$ mm in length was actually $x+0.5$ mm long. The first addition led to the value 316988 mm, the second to 317032 mm—a difference of one part in 7200. Moreover, the second sum would be expected to be slightly the larger of the two on account of the simplifying assumption introduced.

A Further Experiment—The Effect of Intense γ Radiation

In the account already referred to Pokrowski[6] has described a very remarkable experiment in which the irradiation of a weak source of radioactive material with γ-rays was found to produce an increase of 45 percent in the frequency of the scintillations observed. X-ray irradiation was reported to produce a similar effect. It was suggested that the possibility of such an effect was intimately connected with the use of a very weak source, in which the hypothetical mutual action of neighboring atoms was too small to be effective. From this point of view the null result of earlier experiments with stronger sources was obviously explicable.[12]

There should, of course, be an intermediate case with sources a little stronger than Pokrowski's. The writer has carried out the analogous experiment employing a somewhat more concentrated source. To this end the arrangement of the previous investigation was modified in certain particulars; for instance it was no longer thought necessary to employ the maximum possible solid angle and it was recognised that it would be a great convenience to be able to remove the source from time to time. The latter was therefore carried on a rectangular glass plate (a fragment of the original gold leaf was attached to the smooth surface by means of a very thin film of castor oil) which when placed in its holder was maintained in a perfectly definite position, about 1.5 mm distant from the zinc sulphide screen. An area of about 2 mm² of screen was under observation. In the new arrangement the field illumination resulted in the screen being dimly lighted and the rest of the field relatively dark. This is an advantage, for, under sufficiently intense γ-ray irradiation, the screen becomes self-luminous and it is advisable to make conditions in the control experiment as nearly equivalent as possible.

The first comparison was made with a radon tube containing about 25 equivalent milligrams of emanation[13] placed on the axis of the system about

TABLE II.

1 mg	2	3 mm	4 mm	5
22.2	677	9.05	10.30	565
18.6	667	9.07	10.00	585
16.4	758	8.75	10.79	551
13.5	774	8.31	9.82	654
11.3	742	8.98	9.60	679
9.5	750	8.56	8.97	679
7.9	745	8.76	9.55	698
5.5	766	8.65	9.75	672
4.6	830	8.06	8.91	790
3.8	780	9.13	8.62	720
3.2	783	8.37	9.37	728
2.7	811	8.53	8.57	802
2.3	849	8.16	8.27	898
1.9	816	8.93	8.64	784
Totals	10748			9805

[12] When the present account was in the final stages of preparation Pokrowski's second paper (Zeits. f. Physik **59,** 427 (1930)) appeared in this country. In it a more detailed point of view is adopted and experimental data, obtained with a source of radium together with its subsequent products, put forward in its support. However, the experiments newly described differ in essential particulars from those of the writer, so that there is at present no basis of comparison. Nevertheless it may be remarked that if, as Pokrowski assumes, this process of induction is "saturated" in the ordinary case of a concentrated source, and if the mechanism is similar to that which he suggests, then the α-particles from such a source would not be homogeneous in velocity, but a distribution of velocities greater than the normal would also be present.

[13] The writer desires to thank Dr. C. F. Burnam of the Kelly Hospital, Baltimore, for providing him with this material.

3.7 cm distant from the source. The mean length of 1240 intervals recorded with the tube in position was 9.18 mm, 1181 intervals recorded when the tube was removed gave 9.33 mm. Within the probable error of the measurements, therefore, no difference was found. With this arrangement, however, the β and γ-ray luminosity of the screen was almost inappreciable and a further comparison was determined upon with more intense irradiation. The γ-ray tube was moved up in contact with the glass plate supporting the source.[14] Under these conditions the zinc sulphide screen fluoresced brightly and observations with the γ-ray tube in position[15] invariably led to greater values of the mean interval than those carried out when the tube had been removed. It was obvious that scintillations were being missed under the former conditions of observation. Table III summarises the comparisons made on successive days as the activity of the γ-ray tube decayed. A constant geometrical arrangement was used throughout. The strength of the γ-ray source is given in column 1. Columns 2 and 5 show the numbers of intervals

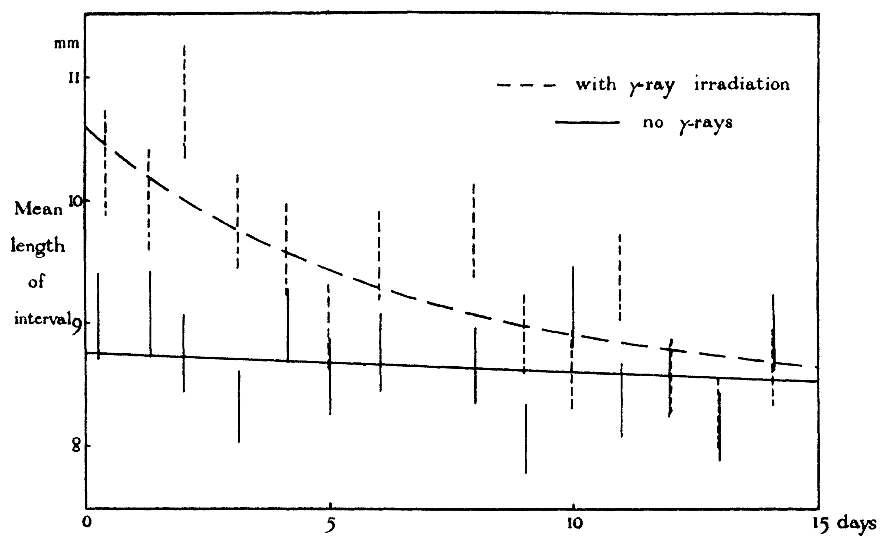

Fig. 3.

measured, in the first case with the γ-ray tube removed, in the second case with the tube in position. Columns 3 and 4 give the values of the mean interval deduced from these measurements. When the γ-ray activity had fallen to 4.6 equivalent milligrams the brightness of fluorescence which it caused was roughly the same[16] as the brightness of artificial illumination employed when the tube was absent; thereafter the very weak fluorescence with the tube in position was supplemented by a certain amount of artificial illumina-

[14] The thickness of the plate was 1.8 mm. The radon tube was contained in a brass case about 0.7 mm thick.

[15] The polonium source was exposed to the γ-rays only during the periods of observation, amounting to about half an hour daily.

[16] This comparison is necessarily inaccurate on account of the difference in quality of the light in the two cases.

tion also. In Fig. 3 the results are shown graphically. The lengths of the vertical lines indicate the magnitude of the probable error in each case. The gradual increase in the α-particle activity of the source during the period of the observations is clearly evident from the decrease in the mean interval determined with the source alone.[17] The broken line, which represents sufficiently well the trend of the apparent activity of the source under γ-ray irradiation, has been drawn in such a way that the difference between corresponding ordinates on this and on the full line, respectively, decreases according to the same law as the activity of the radon tube employed as the source of γ-rays. Whilst no great emphasis is laid on this way of exhibiting the results,[18] it does show at least that, as long as the effect is relatively slight the percentage of scintillations missed when observations are carried out in the presence of intense γ radiation is roughly proportional to the strength of that radiation.

Throughout the present experiments the observed effect of the irradiation of the source has been an apparent decrease in its α-particle activity. It was thought possible, however, that over a short period, during the decay of the γ-ray source employed, an apparent increase would be found. This might have been expected when the intensity of the residual primary β-particles and the secondary electrons produced by the γ-rays was just of the right order of magnitude to give rise to the type of multiple-β-particle scintillations described by Chariton and Lea.[19] Probably the requisite β-particle intensity was reached sometime between the tenth and the fifteenth day, but it is very doubtful indeed whether any trace of this phenomenon has been observed in the present case. Chariton and Lea, observing with a microscope of numerical aperture 0.65, classify this type of scintillation as very faint: it seems likely therefore that, observed with a less powerful microscope, they would only be recorded occasionally when α-particle scintillations were present as well.

In conclusion, therefore, it may be said that no effect has been observed which can in any way be attributed to the induction of disintegration in the polonium nucleus by any of the γ-rays of radium B+C. Very obviously this statement refers only to the concentration of source here employed, but it is interesting to remark that supposing a 10 percent effect had been observed at the beginning of the experiment, when the γ-ray source was most intense, then that would have corresponded to a reaction with the nucleus more than a million times more probable than the recognised types of reaction with the extra-nuclear electrons—and, moreover, this calculation is based on the supposition that γ-rays of all wave-lengths are capable of producing the nuclear change. The factor would be even greater if a selective effect were involved.

[17] In most cases observations with the source alone were carried out immediately after those under conditions of γ-ray irradiation. Occasionally this order was reversed, as may be seen from the figure, but without any systematic difference appearing.

[18] It may be remarked, for comparison with the earlier discussion, that of the 14 points belonging to each curve in Fig. 3, in each case 9 fall within the limits of probable error and 5 fall outside those limits.

[19] Chariton and Lea, Proc. Roy. Soc. **122A,** 335 (1929).