RP419

FLUCTUATIONS OF THE RATE OF EMISSION OF α-PARTICLES FOR WEAK SOURCES AND LARGE SOLID ANGLES

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ABSTRACT

An investigation of the fluctuations in the rate of emission of α-particles from very weak polonium sources (of the order of 3×10^{-12} curie/mm²) for solid angles approximately equal to 4π has been made, using automatic registration and a special double Geiger point counter. The object was to try to detect the large deviations for weak preparations reported by Pokrowski (Z.S. für Phys., vol. 58, p. 706, 1929; and vol. 59, p. 427, 1930). The total number of particles counted for two different sources was about 306,700 recorded in 14 different sets of observations at average rates of from 300 to 45 per minute, representing practically the total rate of emission from each source. The values of Q^2 for each set of observations were computed from the expression

$$Q^2=\frac{\sum\limits_{0}^{x} l_x x^2}{L}-m$$

where l_x is the number of equal intervals, each containing x α-particles, m is the the average number per interval and L is the number of α-particles observed. This divergence coefficient, which for a random distribution equals unity, varied between 0.828 and 1.110 for the 14 different sets. The average value of Q^2 obtained, however, is 1.004 which indicates that under these conditions there is no appreciable deviation from a purely random distribution.

CONTENTS

I. INTRODUCTION

Investigations of the accuracy with which the distribution of the rate of emission of α-particles conforms to that required by the law of probability have quite generally failed to show any perceptible deviation from the probability distribution. Renewed interest in this problem has resulted in several recent studies intended to detect small deviations from random distributions. In addition to observations on sources acted upon by some external stimulus, such as X rays or γ rays, considerable attention has been paid to radioactive preparations under normal conditions in an effort to discover anomalous effects. Attempts of this kind have been encouraged, in part at least, by the development of improved methods of automatic registration which might be expected to reveal small discrepancies which formerly were missed as the result of the subjective errors possible in the scintillation method of studying the problem, which, in the early investigations, was the only method available.

The investigation here described deals only with preparations in the absence of external stimulus. Furthermore it is not concerned with those deviations from random distributions such as have been reported by Kutzner and others,[1] using fairly concentrated preparations and which may be explained as the result of aggregate recoil. The present work was undertaken to test, by a different method from those previously employed, the suggestion of Pokrowski[2] that very weak sources might show deviations from a probability distribution. The experiments which he performed to test this idea were done with zinc sulphide screens artificially "contaminated" with a small amount of radioactive material (about 10^{-12} gm/mm^2). He reports decided deviations from a random distribution of such nature that very short intervals between appearances of particles occur much more frequently than is demanded by probability. On the other hand, two subsequent studies have been described, one by Herszfinkiel and Dobrowolska[3] and the other by Feather,[4] both using methods quite similar to those described by Pokrowski, but failing to confirm his results.

Since the experiments just mentioned were all done by the scintillation method it was thought possible that the disagreement which they exhibit could be referred in some way to the difficulties of making such observations and the possiblity for errors, particularly where a large number of scintillations are counted by a single observer. This consideration led the writer to undertake a few experiments, described in this paper, to test the weak source hypothesis of Pokrowski by a method using entirely automatic registration.

II. DESCRIPTION OF APPARATUS

In making a test of this hypothesis it is necessary to make certain of including as large a solid angle as possible since Pokrowski postulates that this anomalous behavior of weak preparations is the result of two somewhat independent phases of the condition which exists in a weak preparation. He assumes that the reaction of a disintegrating atom may increase the probability of one of its neighbors disintegrating even to the extent of causing it to disintegrate simultaneously; and, since there is little likelihood of both particles thus ejected travelling in the same direction, one of the particles would fail to be counted if, as usual, particles emitted within a small solid angle are observed. In his experiments he imbedded the radioactive material in the zinc sulphide screen to avoid this difficulty. If anything of this kind occurs it can best be detected by observations which include practically all the particles emitted by a preparation so that, if peculiar spatial distributions are present, these can also affect the results.

The apparatus used in the present experiments was designed to count as nearly as is practicable all the particles emitted by the source in all directions. A special double Geiger point counter was used for this purpose. It is shown diagrammatically in Figure 1. The two counting chambers are separated by a thin aluminum foil, *F*, of about 5 mm stopping power. The polonium source, *P*, is formed by allow-

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[1] Kutzner, W., Z. S. für Phys., vol. 21, p. 281, 1924. Curtiss, L. F., B. S. Jour. Research, vol. 4 (RP 166), p. 595, 1930.
[2] Pokrowski, G. I., Z. S. für Phys., vol. 58, p. 706, 1929; vol. 59, p. 427, 1930.
[3] Herszfinkiel, H., and Dobrowolska, H., Z. S. für Phys., vol. 62, p. 432, 1930.
[4] Feather, N., Phys. Rev., vol. 35, p. 705, 1930.

ing a small drop of a very dilute solution of polonium to evaporate on the foil. This solution was prepared by depositing a little polonium from a RaD solution on a bit of silver foil and then dissolving the foil in nitric acid. This solution was then diluted until sources of the desired strength could be obtained from it. The aluminum foil on which the source is deposited is mounted on a metal slide, *H*, and completely covers the circular opening therein. Thin sheet metal shutters, not shown, were provided which could be inserted in the slots, *S*, *S*, one on each side of the foil and close to it. These shutters were useful in determining the proper voltage for the counting chambers and in making certain that the chambers were working properly. Since the chamber walls of both counters were in metallic contact, any difference in operating voltages was taken care of by a biasing potential applied to one needle. *G*, *G* are guard rings connected to earth. In operation, the whole counter was inclosed in an air-tight copper box filled with dry air at atmospheric pressure.

To make certain that as large a proportion as possible of the total number of particles emitted by the source would be counted, specially sharpened steel points were used. The actual point had such a

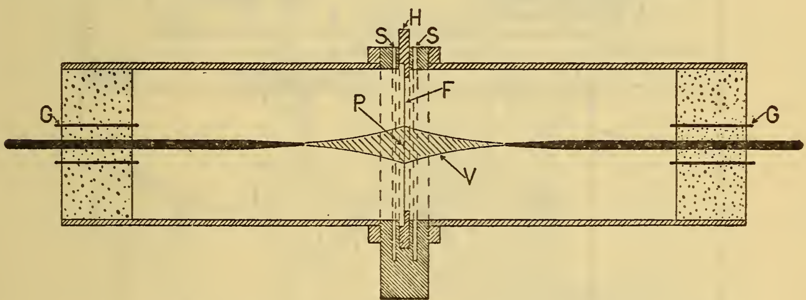

FIGURE 1.—*Diagram of double point counter*

curvature that the active volume, *V*, as indicated by the shaded region in Figure 1, was a cone which at its base was five or six times the diameter of the source itself. The diameter of the source was about 2 mm. Under these circumstances it can be seen that only particles traveling in the plane of the foil will fail to be counted since those which diverge even a very small amount from this plane must traverse a sufficient section of the active volume to produce a response in the counting chamber since a fraction of a millimeter of the path of the α-particle falling within the active volume will do this. Careful tests were made to insure that the active volume fulfilled the requirements set above. Although it is difficult to estimate what fraction of the particles emitted near the plane of the foil will not be counted, it seems probable that this can not amount to more than 5 per cent.

Since it was found simpler to use a single amplifier in connection with the registration of the impulses of the counter, the two points were connected together and all impulses from both points were fed into one recording system. This method of making the record will adequately serve to reveal the deviations in question, if they exist; for, although by this arrangement the simultaneous emission of the particles in opposite directions can not be observed, the failure to register these will increase the error of the observations and consequently will increase the divergence from a probability distribution. In a like manner if, as Powkrowski assumes, there is a great preponderance of very short intervals some of these particles will fail to be recorded if these intervals are less than the resolving power of the recording system. On the other hand, this use of a single amplifier eliminates all disturbing effects of recoil atoms, should any have sufficient energy to be counted, since the effect of the recoil atom will be included simultaneously with that of the α-particle which produced it. As a matter of fact, very little if any effect of recoil atoms could be detected. This was tested by connecting one counting chamber with the amplifier and introducing thin mica screens just in front of the aluminum foil separating the two counters. If any recoil radiation

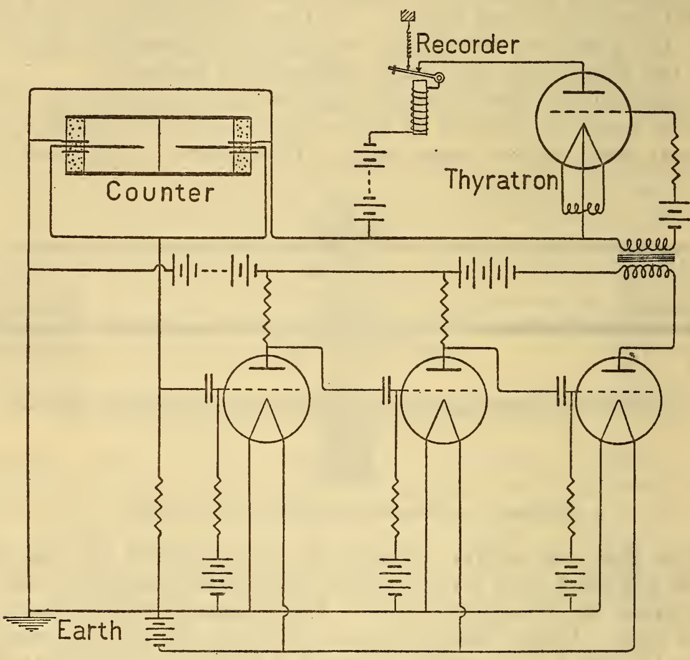

Figure 2.—*Vacuum tube amplifier and connections*

were present at all it would be very easily absorbed so that the introduction of a screen of a few millimeters stopping power should produce a decrease in the rate of counting. No such effect was observed.

The connection of the counter with the three-stage resistance-capacity coupled amplifier is shown in Figure 2. The time constants of the various stages were chosen to give satisfactory results at fairly high counting rates in order to make certain that the "resolving power" of the arrangement was limited only by that of the mechanical recorder used. In determining the best values for these constants it was found that the final choice could best be made by actual trial in the amplifier in agreement with the experience of Fränz.[5] In fact, the chief difference between this amplifier and that used by Franz is the use of a blocking condenser in the input stage. In order to elimi-

[5] Fränz, H., Z. S. für Phys., vol. 63, p. 373, 1930.

nate difficulties which are common with mechanical relays, a thyratron, connected as shown, was used to amplify the output sufficiently to give sharp powerful action in the mechanical recorder.

The mechanical recorder was of special construction shown in Figure 3, designed to reduce the inertia of moving parts as much as possible and to be suitable for use with a thyratron. A thin iron armature, *A*, is pivoted so that it may be drawn downward by the electromagnet, *E*. The spring, *S*, which holds the armature up against the support for the contact buttons is mounted in such a way that its tension can be adjusted. A thin phosphor bronze spring, *P*, carrying an intermediate contact button is mounted as shown, in electrical contact with the support carrying the fixed contact. This spring and intermediate contact is required because provision must be made to

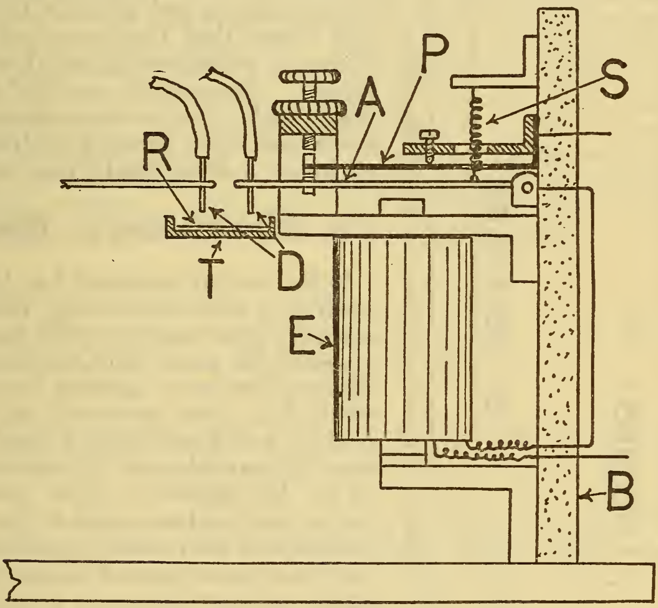

Figure 3.—*Diagram of mechanical recorder*

interrupt the plate circuit of the thyratron to reset it for the next signal. However, the break can not be made at the instant the armature starts downward, as would occur if the fixed contact alone were used, since then the action would be too feeble. By means of the intermediate contact the plate circuit can be interrupted at different points on the down stroke of the armature depending on the position of the adjusting screw which regulates the position of *P*. The wires protruding through the hard rubber base, *B*, are connected, one to the plate of the thyratron, and the other to the positive terminal of the thyratron plate battery, as shown in Figure 2. The outer end of the armature, *A*, carries a short metal tube, *D*, about 0.5 mm diameter. This serves as a dotting pen and is connected with a stationary ink reservoir by a fine rubber tube. When the armature is pulled down the pen makes a dot on the paper ribbon, *R*, moving at right angles to the plane of the figure in the trough shown in cross section at *T*. Another similar dotting pen records time signals.

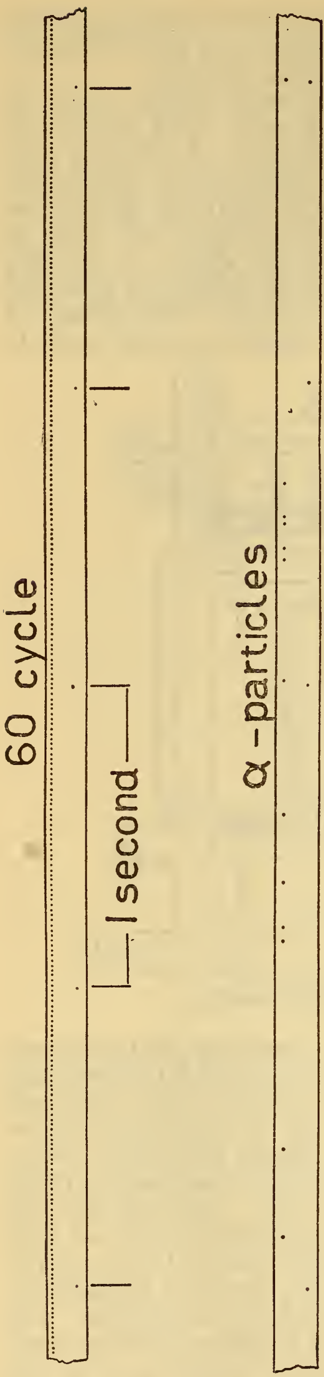

FIGURE 4.—*Typical records made with recording system*

This device is capable of recording signals at the rate of more than 100 per second. An example of a record is shown in Figure 4. The upper record is made from the 60 cycle a. c. supply and the lower is part of a record where α-particles are being recorded at an average rate of about 300 per minute. The time dots, in this case at one second intervals, also appear on each record. It was found convenient to use records made from the 60 cycle supply in adjusting the recorder. When careful adjustments were not made and tested in this way it was found that the recording system missed a considerable number of the signals. Records of α-particles made under such circumstances showed wide deviations from random distributions, always of a subnormal character.

III. EXPERIMENTAL RESULTS

Since special emphasis has been put upon very weak sources by Pokrowski and also upon sources which have been prepared for some time, the final set of observations were made on two sources which had been prepared for several months and which gave a total of less than 100 particles per minute as recorded by the apparatus here described. Such a preparation seems to fall within the range of Pokrowski's experiments, at least as regards order of magnitude. It can be estimated that one α-particle per second in a solid angle of 4π from a polonium source equals approximately 2×10^{-11} curies.[6] Since the area of both faces of the source is about 8 mm^2 this gives for the sources used approximately 2.5×10^{-12} curie/mm^2 as compared with the concentration given by Pokrowski (Z. S. für Phys., vol. 58, p. 706, 1929) of "about 10^{-12} gm/mm^2." Of the records made, the longest contained about 40,000 particles and the shortest 7,000. The longer records were made at an average rate of about 100 per minute so that no set of observations required more than seven hours. Therefore, no correction was made for the natural rate of decay of the polonium source since for this period it is negligible.

[6] Gregoire, C. R., vol. 193, p. 42, 1931.

The resolving power of the apparatus was estimated at 1/100 second. This was deduced from its performance, as shown in Figure 4, in counting 60 cycle pulses. For this resolution the accuracy for an average counting rate at 1 per second is 99 per cent. For a counting rate of 5 per second, it is 95 per cent. Another factor which, of course, must be considered in comparing the results with those required by probability is the number of particles included in a record. For this reason those records which have fewer particles may be expected to show greater variations than the longer records. However, as will be seen, even the shortest records give reasonable agreement with probability distributions it is assumed that all records contain a sufficient number of particles to justify a comparison with a random distribution.

The comparison of the results with those to be expected according to probability was made by computing the Lexis divergence coefficient, Q^2, as explained elsewhere.[7] For a normal distribution $Q^2=1$. For a supernormal distribution, such as Pokrowski found, where the number of intervals differing considerably from the mean interval are in excess, $Q^2>1$. The results are tabulated in Table 1 where Σl_x is the total number of equal intervals for which α particles were counted; x is number of particles in an interval, hence $\Sigma x l_x$ is the total number of particles counted. $\Sigma x^2 l_x$ and m, the average number of particles per interval, are used in the expression for obtaining Q^2

$$Q^2=\frac{\overset{x}{\underset{0}{\Sigma}} x^2 l_x}{\Sigma x l_x}-m$$

TABLE 1

Date	Σl_x	$\Sigma x l_x$	$\Sigma x^2 l_x$	m	Interval	Q^2
1931					*Second*	
October 8	4,926	26,653	166,263	5.411	1	0.828
	7,160	39,307	254,389	5.490	1	.982
October 9	3,634	16,242	89,070	4.470	1	1.014
October 15	2,050	11,972	83,210	5.840	5	1.110
October 16	2,234	12,213	79,623	5.467	5	1.052
	1,977	8,155	42,061	4.125	5	1.033
October 17	1,473	7,281	42,631	4.943	5	.912
October 20	4,219	22,902	145,850	5.428	5	.940
October 21	4,236	22,873	145,949	5.400	5	.982
October 22	3,755	25,306	195,294	6.739	5	.978
October 28	4,241	34,863	324,689	8.221	5	1.092
October 29	3,399	26,423	234,419	7.774	5	1.098
October 30	3,851	31,981	309,969	8.348	5	1.038
October 31	2,243	20,524	208,400	9.150	5	1.004

A test of whether a set of Q^2 shows a genuine departure from the values to be expected is given by Bortkiewicz.[8] This test consists of comparing the mean value of the Q^2 with mean error of the Q^2 defined by

$$M\left(1/\nu\sum_{1}^{\nu}{}_{i}Q_i^2\right)=1/\nu\sqrt{\sum_{1}^{\nu}{}_{i}M^2(Q_i^2)}$$

[7] Curtiss, L. F., B. S. Jour. Research, vol. 4 (RP166), p. 595, 1930.
[8] v. Bortkiewicz, L., Die Radioaktive Strahhlung als Gegenstand wahrscheinlichkeitstheoretischer Untersuchungen, Julius Springer, Berlin, 1913.

where ν equals the number of sets of observations from which the values of Q_i^2 ($_i^2=1, 2 \ldots \nu$) where determined and $M^2(O_i^2)=\frac{2m_j+1}{L_j}$ where L_j = total number of particles counted for that set of observations. Table 2 contains quantities required to determine the mean error for the observations recorded in Table 1.

TABLE 2

Q^2	m	L	$M^2(Q_i^2)$
0.828	5.411	26,653	0.000406
.982	5.490	39,307	.000302
1.014	4.470	16,242	.000612
1.110	5.840	11,972	.001060
1.052	5.467	12,213	.000985
1.033	4.125	8,155	.001134
.912	4.943	7,281	.001495
.940	5.428	22,902	.000531
.982	5.400	22,873	.000516
.978	6.739	25,306	.000572
1.092	8.221	34,863	.000500
1.098	7.774	26,423	.000627
1.038	8.348	31,981	.000553
1.004	9.150	20,524	.000940
$\sum_1^{\nu}{}_i (Q_i^2)=14.063$	----------	----------	$\sum_1^{\nu}{}_i M^2(O_i^2)=.010233$

Inserting the numerical values in the expression for the mean error

$$1/\nu\sqrt{\sum_1^{\nu}{}_i M^2(Q_i^2)}=1/14\sqrt{0.010233}=0.00721$$

The average value of $Q^2=\frac{14.063}{14}=1.004$ so that the deviation of the average from the expected value of unity is 0.004. This is approximately one-half the mean error (0.00721) just computed. We can conclude, therefore, that the variations exhibited by the Q^2 fall well within those permitted theoretically and the deviations of the Q^2 from unity are to be regarded as accidental and not systematic.

In view of the foregoing there seems to be little room to doubt that laws of probability are accurately followed even in the low concentrations of active material which are dealt with here. It thus seems logical to conclude that probability laws apply over an enormous range of concentrations, roughly, from 10^{-3} to 10^{-12} gm/mm^2, since the accuracy with which concentrated preparations conform to an exponential rate of decay verifies the application of probability laws in their case. Consequently, in view of the fact that these laws hold for concentrations varying by a factor of about 1,000 million, leaves little room to doubt that they would apply for any concentration which we are able to measure or observe.

The writer wishes to acknowledge the help of Miss C. L. Torrey and L. L. Stockman and B. W. Brown in making and counting records and in the computation of the numerical results.

WASHINGTON, January 18, 1932.