# The General Theory of Fluctuations in Radioactive Disintegration\*

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The chance,  $W_n(0, t)$ , that *n* atoms of a radioactive source will disintegrate in the interval  $(0, t)$  may be found if we know  $f_r(t)dt$ , the chance that a disintegration will occur in dt at t after r have occurred in  $(0, t)$ . Bateman's differential equation is generalized to cover the case in which  $f_r$  depends on  $t$  and  $r$ . The solution is given for the case in which it depends only on  $t$ . A detector of efficiency  $g$ subtends a solid angle  $4\pi A$  at a source (decay constant  $\lambda$ ), containing  $N$  atoms at time zero. The probability of  $n$ *counts* in the interval  $(T_1, T_1+T_2)$  is

$$
P_n(T_1, T_1+T_2) = C_n N (e^{\lambda T_2} - 1)^n e^{-N\lambda (T_1+T_2)}
$$
  
 
$$
\times (gA)^n [1+e^{\lambda T_2} (e^{\lambda T_1} - 1) + (1-gA) (e^{\lambda T_2} - 1)]^{N-n}.
$$

Putting  $g$  and  $A$  equal to 1 we get the probability  $W_n(T_1, T_1+T_2)$  of n disintegrations in this interval. Bortkiewicz's formula  $W_n(0, t) = C_n N(e^{\lambda t} - 1)^n e^{-N\lambda t}$ , is a special case. If we measure a great number,  $r$ , of intervals between the disintegrations of atoms in a single source, the fraction of the intervals that exceed t has the "expected" value

$$
(e^{-N\lambda t}-e^{-(N-r)\lambda t})/r(1-e^{\lambda t})
$$

The problem of determining fluctuations in the disintegration of a single substance from fluctuations in counting is solved for the case in which each disintegration gives a single ray capable of actuating the counter. Fluctuations in counts produced by two or more independent sources are considered. Since gamma-rays and secondary beta-rays are not emitted by every disintegrating atom, the distribution of counts due to such rays is discussed. The effect of the recovery time,  $\tau$ , of the counter is discussed, using at first

the assumption that  $\tau$  is the same for all counts, an approximation useful at low counting rates. Kith a source that would produce f counts per sec. if  $\tau$  were zero, the probability of  $n$  counts in the interval  $(0, t)$  is obtained for two cases: (1) The counter is not clogged at time zero; (2) it is clogged. The second case has practical interest; the probability of an interval greater than  $t$  between counts is l if  $t \leq \tau$  and  $e^{-f(t-\tau)}$  if  $t \geq \tau$ . Differential-difference equation for the Auctuation-functions are derived. The Auctuations of counts due to a constant source, in a counter with variable recovery time, are obtained, using Skinner's formula for the frequency distribution of recovery times. For  $t$  values greater than the maximum recovery time, the probability of an interval greater than t is  $e^{-f(t-\tau')}$ ;  $\tau'$  is a constant. Formulas are derived for the stock fluctuations of each substance in a source containing several members of a radioactive series, subject to any desired initial conditions; recurrence equations governing the stock probabilities are given. The probability of a stock  $n$  of the daughter of <sup>a</sup> constant parent which yields f disintegrations per sec. is  $S_n = (f/\lambda)^n e^{-(f/\lambda)}/n!$ ; here  $\lambda$  is the decay constant of the daughter. General methods for finding the fluctuations in the emission of an entire radioactive series, or any part of a series, are given. The disintegration-fluctuations of the daughter of a constant parent obey the formula  $W_n = (ft)^n e^{-ft}/n!$ which applies also to the parent. However, fluctuations of parent and daughter are coupled, so that the Bateman type of formula does not apply to their combined emission. Instead, the probability of an interval greater than  $t$  is  $\exp\left[-ft-(f/\lambda)(1-e^{-\lambda t})\right]$ .

#### A. FLUCTUATIONS IN THE DISINTEGRATION OF <sup>A</sup> SINGLE SUBsTANcE

#### l. <sup>A</sup> generalization of Bateman'8 differential equation

IN 1910 Bateman<sup>1</sup> derived his famous formul  $\blacksquare$  for the probability that *n* atoms will disintegrate in time  $t$ , in a source whose diminution during the experiment can be neglected. It is

$$
W_n = (ft)^n e^{-ft}/n!,\tag{1}
$$

where  $f$  is the average number of disintegrations

A.  $f_r$  is a function of t or a constant, but does not depend on  $r$ , so that we may omit the subscript.

<sup>\*</sup> Preliminary communication, see Phys. Rev. 48, 772  $(1935).$ <sup>1</sup> Bateman, Phil. Mag. 20, 698 (1910).

per unit time. Bortkiewicz', gave the formula which applies to a decaying source. We shall present a differential equation which covers a much broader range of possibilities, yielding the formulas of Bateman and Bortkiewicz as special cases. Suppose we know  $f_r(t)dt$ , the probability that one disintegration will occur in  $dt$  at  $t$  when  $r$  have occurred in the interval  $(0, t)$ . Then there are two cases to be considered:

B.  $f_r$  depends on both  $r$  and  $t$ .

Bortkiewicz, Die Rgdioaktive Strahlung als Gegenstand wahrscheinlichkeitstheoretischer Untersuchungen (Springer, 1913), p. 75.

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FIG. 1. Two independent ways in which  $n$  events may occur in time  $t+dt$ .

In each case the primary problem is to find  $W_n(0, t)$ , the probability that *n* disintegrations occur in the interval  $(0, t)$ . When this is known we can obtain other useful functions, such as  $W_{s, n}(T_1, T_1+T_2)$ , the probability that *n* events occur in the interval from  $T_1$  to  $T_1+T_2$  when s events are known to have occurred in the interval  $(0, T_1)$ . We shall drop the time-arguments when they are not needed for clarity.

To obtain a differential equation obeyed by  $W_n(0, t)$ , we note that the probability of *n* events between 0 and  $t+dt$  is the sum of two terms (Fig. 1): the probability of  $n-1$  events in the interval  $(0, t)$ , times the probability of one event in  $dt$  at  $t$ ; (2) the probability of  $n$  events in the interval  $(0, t)$ , times the probability of no events in  $dt$  at  $t$ . Thus,

 $W_n(0, t+dt)$ 

$$
=W_{n-1}(0, t)f_{n-1}dt+W_n(0, t)\cdot(1-f_ndt);
$$

or

$$
dW_n/dt = f_{n-1}W_{n-1} - f_nW_n; \quad n = 1, 2, \cdots, \quad (2)
$$

When *n* is zero,  $f_{n-1}$  is zero, so that

$$
dW_0/dt = -f_0W_0.
$$
 (2a)

These equations are a generalization of the Bateman equations, reducing to them when f is constant. The initial conditions are  $W_0(0, 0)$  $= 1$ ;  $W_n(0, 0) = 0$  when *n* is not zero. Therefore

$$
W_0 = e^{-\int_0^t f_0 dt},\tag{3}
$$

and we can obtain the other  $W$ 's in succession by using the general solution of (2), namely,

$$
W_{n} = e^{-\int f_{n}dt} \cdot \int_{0}^{t} f_{n-1} W_{n-1} e^{\int f_{n} dt} dt; \qquad n = 1, 2, \cdots. \quad (4)
$$

# 2. General solution when  $f$  is a known function of t

If the chance  $f(t)dt$  of an event in time dt at t depends only on  $t$ , Eq.  $(2)$  becomes

$$
dW_n/dx = W_{n-1} - W_n,\tag{5}
$$

$$
x = \int_0^t f(t)dt.
$$
 (6)

The solution of (5) is

$$
W_n(0, t) = x^n e^{-x}/n!
$$
 (7)

Also, we have 
$$
W_n(t_1, t_1+t) = x^n e^{-x}/n!
$$
, (8)

where now 
$$
x = \int_{t_1}^{t_1+t} f(t)dt
$$

The mean number of events in the interval  $t_1$ ,  $t_1+t$  is found to be x, and the standard deviation (r.m.s. fluctuation about the mean) is  $x^{\frac{1}{2}}$ . The Lexian ratio,—that is (standard deviation)<sup>2</sup>/ mean,—is therefore unity, as it also is for <sup>a</sup> source of constant strength. Eqs. (7) and (8) give approximate information about the Huctuations of a preparation containing a large number of atoms, where variations of the stock are negligible. For a preparation whose decay constant is  $\lambda$ , containing N atoms at time zero, f approaches  $Ne^{-\lambda t} \lambda$  as N increases. Putting  $x = N(1 - e^{-\lambda t})$  in Eq. (7), we may compute approximate values of the Huctuation of ionization current, and other useful quantities. Eqs. (7) and (8) cannot be applied to a source that contains only a small number of atoms and has a decay constant so high that the strength of the source falls off considerably during the experiment. For such a source, the more general Eq. (2) must be used.

#### 3. The case of a decaying source

Probability of n counts in a definite interval beginning at a definite time. If a source contains  $N$  atoms at time 0, and  $n$  disintegrate in time  $t$ , the probability that one of the remaining atoms will disintegrate in t,  $t+dt$  is

$$
f_n dt = (N - n)\lambda dt.
$$
 (9)

Then the solution of (2) is

$$
W_n(0, t) = C_n{}^N (e^{\lambda t} - 1)^n e^{-N\lambda t}, \qquad (10)
$$

where  $C_n^N = N!/(N-n)!n!$ . A simple way to

show this is the following. If we observe the source continuously during the interval  $(0, t)$ we are really making N experiments, each one of which consists in seeing whether a particular atom disintegrates. The chance that a particular atom will disintegrate is  $p=1-q=1-e^{-\lambda t}$ , and the probability of  $n$  disintegrations in  $N$  "trials" is  $C_n^N p^n q^{N-n}$  which may be converted into (10). If we study a great number of identical sources, the "expected" average of *n* is  $pN$ , or  $N(1-e^{-\lambda t})$ , and the standard deviation is

Let us now obtain  $W_n(T_1, T_1+T_2)$ , the probability that  $n$  events will occur in the interval  $(T_1, T_1+T_2)$  when we do not know the number that occurred in the previous interval  $(0, T_1)$ . By Eq. (10), the probability that *n* events occur in  $(T_1, T_1+T_2)$  when s have occurred in  $(0, T_1)$ 1s

$$
W_{s, n}(T_1, T_1+T_2)
$$
  
=  $C_n^{N-s}(e^{\lambda T_2}-1)^n e^{-(N-s)\lambda T_2}$ . (11)

$$
(Npq)^{\frac{1}{2}} = [N(1 - e^{-\lambda t})e^{-\lambda t}]^{\frac{1}{2}}.
$$

Therefore,

$$
W_n(T_1, T_1 + T_2) = \sum_{s=0}^{N-n} W_s(0, T_1) W_{s, n}(T_1, T_1 + T_2)
$$
  
=  $C_n^N (e^{\lambda T_2} - 1)^n e^{-N\lambda (T_1 + T_2)} \sum_{s=0}^{N-n} C_s^{N-n} [e^{\lambda T_2} (e^{\lambda T_1} - 1)]^s$   
=  $C_n^N (e^{\lambda T_2} - 1)^n e^{-N\lambda (T_1 + T_2)} [1 + e^{\lambda T_2} (e^{\lambda T_1} - 1)]^{N-n}$ . (12)

The average number of disintegrations and the standard deviation may be obtained from Eqs. (18a) and (18b), as special cases.

Methods for testing fluctuation theory. Eq. (12) can be used to test the Huctuation theory for substances of any desired half-life, whereas previous work has dealt with approximately constant sources. The appropriate procedures for experimentation and for summarizing the data are somewhat different from those employed in the past. When dealing with a constant source, one obtains chronograph records showing the times of occurrence of a great number of disintegrations, and then two convenient methods of preparing the data for statistical analysis are available, associated with the names of Marsden and Barratt, and of Rutherford and Geiger, respectively. They are as follows:

(2) The record is divided into equal intervals of length  $t$ , and the fraction of the intervals containing  $n$  events is determined, for different values of  $n$ .

In dealing with decaying substances, several plans are available. One may obtain chronograph records from a number of sources, all of the same initial strength as nearly as possible. Then, referring to Eq. (12), one may fix the value of  $T_2$ and find the fraction of the sources that give  $n$ particles in the interval  $(0, T_2)$ ; *n* particles in the interval  $(T_2, 2T_2)$ ; and so on, taking  $T_1=2T_2$ ,  $3T_2$ , etc., in succession. The process can be repeated for other values of  $T_2$ , and the dependence of the results on  $n$ ,  $T_1$ , and  $T_2$  can then be compared with Eq. (12). This method is obviously desirable when one must deal with weak sources that decay rapidly, so that only a limited number of counts can be obtained from each source. On the other hand, when sufficiently strong preparations are available, one will desire to analyze records obtained from a single source.

<sup>(1)</sup> The fraction  $F_0(t)$  of the intervals between disintegrations that are longer than  $t$  is determined for various values of t. These fractions should obey the law  $F_0(t) = e^{-ft}$ , according to Eq.  $(1).$ <sup>3</sup>

<sup>&</sup>lt;sup>3</sup> This statement is often questioned by those who meet it for the first time. Eq. (1) with  $n=0$ , really gives the probability that zero events will occur in a time t after an arbitrary initial instant. Therefore it seems unfair in analyzing data, to make every interval begin at the instant when

a disintegration occurs. This mental difficulty may be dispelled by noting that the probability  $fdt$  for an event to  $\alpha$  occur in dt is not affected by the occurrence or nonoccurrence of an event at the beginning of dt. The constant source may be considered as one having an "infinite" stock of atoms and an "infinitesimal" decay constant.

Assigning to  $T_2$  a fixed value t, one counts the number of particles in the intervals  $0, t; t, 2t;$ etc., as before and determines the fraction of the *intervals* in which  $n$  particles occur. Let there be  $r$  intervals altogether. In the jth interval, the probability of *n* particles is  $W_n(\lceil j-1 \rceil t, jt)$ . For fixed values of  $n$  and  $t$ , these probabilities depend only on  $j$ , and by a known theorem,<sup>4</sup> this means that out of  $r$  intervals the fraction containing *n* particles has the expected value

$$
r^{-1} \sum_{j=1}^{r} W_n \left( \left[ j-1 \right] t, j t \right)
$$
  
=  $r^{-1} C_n N (e^{\lambda T_2} - 1) n \sum_{j=1}^{r} e^{-j N \lambda T_2} (1 - e^{\lambda T_2} + e^{j \lambda T_2})^{N-n}$ 

Unfortunately this sum cannot be simplified, as one may see by examining the trivial case  $r=2$ . For practical purposes, it is necessary to use another method of analysis, now to be explained.

Size-distribution of intervals between emissions from a single source. It is possible to find a formula for the fraction  $F_0(t)$  of the intervals between emissions that exceed  $t$ . Let the source contain  $N$  atoms initially. By Eq. (10), the probability that the first interval will exceed  $t$ is  $e^{-N\lambda t}$ . After the first disintegration has occurred, the probability that the second will exceed t is  $e^{-(N-1)\lambda t}$ , and so on. For a given value of  $t$ , each of these probabilities is fixed, so that out of  $r$  intervals the expected number exceeding

t is  $\sum_{j=1}^r e^{-(N-j+1)\lambda t}$ ; that is, the fraction of the r

 $intervals that exceed t has the expected value$ 

$$
F_0 = \frac{e^{-N\lambda t} - e^{-(N-r)\lambda t}}{r(1 - e^{\lambda t})}.
$$
 (13)

This generalization of Bateman's formula makes it possible to carry out a Marsden-Barratt analysis of the fluctuations of a single decaying source.

#### B. FLUCTUATIONS IN COUNTING WITH <sup>A</sup> DETEC-TOR OF NEGLIGIBLE RECOVERY TIME

# 4. Effect of solid angle subtended at source by the detector, and of detector efficiency

So far we have dealt with the time-distribution of all the disintegrations occurring in a source. A direct test of the formulas derived would. require a detector of perfect efficiency, which catches all the emitted particles. Therefore we now discuss the inHuence of the solid angle subtended by the detecting device, and of its failure to record all particles that reach it, To bring out clearly the effects of these factors, we assume in Sections 4 to 6 that the finite time-resolving power of the instrument can be neglected, which means that the results in these sections are valid only for low counting rates. In nearly all counting experiments on radioactive Huctuations made up to the present time, the detecting device subtended only a small solid angle at the source,<sup>5</sup> so that randomness in the direction of emission was superposed on the time distribution of the disintegrations. It has often been stated' that experiments in which the detecting device subtends a very small solid angle at the source cannot serve as a test of Huctuation theory. This is easily seen by considering an extreme and fantastic case, a hypothetical source, emitting particles at uniform intervals. If it yields f particles in unit time, the probability that in time  $t$  n particles will pass through a very small solid angle  $\omega$  is, by Poisson's formula,  $(Att)^n e^{-Att}/n!$ , where  $A = \omega/4\pi$ ; but this is exactly what we would expect from a source subject to Huctuations, yielding an average of Af particles per unit time, in the solid angle  $\omega$ . Similarly, if the efficiency, g, of the counter' in detecting a single particle is very small, the time-distribution of counts will follow Bateman's formula even though the counter receives the entire output of the abovementioned hypothetical source.

However, we can prove that if g and  $A$  are not very small compared with unity, a definite test of

<sup>4</sup> Czuber, Wuhrscheinlichkeitsrecknung, Vol. 1, p. 78.

<sup>&</sup>lt;sup>5</sup> There are two exceptions. Kohlrausch and Schweidler (Physik. Zeits. 13, 11 (1912)) attempted to utilize the entire emission, but Kohlrausch says the observations did not get beyond a qualitative stage. Curtiss (Bur. Standards J, Research 8, 329 (1932)) used nearly the entire emission from a polonium source deposited on a thin foil between two counters.

See for example, Kohlrausch, Radioaktivität, p. 789.

<sup>7</sup> Such an efficiency factor may arise in many ways. For example, in recording x-rays with a tube counter, only a part of the rays undergoes absorption or scattering in such a way that counts, occur. The efficiency will of course be different for different kinds of rays or particles. (Section 6.) If several efficiency factors are active in sequence, we simply use an overall factor equal to their product; thus, if  $v$  is the probability that a photon will liberate an electron in the wall of a counter, and  $w$  the probability that this electron will produce a count,  $g=vw$ .



FIG. 2. In a typical case the source emits s particles in the interval  $(T_1, T_1+T_2)$ ; the detector records *n* of them.

the fluctuation theory can be made. For generality, we discuss a decaying source containing N atoms at time zero. The chance that the source will emit s particles or rays in the interval  $(T_1, T_1+T_2)$ is  $W_s(T_1, T_1+T_2)$ , given by Eq. (12); when this event happens, the chance (Fig. 2) that any one of the s particles will be recorded is  $gA$ , and the chance that  $n$  of them will be recorded is

$$
C_n^{\ s}(gA)^n(1-gA)^{s-n}.\tag{14}
$$

Therefore the chance of recording  $n$  particles in the interval  $T<sub>2</sub>$ , however many are emitted by the source, is

$$
P_n(T_1, T_1+T_2)
$$
  
=  $\sum_{s=n}^{N} W_s(T_1, T_1+T_2) C_n^s (gA)^n (1-gA)^{s-n}$   
=  $C_n^N e^{-N\lambda (T_1+T_2)} [1+e^{\lambda T_2} (e^{\lambda T_1}-1)]^N$   
 $\times \left(\frac{gA}{1-gA}\right)^n \sum_{s=n}^{N} \frac{(N-n)!}{(N-s)!(s-n)!} x^s$ , (15)

where

$$
x = \{ (e^{\lambda T_2} - 1)(1 - gA) / \lfloor 1 + e^{\lambda T_2} (e^{\lambda T_1} - 1) \rfloor \}.
$$

The summation in Eq. (15) is  $x^{n}(1+x)^{N-n}$ , so that

$$
P_n(T_1, T_1+T_2) = C_n N e^{-N\lambda (T_1+T_2)} (e^{\lambda T_2}-1)^n (gA)^n \quad \text{and}
$$
  
 
$$
\cdot [1+e^{\lambda T_2}(e^{\lambda T_1}-1)+(1-gA)(e^{\lambda T_2}-1)]^{N-n}. \quad (16)
$$

This is a generalization<sup>8</sup> of Eq.  $(12)$ , and from it certain interesting facts emerge.

(1) If N approaches unfinity, but N $\lambda$  and n remain finite, then  $T_1$  drops out of Eq. (16) and

$$
\left[1 - e^{-\lambda T_r} g A \left(1 - e^{-\lambda t}\right)\right]^{N-r},
$$

we have the Bateman formula for the probability of n counts in any interval of length  $T_2$ :

$$
P_n(T_2) = (N\lambda gA T_2)^n e^{-N\lambda gA T_2}/n! \qquad (17)
$$

(2) While the time-distributions of counts and of disintegrations are of the same form for a "constant" source, this is not true for short-lived sources.

(3) The mean number of counts in the interval  $T_2$  is

$$
\bar{n} = Ne^{-\lambda T_1}(1 - e^{-\lambda T_2})gA, \qquad (18a)
$$

and the mean square deviation is

$$
\overline{n^2} - \overline{n}^2 = Ne^{-\lambda T_1}(1 - e^{-\lambda T_2})gA
$$

$$
\times [1 - e^{-\lambda T_1}(1 - e^{-\lambda T_2})gA]. \quad (18b)
$$

Many writers have used the value of the Lexian ratio, as a criterion for the random character of radioactive disintegration. For sources whose decay during a single experiment can be neglected, this ratio should be unity, whatever solid angle is subtended by the counting device. The case of a decaying source was discussed by Bortkiewicz<sup>9</sup> but he did not consider the influence of counter efficiency and solid angle. His discussion covers the frequency distribution of  $n$ values in equal intervals of time, for a single source; that is,  $T_2$  is constant, and  $T_1$  increases by equal steps. For this case, the expected value of the Lexian ratio is greater than unity. On the other hand, the Lexian ratio given by Eqs. (18a) and (18b), is less than unity. It refers to the case in which we choose fixed values of  $T_1$ and  $T_2$ , make experiments on a great number of sources, and consider the frequency-distribution of *n* values in the interval  $T_2$ . The complicated form of Eqs. (12) and (16) and of similar equations which follow, makes it inconvenient to use the Lexian ratio in studying decaying sources. On the other hand,  $P_0(T_1, T_1+T_2)$  is simple in form, and if we determine by experiment the dependence of this function on  $T_1$  or  $T_2$ , a comparison with theory can be made by a method parison with theory can be made by a method<br>employed by Bortkiewicz.<sup>10</sup> This is the most convenient procedure.

<sup>&</sup>lt;sup>8</sup> Unfortunately Eq. (13) cannot be generalized to take account of g and  $A$ . It can be shown that when *r counts* have been observed, the probability that the interval be-<br>tween the rth and  $(r+1)$ th counts will exceed  $t$  is

where  $T_r$  is the time of occurrence of the rth count. Since this expression depends on  $T_r$  as well as r, the method of proof which led to (13) can no longer be invoked.

<sup>&#</sup>x27; Bortkiewicz, reference 2, p. 72.

Reference 2, pp. <sup>5</sup>—15.

# 5. Determination of fluctuations in disintegration when the distribution of counts is a known function

Suppose the functions  $P_n(T_1, T_1+T_2)$  have been found by experiment. When X is finite, and  $T_1$  and  $T_2$  are given fixed though arbitrary values, Eq. (15) constitutes a set of linear equations that can be solved for the functions  $W_s$  in terms of the functions  $P_n$  and the solution is unique. Thus experiments on the time-distribution of counts can give us a definite test of the probability theory of radioactive decay.

To solve the Eq. (15) when  $T_1$  and  $T_2$  are given fixed values, we omit the arguments of the functions  $W_s$  and  $P_n$ , and write

$$
v_s = W_s (1 - gA)^s; \quad p_n = P_n \left(\frac{1 - gA}{gA}\right)^n. \quad (19)
$$

Then (15) assumes the simple form

$$
\sum_{s=n}^{N} C_n^s v_s = p_n. \tag{20}
$$

The work is done in Appendix 1 and the result is

$$
W_i = (1 - gA)^{-i}(-1)^i
$$
  

$$
\sum_{i=j}^{N} C_i{}^{i} \left(\frac{1 - gA}{gA}\right)^i (-1)^i P_i.
$$
 (21)

Since this result holds for every choice of  $T_1$ and  $T_2$ , it is the general solution of our problem.

# 6. Distribution of counts due to gamma-rays and, secondary beta-rays; counts due to two or more independent sources

Gamma-rays and secondary beta-rays are emitted very quickly after a disintegration, but this does not mean that their time-distribution is the same as that of disintegrations. Some transitions leave the daughter nucleus in the ground state and in general, a given gamma-ray is emitted only in a certain fraction of the disin- -tegrations. The distribution of counts due to gamma-rays is found as follows. Let  $G_i$  be the probability of emission of a gamma-ray of frequency  $v_i$ , when a given atom disintegrates, and  $g_i$  the counter efficiency for this ray. The effective solid angle subtended at the source by the detector may also be different for the various rays,



because of differences in their penetrating power. The value of this solid angle for the *j*th ray may be denoted by  $A_i$ . Then, the probability that a gamma-ray of this frequency will be emitted and will produce a count is  $G_iA_i g_i$ ; and if the gammarays are mutually exclusive the total probability that the given disintegration will result in a count is  $\sum G_i A_i g_j$ . Replacing gA by this sum, the problem is treated exactly as in Section 4.

The situation is different if some of the gammarays are emitted in sequence without measurable time lag, which is often the case. To illustrate this, consider a hypothetical nucleus that emits only three gamma-rays, shown in the energy diagram (Fig. 3). Let the emission probabilities be  $G_1, G_2, G_3$ , where  $G_1$  obviously equals  $G_2$  and let the counter efficiencies be  $g_1$ ,  $g_2$ ,  $g_3$ . Then we can show that the probability a count will result from a given disintegration is  $G_1(A_1g_1)$  $+A_2g_2 - A_1A_2g_1g_2)+G_3A_3g_3$ ; this quantity replaces  $gA$  in the analysis of Section 4.

Let us now discuss the distribution of counts when two or more sources are superposed, or mixed. The superposition of any number of independent sources obeying Bateman's formula gives a resultant distribution obeying Bateman's formula; to be exact, if the average numbers of events caused by the individual sources in time  $t$  are  $x_1$ ,  $x_2$ , etc., then the probability that all of them together will yield  $n$  events in time  $t$  is  $(\sum x)^n e^{-\sum x}/n!$  The sources may be at different distances from the counter, may give rays of different absorbability, or may differ in any other respect; still the formula holds. The situation is quite different for decaying sources. By Eq. (16), the probability of *n* counts due to two such sources in the interval  $T_2$  is

$$
\sum_{j=0}^n P_i{}^{(1)} P_{n-j}{}^{(2)},
$$

where the superscripts refer to the individual

sources. This is not of the form (16), unless the decay constants are equal and the products gA are the same for the two sources. Of course, when these quantities are equal we are dealing essentially with a single source.

# C. DISTRIBUTION OF COUNTS IN <sup>A</sup> DETECTOR WITH FINITE RECOVERY TIME

Preceding sections dealt with the distribution of counts in a detector characterized by constant efficiency factors for the different classes of particles or rays which it receives, but having a negligible recovery time. Now we shall consider the distribution of counts in a detector whose efficiency drops to zero when a count occurs. We shall suppose (1) that it remains zero for a fixed time  $\tau$ ; and (2) that the counter then returns to time  $\tau$ ; and (2) that the counter then returns to<br>a state of perfect efficiency.<sup>11</sup> We wish to find  $P_n(0, t)$ , the probability that *n counts* occur in the interval  $(0, t)$ . The general problem is complex, and we shall deal only with the case of a constant source.

### 7. Counts due to a constant source; counter not clogged at  $t=0$

Let the counter receive  $f$  events per second, on the average. Then  $P_0 = e^{-ft}$ . Up to the time  $t = \tau$ ,  $P_2$ ,  $P_3$ , etc., are zero, for if one count occurs anywhere in this interval, the counter is clogged for the remainder of the interval. Therefore, if  $t \leq \tau$ :

$$
P_1 = 1 - P_0 = 1 - e^{-ft}.\tag{22}
$$

Now, let t be greater than or equal to  $\tau$ , and consider the probability of just one count, occurring between s and  $s+ds$ . There are two cases, as-shown at the bottom of Fig. 4A.

(1) If  $s \leq t - \tau$ , the probability of one count in ds is (prob. of no event up to s) $\times$ (prob. of 1, count in ds) $\times$ (certainty that no further count occurs up to  $s+r$ , the counter being clogged)  $\times$  (prob. of no event from  $s+\tau$  to t)  $=e^{-fs}fds e^{-f(t-s-\tau)}$ .

(2) If  $s \geq t - \tau$  we omit the last factor.



FIG. 4A. Probabilities of no counts and of one count in detector which is not clogged when  $t=0$ ; and diagram illustrating derivation of Eq. (23).

B. Probabilities of no counts and of 1 count in detector which is clogged when  $t = 0$ . The curves are drawn for the case  $f=1/\tau$ .

Therefore, if  $t \geq \tau$ :

$$
P_1 = \int_0^{t-\tau} e^{-f(t-\tau)} f ds + \int_{t-\tau}^t e^{-f s} f ds
$$
  
=  $e^{-f(t-\tau)} [1 + f(t-\tau)] - e^{-f t}.$  (23)

By similar methods  $P_n$  may be computed. The work is greatly shortened by the following facts: (1)  $P_n = 0$  if  $t \leq (n - 1)\tau$ . (2) In the range  $(n - 1)\tau$ . to  $n\tau$ ,  $P_{n+1}$  and all higher P's are zero, so in this range  $P_n = 1 - P_1 \cdots - P_{n-1}$ . Thus we only need to compute  $P_n$  for the case  $t \geq n\tau$ . In this range its analytic form does not change. The work is carried out in Appendix 2, and the result is

where

$$
P_n(0, t) = F_n(t) - F_{n-1}(t),
$$
\n(24)

where  
\n
$$
F_n(t) = e^{-f(t-n\tau)} \{1 + f(t - n\tau) + \cdots + [f(t - n\tau)]^n/n!\}. \quad (25)
$$

If we choose a value of t between  $(s-1)\tau$  and  $s\tau$ .

$$
\bar{n} = sF_s - \sum_{j=0}^{s-1} F_j.
$$
 (26)

<sup>&</sup>lt;sup>11</sup> In a tube or point counter, the recovery time is not constant, and the efficiency increases as recovery proceeds. These matters are discussed in Section 10, but our present purpose is to construct an approximate theory, applicable at low counting rates. When such complications are neglected, we need not retain the factors  $g$  and  $A$  in discussing a constant source, for we may confine our attention to the class of events which would be counted if the recovery time were zero; these events, of course, constitute a Bateman distribution.

#### 8. Counts due to a constant source: count occurs at  $t=0$

The above formulae are not quite what we want for the analysis of experimental data, where it is convenient to know the probability  $P_n'(0, t)$ of  $n$  counts in the interval  $(0, t)$  after an initial *count* at  $t=0$ . Up to time  $\tau$ ,  $P_0' = 1$  because the counter is clogged by the initial count. At time  $\tau$ conditions are the same as at  $t = 0$  in the previous treatment, so that we may write

$$
t \leq n\tau : P_{n}' = 0,
$$
  
\n
$$
n\tau \leq t \leq (n+1)\tau : P_{n}' = 1 - P_{1}' \cdots - P'_{n-1},
$$
  
\n
$$
t \geq (n+1)\tau : P_{n}' = G_{n+1} - G_n, \quad n \neq 0;
$$
  
\n
$$
P_{0}' = e^{-f(t-\tau)}; \quad (27)
$$

where

$$
G_n = e^{-f(t-n\tau)}\{1+f(t-n\tau)+\cdots \text{ we have}
$$
\n
$$
+ [f(t-n\tau)]^{n-1}/(n-1)!\}.
$$
\n(28)\n
$$
dP_n/dt = F_{n-1}P_{n-1} - F_nP_n,
$$
\n(31)

To obtain  $\overline{n}$ , suppose t lies between  $s\tau$  and  $(s+1)\tau$ . Then

$$
P_{s}^{\prime}=1-\sum_{0}^{s-1}P_{n}^{\prime}=1-e^{-f(t-\tau)}-\sum_{1}^{s-1}P_{n}^{\prime},
$$

where the terms in the summation are given by the last line of (27). Therefore

$$
\bar{n} = \sum_{1}^{s-1} n P_n' + s - s e^{-f(t-\tau)} - s \sum_{1}^{s-1} P_n'.
$$

Now,  $\sum_{i=1}^{s-1} P_{n'}$  is a telescoping series whose sum

is 
$$
G_s - G_1
$$
, and  
\n
$$
\sum_{1}^{s-1} n P_n' = \sum_{1}^{s-1} \left[ (n+1)G_{n+1} - nG_n \right] - \sum_{1}^{s-1} G_{n+1};
$$

here the first sum telescopes, and is equal to  $sG_s-G_1$ , so that

$$
\bar{n} = s - \sum_{1}^{s} G_n, \qquad (29)
$$

and by similar methods

$$
\overline{n^2} = s^2 - \sum_{1}^{s} (2n - 1)G_n.
$$
 (30)

For practical purposes, we usually need oniy  $P_0'$ , the fraction of the intervals between adjoining counts which are greater than  $t$ . We may obtain this fraction experimentally for various values of t and plot the results on semi-log paper.<br>Beginning at abscissa  $\tau$ , we have  $\log_{10} P_0' =$  $B = 0.4343f(t - \tau)$ , so the experimental points should lie in the neighborhood of a straight line whose slope is  $-0.4343f$ . Thus f can be determined without a knowledge of  $\tau$ , and the "goodness of fit" of the experimental points can be tested by several methods which need not be explained here.

### 9. General equations governing  $P_n$

 $f(x)$  In setting up general equations for  $P_n$ , one naturally defines a function  $F_n$ , such that  $F_n dt$ is the probability of a count in t to  $t+dt$  when there have been *n* counts between 0 and *t*. Then we have

$$
dP_n/dt = F_{n-1}P_{n-1} - F_nP_n, \tag{31}
$$

the derivation being exactly like that of Eq. (2). However, the  $F$ 's are discontinuous in analytical form, and study of their properties leads one to other equations which will now be obtained directly. Consider the probability of  $n$ counts in time  $t+dt$ . They may be realized in three ways:

- (1) *n* in  $(0, t-\tau)$ ; zero in  $(t-\tau, t)$ ; zero in dt;
- (2)  $n-1$  in  $(0, t-\tau)$ ; zero in  $(t-\tau, t)$ ; 1 in dt;
- (3)  $n-1$  in  $(0, t-\tau)$ ; 1 in  $(t-\tau, t)$ ; zero in dt.

Let  ${}_{n}P_{s}(t-\tau, t)$  be the probability of s counts in  $(t - \tau, t)$ , when *n* have preceded in the interval  $(0, t - \tau)$ . Then a little reflection will show that

$$
P_n(0, t+dt) = P_n(0, t-\tau) \cdot {}_nP_0(t-\tau, t) (1-fdt)
$$
  
+
$$
P_{n-1}(0, t-\tau) \cdot {}_{n-1}P_0(t-\tau, t) fdt
$$
  
+
$$
P_{n-1}(0, t-\tau) \cdot {}_{n-1}P_1(t-\tau, t) (\text{unity}).
$$

Separating the finite terms and the terms containing  $dt$ , we get

$$
P_n(0, t) = P_n(0, t - \tau) \cdot {}_nP_0(t - \tau, t)
$$
  
+ 
$$
P_{n-1}(0, t - \tau) \cdot {}_{n-1}P_1(t - \tau, t); \quad (32)
$$

$$
dP_n(0, t)/dt = fP_{n-1}(0, t-\tau) \cdot {}_{n-1}P_0(t-\tau, t)
$$

$$
-fP_n(0, t-\tau) {}_nP_0(t-\tau, t). \quad (33)
$$

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The evaluation of the functions  $P_0$ , etc., by integration is difficult, so Eqs. (32) and (33) are not very useful as a means of finding the functions  $P_n$ . On the contrary, they enable us to get the functions  ${}_{n}P_{0}$ , etc., from the values of  $P_{n}$ , given in Section 7.

# 10. Distribution of counts in counter with variable recovery time

Skinner<sup>12</sup> has obtained the distribution of recovery times in a counter exposed to a "constant" source. His results are based on Danforth's<sup>13</sup> studies of the way in which the counter voltage V varies when a discharge is initiated, Two classes of counts are considered. In the first, obtained at voltages close to the threshold, V drops with great rapidity to a value below the threshold, and recovers exponentially, with a time constant determined by the capacity of the counter and by the resistance in series with it. The recovery time depends on the time required for  $V$  to rise to the threshold. In the second type, encountered at higher voltages, V falls, recovers to the threshold value and remains close to the threshold for an indefinite period. The mechanism of the second type is not fully understood and we confine our remarks to the first type. Skinner adopts the reasonable view that there is space charge in the counter when effective ionization ceases, and that the voltage goes below the threshold because this'charge is swept to the electrodes. How far V goes below the threshold depends on the voltage existing when the count occurs, and we must refer the reader to Skinner's paper for details. He tacitly assumes that the counter returns to complete efhciency when the voltage rises to the threshold. Of course this is not true, but it is a very desirable simplifying assumption which probably does not affect the results to any great extent at low counting rates.

Bearing in mind that our results depend on these assumptions, let us suppose that the probability of an ionizing particle in  $dt$  is  $fdt$ ,  $f$  being constant. Skinner finds that when the counter is recording such a distribution of events, the probability of a recovery time between  $\tau$  and  $\tau+d\tau$  is

$$
\omega(\tau)d\tau = f(d-1)^{fRC}(d-e^{\tau/RC})^{fRC-1}e^{\tau/RC}d\tau, \quad (34)
$$

where RC is the time constant of the counter, and  $d$  is the maximum voltage drop divided by the difference between operating and threshold voltages; thus  $d$  is a constant which can be determined experimentally. The  $\tau$ 's have all sizes between zero and a certain maximum,  $\tau_{\text{max}}$ , which is  $RC \log_e d$ .

The probability that no count occurs in an interval  $t$ , after a count whose recovery time is between  $\tau$  and  $\tau+d\tau$ , is unity if  $t \leq \tau$ , and  $e^{-f(t-\tau)}$  if  $t \geq \tau$ . Averaging this over all possible recovery times with the aid of Eq. (34), we get the over-all probability  $H_0$ , of no count in an interval t after any count whatsoever. If  $t \geq \tau_{\text{max}}$ , which is the only case of interest here,

$$
H_0 = \int_0^{\tau_{\text{max}}} \omega(\tau) e^{-f(t-\tau)} d\tau = e^{-f(t-\tau'}, \quad (35)
$$
  
where  

$$
e^{f\tau'} = \int_{-\infty}^{\tau_{\text{max}}} \omega(\tau) e^{f\tau} d\tau.
$$

0

This can be evaluated in series, but we only need to note that  $H_0$  is of the same form as  $P_0$ , with  $\tau'$ replacing  $\tau$ . Therefore the introduction of a variable recovery time does not alter the form of the distribution of intervals; but we must remember that variation of efhciency with voltage was neglected.

# D. FLUCTUATIONS IN THE STOCK AND THE DIS-INTEGRATION OF DAUGHTER SUBSTANCES

It was clearly recognized by Adams<sup>14</sup> that the emission from an entire radioactive series has fluctuations which do not obey the Bateman formula, even when the substances in the series are in equilibrium. He showed that if the interval of time for which probabilities are calculated is very short compared with the mean lives of some of the substances, and very long compared with those of others, the fluctuations can be computed approximately in terms of Bateman

<sup>&</sup>lt;sup>12</sup> S. M. Skinner, Phys. Rev. 48, 438 (1935). We are indebted to Dr. Skinner for placing his results at our disposal in advance of publication.

 $13$  Danforth, Phys. Rev. 46, 1026 (1934). The authors have often made visual oscillographic observations which agree in a qualitative way with those of Danforth.

<sup>&</sup>lt;sup>14</sup> N. I. Adams, Phys. Rev. **44**, 651 (1933).

functions. He did this for the alpha-particles of the thorium series, using an interval of 5 minutes which roughly satisfies the above requirements. Adams' analysis refers to the equilibrium case, and he did not consider the inHuence of Huctuations in the stocks of the various substances. We shall now develop methods for dealing with all cases rigorously.

#### 11. Theory of stock fluctuations

At time zero, let a source consist of  $N$  atoms of the parent-substance of a radioactive series whose members are labeled  $A, B, \cdots J$ . The decay constants will be denoted by  $\lambda_A$ ,  $\lambda_B$ ,  $\cdots$ , where the subscript  $A$  belongs to substance  $A$ , and so on. The results of simple disintegration theory give us the probabilities a, b,  $\cdots$  that one of these  $N$  atoms, chosen at random, will be of type  $A, B, \cdots, J$ , respectively, at a later time t. The probability that the stocks of A-atoms, etc., will be  $n_A$ ,  $n_B \cdots$  at time t is

$$
S(n_A, n_B \cdots; t) = \frac{N!}{n_A!, n_B! \cdots n_J!} a^{n_A} b^{n_B} \cdots j^{n_J},
$$
 (36)

and the probability of a stock n of B-atoms, time  $t+dt$ . without regard to the nature of the others, is simply to the nature of the others, is (1) stock = n-1 at t; 1 is formed and 0 decay in dt;<br>
(2) stock = n at t; 0 are formed and 0 decay in dt;<br>
(b) =  $C_n^N b^n (1-b)^{N-n}$ . (37) (3) stock = n+1 at t; 0 are formed and 1 decays in dt

$$
S_n(t) = C_n N b^n (1 - b)^{N - n}.
$$
 (37)

In a series composed of a parent  $A$ , a daughter  $B$ , and an end-product  $C$ , with only the parent present at the start, these probabilities are

$$
a = e^{-\lambda_A t}; \quad b = \frac{\lambda_A}{\lambda_B - \lambda_A} (e^{-\lambda_A t} - e^{-\lambda_B t});
$$

$$
c = \frac{\lambda_B}{\lambda_B - \lambda_A} (1 - e^{-\lambda_A t}) - \frac{\lambda_A}{\lambda_B - \lambda_A} (1 - e^{-\lambda_B t}), \quad (38)
$$

which will be useful farther on. To treat the important case in which the parent  $A$  is very longlived, and is the only substance present initially, we let N approach infinity and  $\lambda_A$  approach zero, keeping  $N\lambda_A$  equal to a constant f. Then a approaches 1 and  $b$  approaches 0, but

$$
Nb \to (f/\lambda_B)(1 - e^{-\lambda_B t}) \equiv x,\tag{39}
$$

$$
S_n(t) = x^n e^{-x}/n!,
$$
\n(40)

x being the "expected" number of B-atoms at time  $t$ . Thus, when  $t$  approaches infinity, so that equilibrium would exist according to simple disintegration theory, the stock Huctuations of B-type atoms are given by  $S_n = (f/\lambda_B)^n e^{-f/\lambda_B}/n!$ Similarly, the stock-Huctuations of all substances except the parent and the end-product obey the formula

$$
S_n = x^n e^{-x}/n!, \qquad (41)
$$

where  $x$  is the "expected" value of the stock.<sup>15</sup>

# 12. Recurrence equations governing stock fluctuations

A differential equation governing the functions  $S_n(t)$  will now be obtained. Consider a given kind of atom, say the kind B, and let  $f_n dt$  and  $p_n dt$  be the probabilities that an atom of this kind wi11 be formed, or will decay, respectively, in the interval dt at t, if the stock of B-atoms is n. (The function  $f_n$  depends on this stock, because the probable stock of the parent  $A$  depends on  $n$ , among other variables.) Now let us consider the value of  $S_n(t+dt)$ . There are three ways<sup>16</sup> in which a stock of  $n$  B-atoms can be realized at

- (1) stock =  $n-1$  at t; 1 is formed and 0 decay in dt;
- (2) stock = n at t; 0 are formed and 0 decay in  $dt$ ;
- 

The probability sought is the sum of three probabilities:

$$
S_n(t+dt) = S_{n-1}(t) f_{n-1} dt + S_n(t) [(1 - f_n dt) (1 - p_n dt)] + S_{n+1}(t) [(1 - f_{n+1} dt) p_{n+1} dt].
$$
 (42)

It follows that

$$
dS_n/dt = f_{n-1}S_{n-1} - (f_n + p_n)S_n + p_{n+1}S_{n+1}.
$$
 (43)

When  $n=0$ , the first term drops out. Because of the presence of  $S_{n+1}$ , this is not a differential

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and

 $*$  <sup>15</sup> When  $t$  approaches infinity, the amount of the parent approaches a value which depends on the value assigned to  $\lambda t$ . Also, the amount of the end product is infinite and fluctuations of these substances have no meaning and no interest

 $<sup>16</sup>$  Cases in which several  $B$ -atoms are formed and several</sup> decay in *dt* can be neglected because they give rise to infinitesimals of higher order than those which must be retained.

equation which we can integrate to get  $S_n$  when we know  $S_{n-1}$ . It is really a recurrence relation for the *seriatim* computation of  $S_1$ ,  $S_2$ , etc., when we know  $S_0$ .

### 13. Fluctuations in the disintegration of a daughter atom and of a series

When a parent substance and its descendants are present in the same source the fluctuations in the total emission do not follow the simple formulas derived in Sections 3 to 6. The problems which arise are very numerous, and when the decay of the parent must be taken into account the algebra is complex. To illustrate as comprehensively as possible the methods of solving such problems, we limit the discussion to a hypothetical three-member series composed of parent, daughter and end-product substances, called A, B, and C, respectively. It is convenient to refer to the disintegrations of parent and daughter as A-emissions and B-emissions, respectively.

The new feature that arises is this: if the stocks of A- and B-atoms are  $n_A$  and  $n_B$  at time zero,  $B$ -emissions in the interval  $(0, t)$  may arise in two ways.  $(1)$  Some of the B-atoms present at time zero may disintegrate, the Huctuations being given by Eq.  $(10)$ ; and  $(2)$  some A-atoms may undergo two disintegrations, thus contributing to the  $B$ -emissions. The problem is to find the Huctuations in the B-emission, and also the Huctuations in the total emission. We shall solve a simple problem in which the decay of the parent stock must be allowed for, and shall then deal with cases in which the parent is very longlived.

Fluctuations when all atoms are initially of  $kind A$ . Let there be  $NA$ -atoms initially. The probability of *n* B-emissions in the interval  $(0, t)$ is the probability that  $n$  of the  $A$ -atoms change into C-atoms in time *t*. This is<br>  $W_{n, B}(0, t) = C_n N c^n (1 - c)^{N-n},$  (44)

$$
W_{n, B}(0, t) = C_n N c^n (1 - c)^{N - n}, \tag{44}
$$

where  $c$  is the probability that any given atom will be of kind  $C$ ; c is given by Eqs. (38). To get the probability of  $n$  emissions from the whole source, we consider a typical case in which  $k A$ atoms go into the B-state and remain there until time t at least, and  $l$  A-atoms go into the C-state; it is understood that  $k+2l=n$ . We sum the

probabilities of all such cases. If  $n$  is even, the result is  $\ddot{\phantom{0}}$ 

$$
W_{n, A+B}(0, t) = \sum_{k} \frac{N!}{(N-k-(n-k)/2)!k!((n-k)/2)!}
$$
  
 
$$
\times a^{N-k-1(n-k)}b^{k}c^{1(n-k)}, \quad (45)
$$

where  $k=0, 2, \cdots n$ . If *n* is odd the summand is the same but  $k = 1, 3, \cdots n$ .

To obtain the Huctuations in B-emission when the parent is very long-lived, and there are no B-atoms initially, we allow  $N$  to approach infinity and  $\lambda_A$  to approach zero in such a way that  $N\lambda_A = f$ . Then (44) is replaced by

$$
W_{n, B} = x^{n} e^{-x} / n!; \quad x = ft - (f/\lambda_{B})(1 - e^{-\lambda_{B}t});
$$
 (46)

x is simply the number of atoms which should pass into the C-state in time  $t$  according to disintegration theory.

Fluctuations of emission by daughter in equi $librium$  with long-lived parent. Suppose that the daughter  $B$  should be in equilibrium with its parent, but that owing to Huctuations the daughter stock at time zero is  $n$ , a value which may or may not be the equilibrium one. Suppose  $k$  of these  $B$ -atoms emit in time  $t$ , and that  $l$  of the parent atoms change into C-atoms; and let  $k+l=s$ . By using (40) and (46), and replacing  $\lambda_B$  by  $\lambda$  for simplicity, the probability of this composite event is

$$
S_n C_k^{n} (e^{\lambda t} - 1)^k e^{-n\lambda t} x^l e^{-x} / l! \qquad (47)
$$

To get the total probability of  $s$  B-emissions we must sum (47) over all allowable values of  $n$ and  $k$ . Obviously,  $n$  must be at least equal to k so n runs from k to  $\infty$  and k runs from 0 to s. Replacing l by  $s-k$ , and n by  $m+k$ , we have, after some reductions,

$$
W_{s, B} = \frac{e^{-f/\lambda}e^{-x}}{s!} \sum_{k=0}^{s} C_k^s \left(\frac{f}{\lambda}\right)^k
$$

$$
\cdot \left(\frac{1-e^{-\lambda t}}{x}\right)^k x^s \sum_{m=0}^{\infty} \frac{(fe^{-\lambda t}/\lambda)^m}{m!}
$$

$$
= (ft)^s e^{-f t}/s!
$$
(48)

Thus the emission of the daughter atom fluctuate  $\begin{aligned} \n\mathbf{y} &= (ft)^s e^{-ft}/s! \tag{48} \n\end{aligned}$ <br>
Thus the emission of the daughter atom fluctuates<br>
in the same way as that of the long-lived parent.<sup>17</sup><br>  $\begin{aligned} \n\mathbf{y} &= \mathbf{y} \mathbf{y} \mathbf{y} + \mathbf{y} \mathbf{y} \mathbf{y} + \mathbf{y} \mathbf{y} \mathbf$ 

 $17$  The simplicity of this result is surprising, when we consider that Eq. (48) results from stock-variations and

Fluctuations of total emission under equilibrium conditions. It must not be supposed that the total emission of parent and daughter follows the Bateman formula. In Section 6 we remarked that the superposition of *independent* sources fluctuating according to the Bateman formula gives rise to a distribution of events obeying that formula, but in the present instance the sources are not independent; fluctuations in the emission by the parent have a direct influence on the subsequent emission of the daughter. To illustrate this, if for a time the parent disintegrates at a rate higher than the average the stock of the daughter will be raised and it will emit at a rate higher than average. Before working out the general case, let us clarify our ideas by getting  $W_0$ , the chance of no emission by both parent and daughter in time  $t$ . Now, the probability that (1) there are  $n \text{ } B$ -atoms at time zero; (2) no A-atoms disintegrate in time  $t$ ; and (3) no B-atoms disintegrate in time t, is  $S_n e^{-ft} e^{-nkt}$ , and averaging over  $n$ ,

$$
W_0 = e^{-ft - (f/\lambda)(1 - e^{-\lambda t})}.
$$
 (49)

When  $\lambda t$  approaches zero, this function behaves like  $e^{-2ft}$ , and when  $\lambda t$  is large compared with unity, it behaves like  $e^{-ft}$ . It is instructive to consider the limiting case in which the life of the daughter is very short compared with the average interval between disintegrations of parent atoms. Then nearly all the events occur in close pairs. The average spacing of the events composing a pair is of the order  $1/\lambda$ , and the longer intervals between pairs are distributed approximately according to the law  $e^{-ft}$ , being governed mainly by disintegrations of the parent atoms.

Now we shall obtain the probability of s emissions by both parent and daughter. Consider the case in which there are  $n \, B$ -atoms at time 0, and in time t, (1) k B-atoms disintegrate; (2)  $l A$ atoms undergo one disintegration each; (3) m A-atoms undergo two disintegrations each. We first require the probability that both the events (2) and (3) occur. Consider a source containing  $N A$ -atoms with finite decay constant, and pass to the limit  $N \rightarrow \infty$ ,  $\lambda_A \rightarrow 0$ ,  $N\lambda_A = f$ . The probability that both (2) and (3) occur is

$$
[N!/(N-l-m)!l!m!]a^{N-l-m}b^lc^m,
$$

where  $a$ ,  $b$ , and  $c$  are given by Eqs. (38), and in the limit this becomes

$$
e^{-ft}y^lx^m/l!m!,\tag{50}
$$

where y is an abbreviation for  $(f/\lambda)(1-e^{-\lambda t})$ ,  $x=ft-\gamma$ , and  $\lambda$  is written in place of  $\lambda_B$ . Finally, replacing  $l$  by  $s-k-2m$ , the total probability of s disintegrations by both parent and daughter is, when s is even,

$$
W_{s, A+B} = \sum_{m=0}^{s/2} \sum_{k=0}^{s-2m} \sum_{n=k}^{\infty} S_n C_k^{n} (e^{\lambda t} - 1)^k e^{-n\lambda t}
$$
  
 
$$
\times e^{-f} y^{s-k-2m} x^m / (s - k - 2m) \, \text{Im} \, \text{I}.
$$

Putting  $n=k+j$  for convenience, the result is this:

$$
W_{s, A+B} = (2y)^{s} e^{-ft - y} \sum_{m=0}^{s/2} (x/4y^2)^m / (s - 2m) \ln l. (51)
$$

When s is odd, m runs from 0 to  $(s-1)/2$ .

The results of this section can be tested in a very direct way. Suppose we prepare a source of an alpha-raying substance whose daughter is an alpha-rayer of short life, adjusting the strength so that both parent and daughter give only a few particles per hour when equilibrium is attained. Using a tube electrometer, the particles emitted by these two substances can be distinguished from those due to their descendants, and also from each other. Thus we can obtain all the data needed for tests of Eqs. (48) and (51), and also of Eq. (10).

In conclusion, the formulae derived in this paper are sufhcient in scope to facilitate the solution of a wide variety of fluctuation problems. To keep the paper within bounds, we have omitted applications to experimental data and have made no mention of Huctuation-velocities, an important though neglected subject. It is a p]easure to acknowledge our indebtedness to Professor Forrest Western, who collaborated in experimental work which suggested the writing of this paper.

emission-Huctuations of the kind considered above. The reasoning has been checked by passing to the limit after setting up the problem for the case of a parent having a finite decay constant.

#### APPENDIX 1. SOLUTION OF EQ. (20)

The problem is to find the quantities  $v_s$  from the equations

$$
\sum_{s=n}^{N} C_n^s v_s = p_n; \quad n = 0, 1, \cdots N. \tag{20}
$$

If we differentiate the identity

$$
(a+b)^s = \sum_{n=0}^s C_n^s a^{s-n} b^n
$$

with respect to  $b$ ,  $j$  times, we get

$$
s(s-1) \cdots (s-j+1)(a+b)^{s-j}
$$
  
= 
$$
\sum_{n=j}^{s} C_n s a^{s-n} n(n-1) \cdots (n-j+1) b^{n-j}
$$

Putting  $a = 1$ , and  $b = -1$ , we find that

$$
\sum_{n=j}^{s} C_n{}^{s} C_j{}^{n} (-1)^{n-j} = \delta_{sj}
$$

where  $\delta_{sj}$  is 0 if  $s \neq j$  and 1 if  $s = j$ . This suggests that to get  $v_i$ , we should multiply the typical equation in (20) by  $C_i^{n}(-1)^{n-j}$ . (Since  $C_i^{n}$  has no meaning if n is less than j, we begin, of course, with the jth equation.) Doing this and adding the results we have

$$
\sum_{n=j}^{N} \sum_{s=n}^{N} C_n {}^{s}C_j {}^{n}(-1)^{n-j}v_s = \sum_{n=j}^{N} C_j {}^{n}(-1)^{n-j}p_n.
$$

Interchanging the order of summation on the left, we have

$$
\sum_{s=j}^{N} v_s \sum_{n=j}^{s} C_n {}^{s}C_j {}^{n}(-1)^{n-j}.
$$

By the identity derived above, the coefficient of  $v_s$  vanishes unless  $s = j$ , and we have

$$
v_j = \sum_{n=j}^{N} C_j^{n} (-1)^{n-j} p_n.
$$

APPENDIX 2. COUNTS IN <sup>A</sup> DETECTOR WITH CONSTANT RECOVERY TIME,  $\tau$ .

Let the counter receive, on the average,  $f$  events per second, distributed according to the Bateman formula. We wish to obtain  $P_n$ , the probability that *n* counts occur in the interval  $(0, t)$ . The counter is not clogged at  $t=0$ ; and in accord with Section 7 we are interested only in the case where  $t \geq n\tau$ . We shall call a period during which the



counter is clogged an interlude. Two cases must be considered, as shown in Fig. 5. In Case 1, the nth interlude ends before the instant  $t$ , and in Case 2, it extends beyond this instant.

We begin by writing the probability that the  $n$  counts will occur, respectively, in the intervals  $(s_1, s_1+d s_1);$  $(s_2, s_2+d s_2), \cdots, (s_n, s_n+d s_n),$  where it is understood, of course, that  $s_2$  is greater than or equal to  $s_1+\tau$ , and so forth. In Case 1, this probability is  $e^{-fs}$   $fds_1e^{-f(s_2-s_1-\tau)}f ds_2$  $e^{-f(s_n-s_{n-1}-\tau)}f ds_n e^{-f(t-s_n-\tau)} = e^{-f(t-n\tau)}f^n ds_1 \cdots ds_n$ . This must be integrated for all possible positions of the counts that can occur in Case 1, ranging from the case in which all the interludes are closely packed at the left in Fig. 5, upper diagram, to that in which they are closely packed at the right, with the end of the nth interlude at the instant t. For example, the limits for  $s_1$  are 0 and  $s_2-r$ ; for  $s_{n-1}$  they are  $(n-2)\tau$  and  $s_n-\tau$ ; and for  $s_n$  they are  $(n-1)\tau$  and  $t-\tau$ .

Similarly, in Case 2, the probability of having counts in  $ds_1$  at  $s_1$ , etc., is

$$
e^{f(n-1)\tau}e^{-f s_n}f^n ds_1\cdots ds_n,
$$

and the limits are the same as before, except for  $s_n$ , whose limits are now  $t-\tau$  to t. Therefore

$$
P_n = e^{-f(t-n\tau)} \cdot \int_{(n-1)\tau}^{t-\tau} \int_{(n-2)\tau}^{s_n-\tau} \cdots \int_{\tau}^{s_3-\tau} \int_{0}^{s_2-\tau} \int_{0}^{n} ds_1 \cdots ds_n
$$
  
+  $e^{f(n-1)\tau} \int_{t-\tau}^{t} \int_{(n-2)\tau}^{s_n-\tau} \cdots \int_{\tau}^{s_3-\tau} \int_{0}^{s_2-\tau} e^{-f s_n f^n} ds_1 \cdots ds_n$ 

The substitution  $x_i = f[s_i - (i-1)\tau]$  makes it simple to carry out the integration over  $s_1$ ,  $s_2 \cdots s_{n-1}$ , and the result is  $f^{n-1}[s_n - (n-1)\tau]^{n-1}/(n-1)!$ , so finally

$$
P_n = e^{-f(t-n\tau)} f^n \int_{(n-1)\tau}^{t-\tau} [s_n - (n-1)\tau]^{n-1} / (n-1)! \cdot ds_n
$$
  
+  $e^{f(n-1)\tau} f^n \int_{t-\tau}^t e^{-f s_n} [s_n - (n-1)\tau]^{n-1} / (n-1)! ds_n$   
=  $e^{-f(t-n\tau)} [1 + f(t-n\tau) + \dots + f^n (t-n\tau)^n / n!]$   
-  $e^{-f(t-(n-1)\tau)} [1 + f(t-(n-1)\tau) + \dots + f^{n-1} (t-(n-1)\tau)^{n-1} / (n-1)!]$ ,

which is Eq. (24).