

## On the Correlation Method for the Determination of the Absolute Yield of Nuclear Reactions

V. I. GOL'DANSKII AND M. I. PODGORETSKII

*P. N. Lebedev Physical Institute, Academy of Sciences, USSR*

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An examination has been made of the statistical method of investigating nuclear transformations and of determining their absolute yield and mechanism. The method is based on measurement of the magnitude of the correlation function between the observed transformations of various types. The correlation function and its dispersion have been calculated (in the absence and in the presence of background), and the question of the accuracy of the method has been examined. The well-known method of coincidences is a special case of the correlation method, which is also applicable for longer time intervals between related transformations at high radiation intensities.

### 1. INTRODUCTION

ONE of the important methods of experimental physics of the last few years has been the method of coincidences, based on the recording (by means of special circuits) of nuclear particles emitted in an individual nuclear transformation or in several successive transformations separated by very short time intervals. Coincidence circuits are used both in those cases when an angular correlation exists between the recorded particles, and also in the absence of such a correlation. In particular, the recording of the coincidences permits one to determine the absolute number of such transformations independently of the efficiency with which they are recorded. An example of such a kind is afforded by the well-known method of  $\beta\gamma$  and  $\gamma\gamma$  coincidences<sup>1</sup> which may be illustrated by the following very simple case.

If in the course of the radioactive decay of an atom of a given isotope (for example,  $\text{Al}^{28} \xrightarrow{\beta^-} \text{Si}^{28*} \xrightarrow{\gamma} \text{Si}^{28}$ ) one  $\beta$ -particle and one  $\gamma$ -quantum are emitted, then, if  $M$  individual disintegrations occur in the course of the experiment in the absence of background, the  $\beta$ -counter will record

$$M_{\beta} = g_{\beta} M \quad \beta\text{-particles,} \quad (1)$$

while the  $\gamma$ -counter will record

$$M_{\gamma} = g_{\gamma} M \quad \gamma\text{-quanta} \quad (2)$$

and the coincidence circuit will record

$$M_{\beta\gamma} = g_{\beta} g_{\gamma} M + \frac{2\tau}{T} g_{\beta} g_{\gamma} (1 - g_{\beta}) \quad (3)$$

$\times (1 - g_{\gamma}) M^2$  coincidences.

Here  $g_{\beta}$  and  $g_{\gamma}$  are unknown efficiencies of recording, and  $\tau$  is the resolving time of the coincidence circuit. The first term in Eq. (3) characterizes the recording of true counts, the second term characterizes the recording of chance coincidences determined by the disintegration being investigated. In a practical application of the method of  $\beta\gamma$  and  $\gamma\gamma$  coincidences the quantity  $\tau$  is usually chosen to be so small that the second term in Eq. (3) is negligible, and

$$M_{\beta\gamma} \approx g_{\beta} g_{\gamma} M. \quad (4)$$

From Eqs. (1)-(3) it follows that the true number of individual radioactive disintegrations may be determined from

$$M = M_{\beta} M_{\gamma} / M_{\beta\gamma}. \quad (5)$$

Knowing the value of  $M$ , one can then determine the efficiencies of recording the disintegrations, and also the cross section of the nuclear reaction leading to the formation of the given isotope if the flux of the activating radiation is known (or, conversely, one can determine the flux of the activating radiation from the known cross section of the nuclear reaction).

The recording of coincidences is also utilized, as is well known, for the investigation of the mechanism of nuclear disintegration. A number of appropriate examples is given by Mitchell<sup>1</sup>. The above illustrates sufficiently clearly the possibilities of the method of coincidences because of which it has received wide acceptance.

<sup>1</sup> A. C. G. Mitchell, Rev. Mod. Phys. **20**, 296 (1948).

Let us now turn to a description of the essence of the method of correlations in which, instead of measuring the number of coincidences, one should measure the correlation function between the counts of the various radiations being recorded. This method can yield exactly the same results as the method of coincidences and represents an alternative experimental possibility of obtaining quantities which are presently determined by means of coincidence circuits.

However, it is important to note that in the method of correlation the time interval between successive disintegrations may be in principle arbitrarily large. Its allowable magnitude is determined only by practical considerations, and may be as large as one second. This circumstance provides the main advantage of the method of correlations which allows it to be used even in those cases when the method of coincidences is quite inapplicable because of the overloading of the coincidence circuit.

## 2. GENERAL BASIC ASSUMPTIONS. DEFINITION OF THE CORRELATION FUNCTION AND THE CORRELATION COEFFICIENT. THE THEORY OF THE METHOD.

Let us consider the chain of daughter products  $B, C, D, \dots, L$ , the first of which is formed either by the decay of a radioactive substance  $A$ , the amount of which practically does not change during the experiment, or as a result of a nuclear reaction occurring when a target is irradiated externally by particles of some kind. Then the probability  $P_{bc\dots l}$  of finding at some instant of time  $t$ ,  $n_b, n_c, n_d, \dots, n_l$  atoms, respectively, of the various types is determined by the many-dimensional Poisson law:

$$P_{bc\dots l} = \frac{e^{-\bar{B}} \bar{B}^{n_b}}{n_b!} \frac{e^{-\bar{C}} \bar{C}^{n_c}}{n_c!} \dots \frac{e^{-\bar{L}} \bar{L}^{n_l}}{n_l!}, \quad (6)$$

where  $\bar{I}$  is the mean number determined by the laws of radioactive decay of atoms of the kind  $I$  which exists at the time  $t$  for a given intensity of the primary radiation.

From (6) follow the relations between the dispersions and the mean values of the products:

$$D \left( \sum_{i=B}^{i=L} n_i \right) = \sum_{i=B}^{i=L} D(n_i), \quad (7)$$

$$\prod_{i=B\dots L} (n_i) = \prod_{i=B\dots L} (\bar{n}_i), \quad (8)$$

where  $n_i$  is the number of atoms which are found

at the given instant of time in the state  $i$ .

The many-dimensional Poisson distribution in this case follows physically from the complete statistical independence of the decay of the individual atoms; each of the atoms may be found at the time  $t$  in some one definite state independently of the behavior of the others, and in virtue of the lack of limitation on the total number of atoms of the original substance, the presence of a certain number of atoms in one state does not impose any restrictions on the possible quantities of atoms in other states. The complete statistical independence of the decay of individual atoms which makes it possible to apply Eqs. (6) to (8) is realized, in particular, in an equilibrium radioactive mixture.

Let us now turn to the relations between the disintegrations of various kinds observed in such mixtures. Let us denote by  $m_i$  the actual number of disintegrations of the  $i$ th type during a certain time interval, and by  $r_i$  the recorded number of such disintegrations, with

$$r_i = g_i \bar{m}_i, \quad (9)$$

where  $g_i$  is the efficiency of recording the disintegrations of the  $i$ th kind. Evidently each of the quantities  $m_i$  and  $r_i$  is distributed in accordance with the one-dimensional Poisson law, but relations of the type (7) and (8) are no longer applicable, for here the events are no longer independent--the same atom may, during the course of the experiment, undergo disintegrations of the  $i$ th,  $(i+1)$ th,  $\dots$   $k$ th type. In this case

$$D(m_l + m_k) = D(m_l) \quad (10)$$

$$+ D(m_k) + 2(\overline{m_l m_k} - \bar{m}_l \bar{m}_k),$$

$$D(r_l + r_k) = D(r_l) \quad (10')$$

$$+ D(r_k) + 2(\overline{r_l r_k} - \bar{r}_l \bar{r}_k).$$

The last terms in (10) and (10')--the so-called correlation functions--play a determining role in the proposed method. Thus the problem arises of the calculation of the correlation functions

$$\Phi_{lk} = \overline{r_l r_k} - \bar{r}_l \bar{r}_k \quad (11)$$

or of the correlation coefficient

$$R_{lk} = \frac{\overline{r_l r_k} - \overline{r_l} \cdot \overline{r_k}}{\sqrt{D(r_l) D(r_k)}} = \frac{\overline{r_l r_k} - \overline{r_l} \cdot \overline{r_k}}{\sqrt{\overline{r_l r_k}}}. \quad (12)$$

Because of its importance for the following let us begin with an examination of the case when  $g_l = g_k = 1$  which is seldom met with in practice, but which demonstrates the principle most clearly.

It is evident that the correlation functions and the correlation coefficients are essentially different in the cases when the radiation of the  $l$ th and  $k$ th type (i.e., the disintegrations of the  $l$ th and  $k$ th type) occur independently of one another, or when, on the contrary, radiation of one type (for example of the  $l$ th type, or the  $l$ th disintegration) is necessarily accompanied at the same instant or later by radiation of another kind (for example, of the  $k$ th kind, i.e., by the  $k$ th disintegration).

If the emissions of the  $l$ th and the  $k$ th radiation are independent of one another (for example, in the parallel disintegration in a radioactive chain), naturally

$$\Phi_{lk} = R_{lk} = 0. \quad (13)$$

A different situation will exist in the case of related disintegrations. Let us suppose for the sake of simplicity that the length of the chosen time interval  $t$  for the measurement greatly exceeds the mean time interval between the related disintegrations of the  $l$ th and  $k$ th type (in the case of a chain of radioactive transformations).

$$\sum_{i=l+1}^k \frac{1}{\lambda_i t} \ll 1. \quad (14)$$

Then each disintegration of the  $l$ th type is necessarily accompanied by a disintegration of the  $k$ th type, and conversely, i.e.,  $\overline{m}_l = \overline{m}_k = m$ . It then follows that

$$\Phi_{lk} = \overline{m_l m_k} - \overline{m_l} \overline{m_k} = \overline{m^2} - (\overline{m})^2 = \overline{m} \neq 0, \quad (15)$$

$$R_{lk} = \frac{\overline{m}}{D(m)} = 1, \quad (16)$$

which could have been written down immediately since we are dealing with completely correlated random variables.

Thus it is evident that the measurement of the correlation coefficient allows one to distinguish between the cases of mutually unrelated and related disintegrations. The related disintegrations may be either simultaneous, or separated by con-

siderable intervals of time. However, for the sake of generality of treatment, we shall consider in the following the case of two related disintegrations, the  $l$ th disintegration (emission of particles of the  $l$ th type) preceding in time the  $k$ th disintegration (emission of particles of the  $k$ th type). Let us now examine the case when  $g_l$  and  $g_k$  differ from unity. If the  $l$ th and the  $k$ th disintegrations are independent, then evidently Eq. (13) remains valid. However, if these disintegrations are related, then among the  $r_l$  disintegrations of the  $l$ th type recorded in the time interval under consideration there will be registered  ${}_k r_l$  such disintegrations which also give rise to disintegrations of the  $k$ th type within the same interval (irrespective of whether these disintegrations are recorded or not) and  $r'_l$  such disintegrations which are not followed by disintegrations of the  $k$ th type.

In its turn the number  $r_k$  of recorded disintegrations of the  $k$ th type may be divided into  ${}_l r_k$ —the number of disintegrations of the  $k$ th type originating from the disintegrations of the  $l$ th type (irrespective of whether they were recorded or not) which occurred in the same time interval, and  $r'_k$ —the number of disintegrations of the  $k$ th type which were not preceded by a disintegration of the  $l$ th type within the given time interval. If one denotes by  ${}_k m_l$  not merely the recorded but the total number of disintegrations of the  $l$ th type in the given time interval which were followed by disintegrations of the  $k$ th type, and by  ${}_l m_k$  the total number of disintegrations of the  $k$ th type within the given interval which were preceded by disintegrations of the  $l$ th type, then it is evident that

$${}_k m_l = {}_l m_k = m_{kl}, \quad (17)$$

$$\overline{{}_k r_l} = g_l \overline{m_{kl}}, \quad (18)$$

$$\overline{{}_l r_k} = g_k \overline{m_{kl}}. \quad (18')$$

In the expressions

$$r_l = {}_k r_l + r'_l \quad (19)$$

and

$$r_k = {}_l r_k + r'_k, \quad (19')$$

only the terms  ${}_k r_l$  and  ${}_l r_k$  are mutually dependent, while all the others are independent of one another.

From all the above we obtain

$$\begin{aligned} \Phi_{lk} &= \overline{r_l r_k} - \overline{r_l} \overline{r_k} = \overline{{}_k r_l r_k} - \overline{{}_k r_l} \overline{{}_l r_k} = \\ &= g_l g_k \overline{m_{kl}^2} - g_l g_k (\overline{m_{kl}})^2 = g_l g_k \overline{m_{kl}}, \end{aligned} \quad (20)$$

since for the quantity  $m_{kl}$  we have

$$D(m_{kl}) = \overline{m_{kl}}. \tag{21}$$

In what follows below we shall take the length of the time interval  $t$  such that Eq. (14) is satisfied, and then in the absence of branching between disintegrations of the  $l$ th and  $k$ th type

$$\overline{m_{kl}} = \overline{m_l} = \overline{m_k} = \overline{m}, \tag{22}$$

from which we obtain for the correlation function and the correlation coefficient

$$\Phi_{lk} = g_l g_k \overline{m}, \tag{23}$$

$$R_{lk} = g_l g_k \overline{m} \sqrt{g_l \overline{m} g_k \overline{m}} = \sqrt{g_l g_k}. \tag{24}$$

We note that the expressions for the correlation function and for the number of true coincidences in a given time interval are the same. It may be easily shown that the corresponding expressions are analogous also in the presence of an angular correlation of the radiations.

Thus, if, instead of determining the numbers of disintegrations  $M_l$  and  $M_k$  that have taken place in the course of the whole experiment, and the number of coincidences  $M_{lk}$ , we determine the average number of disintegrations  $\overline{r_l}$  and  $\overline{r_k}$  recorded during one interval, and the correlation function  $\Phi_{lk}$ , we can solve all the problems which can be solved by the method of coincidences; in particular we can determine the absolute number  $\overline{m}$  of disintegrations during one interval from the relation

$$\overline{m} = \overline{r_l r_k} / \Phi_{lk}, \tag{5'}$$

wholly analogous to Eq. (5). This constitutes the essence of the correlation method.

In examining the practical applicability of the correlation method two questions arise. First of all we must determine the accuracy with which the correlation coefficient can be measured. If  $g_l$  and  $g_k$  are small, then the question of the accuracy of determination of  $R_{lk}$  becomes particularly important, for the quantity  $R_{lk}$ , in accordance with Eq. (24), cannot differ appreciably from  $R_{lk} = 0$  which corresponds to the case of two mutually independent processes. The second important question is the elucidation of the role which the background plays in measurements using the correlation method. Therefore, let us turn now to the examination of the accuracy of the correlation

method both in the absence and in the presence of background.

### 3. DISPERSION OF THE CORRELATION FUNCTION. COMPARISON OF THE CORRELATION METHOD WITH THE METHOD OF COINCIDENCES.

In accordance with the definition (11) of the correlation function its value obtained as a result of measurements in  $N$  intervals is:

$$\Phi_{lk} = \frac{\sum_{i=1}^N r_{li} r_{ki}}{N} - \frac{\sum_{i=1}^N r_{li}}{N} \frac{\sum_{i=1}^N r_{ki}}{N}. \tag{25}$$

It is evident that

$$\overline{\Phi_{lk}} = \frac{\overline{\sum r_{li} r_{ki}}}{N} - \frac{\overline{\sum r_{li}} \overline{\sum r_{ki}}}{N^2}, \tag{26}$$

$$\overline{\Phi_{lk}^2} = \frac{\overline{(\sum r_{li} r_{ki})^2}}{N^2}. \tag{27}$$

$$- 2 \frac{\overline{\sum r_{li} r_{ki} \sum r_{li} \sum r_{ki}}}{N^3} + \frac{(\overline{\sum r_{li} \sum r_{ki}})^2}{N^4}$$

(here and in the following, in writing the summation signs we shall omit the summation index  $i = 1, \dots, N$ ). For further calculations we shall need the following equations

$$\overline{\sum r_{li} r_{ki}} = N \overline{r_l r_k}; \tag{28}$$

$$\overline{\sum r_{li} \sum r_{ki}} = N \overline{r_l r_k} + N(N-1) (\overline{r_l} \overline{r_k}); \tag{29}$$

$$\overline{(\sum r_{li} r_{ki})^2} = N \overline{r_l^2 r_k^2} + N(N-1) (\overline{r_l r_k})^2; \tag{30}$$

$$\overline{\sum r_{li} r_{ki} \sum r_{li} \sum r_{ki}} = N \overline{r_l^2 r_k^2} \tag{31}$$

$$+ N(N-1) [\overline{r_l r_l r_k^2} + \overline{r_k r_k r_l^2} + (\overline{r_l r_k})^2]$$

$$+ N(N-1)(N-2) \overline{r_l} \overline{r_k} \overline{r_l r_k};$$

$$\overline{(\sum r_{li} \sum r_{ki})^2} = N \overline{r_l^2 r_k^2} + N(N-1) \overline{r_l^2} \overline{r_k^2} \tag{32}$$

$$+ 2N(N-1) [\overline{r_l} \overline{r_l r_k^2} + \overline{r_k} \overline{r_k r_l^2} + (\overline{r_l r_k})^2].$$

$$+ N(N-1)(N-2)(N-3) (\overline{r_l})^2 (\overline{r_k})^2$$

$$+ N(N-1)(N-2) [\overline{r_l^2} (\overline{r_k})^2$$

$$+ \overline{r_k^2} (\overline{r_l})^2 + 4 \overline{r_l} \overline{r_k} \overline{r_l r_k}].$$

Equations (28)-(32) are considerably simplified if the disintegrations of the  $l$ th and  $k$ th type are independent, for in that case,

$$\overline{r_l^2 r_k^2} = \overline{r_l^2} \overline{r_k^2}, \quad (33)$$

$$\overline{r_l r_l r_k^2} = (\overline{r_l})^2 \overline{r_k^2}, \quad (33')$$

$$\overline{r_k r_k r_l^2} = (\overline{r_k})^2 \overline{r_l^2}, \quad (33'')$$

and

$$(\overline{r_l r_k})^2 = (\overline{r_l})^2 (\overline{r_k})^2. \quad (33''')$$

In this case  $\overline{\Phi_{lk}} = 0$ , and

$$\begin{aligned} \overline{\Phi_{lk}^2} = \frac{N-1}{N} [\overline{r_l^2 r_k^2} - (\overline{r_l})^2 \overline{r_k^2} - (\overline{r_k})^2 \overline{r_l^2} \\ + (\overline{r_l})^2 (\overline{r_k})^2] = \frac{N-1}{N^2} \overline{r_l r_k}. \end{aligned} \quad (34)$$

Thus the dispersion of the correlation function in the case of independence of disintegrations of the  $l$ th and  $k$ th type is given by

$$\begin{aligned} D_{\text{indep}}(\Phi_{lk}) = \overline{\Phi_{lk}^2} = \frac{N-1}{N^2} \overline{r_l r_k} \\ = \frac{N-1}{N^2} g_l g_k (\overline{m})^2. \end{aligned} \quad (35)$$

In the case of related disintegrations (we shall assume here that there is no branching between these disintegrations) it follows from (22), (26), (28) and (29) that the average value of the correlation function is equal to

$$\overline{\Phi_{lk}} = \frac{N-1}{N} (\overline{r_l r_k} - \overline{r_l} \overline{r_k}) = \frac{N-1}{N} g_l g_k \overline{m}. \quad (36)$$

For  $N \rightarrow \infty$  we obtain the "theoretical" mean value  $g_l g_k \overline{m}$ . For the determination of the mean square value  $\overline{\Phi_{lk}^2}$  we must obtain the values

$\overline{r_l^2 r_k^2}$ ,  $\overline{r_l^2 r_k}$  and  $\overline{r_l r_k^2}$ . We shall explain the method

of obtaining these quantities by using

$\overline{r_l r_k^2}$  as an example, and by noting first of all that for the determination of the correlation function we are interested only in those disintegrations of the  $l$ th type which are followed by disintegrations of the  $k$ th type in the given interval, and only in those disintegrations of the  $k$ th type which are preceded in the given interval by disintegrations of the  $l$ th type. In satisfying condition (14) we can consider, neglecting distortions near the boundary of the interval, that each disintegration of the  $l$ th type necessarily leads to a disintegration of the  $k$ th type in the same interval.

Let the values of  $r$  at a time  $t$  at the end of the interval be, respectively,  $r_l$  and  $r_k$ . Let us add an infinitesimal time  $dt$  at the beginning of the interval. Then we shall have the probability  $adt$ , where  $a$  is the equilibrium activity of all the members of the series, that during this infinitesimal time a disintegration of the type  $l$  will take place necessarily followed by a disintegration of the type  $k$  within the interval, while  $1-adt$  gives us the probability that a disintegration of the type  $l$  will not occur. Therefore,

$$\begin{aligned} \overline{r_l r_k^2}(t+dt) &= \overline{r_l r_k^2}(t) + \frac{d \overline{r_l r_k^2}}{dt} dt \\ &= (1-adt) \overline{r_l r_k^2} + adt \{g_l g_k (\overline{r_l + 1})(\overline{r_k + 1})^2 \\ &\quad + g_l (1-g_k) (\overline{r_l + 1}) \overline{r_k^2} + g_k (1-g_l) \overline{r_l} (\overline{r_k + 1})^2 \\ &\quad + (1-g_l)(1-g_k) \overline{r_l r_k^2}\}. \end{aligned} \quad (37)$$

From this it follows that

$$\begin{aligned} \frac{d \overline{r_l r_k^2}}{dt} &= 2a g_k \overline{r_l r_k} + a g_l \overline{r_k^2} \\ &\quad + a g_k \overline{r_l} + 2a g_l g_k \overline{r_k} + a g_l g_k. \end{aligned} \quad (38)$$

Similarly

$$\begin{aligned} \frac{d \overline{r_k r_l^2}}{dt} &= 2a g_l \overline{r_l r_k} + a g_k \overline{r_l^2} \\ &\quad + a g_l \overline{r_k} + 2a g_l g_k \overline{r_l} + a g_l g_k, \end{aligned} \quad (39)$$

$$\begin{aligned} \frac{d \overline{r_l^2 r_k^2}}{dt} &= 2a g_k \overline{r_k r_l^2} + 2a g_l \overline{r_l r_k^2} \\ &\quad + a g_k \overline{r_l^2} + a g_l \overline{r_k^2} + 4a g_l g_k \overline{r_l r_k} + \\ &\quad + 2a g_l g_k \overline{r_l} + 2a g_l g_k \overline{r_k} + a g_l g_k. \end{aligned} \quad (40)$$

Integration of Eqs. (38)-(40) gives

$$\begin{aligned} \overline{r_l^2 r_k^2} &= g_k^2 g_l (\overline{m})^3 + 2g_k^2 g_l (\overline{m})^2 \\ &\quad + g_l g_k (\overline{m})^2 + g_l g_k \overline{m}, \end{aligned} \quad (41)$$

$$\begin{aligned} \overline{r_k r_l^2} &= g_l^2 g_k (\overline{m})^3 + 2g_l^2 g_k (\overline{m})^2 \\ &\quad + g_l g_k (\overline{m})^2 + g_l g_k \overline{m}, \end{aligned} \quad (42)$$

$$\overline{r_l^2 r_k} = g_l^2 g_k (\overline{m})^4 + 4g_l^2 g_k (\overline{m})^3 + \quad (43)$$

$$\begin{aligned}
& + g_l^2 g_k (\bar{m})^3 + g_l g_k^2 (\bar{m})^3 \\
& + 2g_l^2 g_k^2 (\bar{m})^2 + 2g_l^2 g_k (\bar{m})^2 \\
& + 2g_l g_k^2 (\bar{m})^2 + g_l g_k (\bar{m})^2 + g_l g_k \bar{m}.
\end{aligned}$$

We can now turn to the direct calculation of the mean square of the correlation function which gives

$$\begin{aligned}
\Phi_{lk}^2 &= \frac{N-1}{N} g_l^2 g_k^2 (\bar{m})^2 \\
& + \frac{N-1}{N^2} g_l g_k (\bar{m})^2 + \frac{(N-1)^2}{N^3} g_l g_k \bar{m}.
\end{aligned} \quad (44)$$

The dispersion of the correlation function is given by

$$\begin{aligned}
D(\Phi_{lk}) &= \Phi_{lk}^2 - (\bar{\Phi}_{lk})^2 \\
&= \frac{N-1}{N^2} \left[ 1 + g_l g_k + \frac{N-1}{N\bar{m}} \right] g_l g_k (\bar{m})^2,
\end{aligned} \quad (45)$$

where  $N$  is the number of intervals in which readings are taken,  $\bar{m}$  is the mean value of disintegration of each kind during one interval. The relative error in determining  $\Phi_{lk}$  is

$$\begin{aligned}
\delta(\Phi_{lk}) &= \sqrt{D(\Phi_{lk})/\bar{\Phi}_{lk}} \\
&= \sqrt{\frac{1}{N-1} + \frac{1}{g_l g_k N \bar{m}} + \frac{1}{g_l g_k (N-1)}}.
\end{aligned} \quad (46)$$

If  $\bar{m} \gg 1$  and  $N \gg 1$ , then

$$\delta(\Phi_{lk}) \approx \sqrt{\frac{1 + g_l g_k}{g_l g_k N}}. \quad (46')$$

If, on the other hand,  $\bar{m} \ll 1$ , then

$$\delta(\Phi_{lk}) \approx \frac{1}{\sqrt{g_l g_k N \bar{m}}} = \frac{1}{\sqrt{g_l g_k M}}, \quad (46'')$$

where  $M$  is the total number of disintegrations of each kind during the experiment.

We may compare the expressions which characterize the general content and the accuracy of the correlation method and the method of coincidences. The effective length of one interval of measurement in the method of coincidences is obviously equal to  $2\tau$ . The mean number of coincidences during one interval  $S_{lk}$  in the case of  $g_l \ll 1$  and  $g_k \ll 1$ , which is the most interesting one practically, is equal to

$$S_{lk} = g_l g_k \bar{m} + g_l g_k (\bar{m})^2 = \Phi_{lk} + \bar{r}_l \bar{r}_k. \quad (47)$$

It is evident that for  $\bar{m} \ll 1$ , when the chance coincidences are negligible,

$$S_{lk} = \Phi_{lk}. \quad (47')$$

The relative error in the determination of the correlation function is given in this case (for  $N \gg 1$ ) by the relation (46''), i.e., it is equal to the relative error in counting the number of true coincidences in the course of the whole experiment.

Thus for  $m \rightarrow 0$  and  $N \rightarrow \infty$ , the correlation method is identical with the method of coincidences. This circumstance has a clear physical significance. Indeed, for  $\bar{m} \ll 1$  a fortiori  $\bar{r}_l \ll 1$  and

$\bar{r}_k \ll 1$ , and hence  $\Phi_{lk} = \bar{r}_l \bar{r}_k - \bar{r}_l \bar{r}_k \approx \bar{r}_l \bar{r}_k$ . The

quantity  $\bar{r}_l \bar{r}_k$  is either zero if one of the  $r$  is zero, of unity if  $r_l = r_k = 1$ . But in the first instance the coincidence circuit would not record a coincidence; it would record a coincidence in the second instance. This shows that the two methods are identical if the counters are not heavily loaded. Of course we are discussing the equivalence of the methods only in principle, and not in practice, even in the region  $\bar{m} \ll 1$ .

If  $\bar{m}_l \gtrsim 1$ , i.e., if the chance coincidences cannot be neglected, then in the method of coincidences the second term in Eq. (47) is measured or calculated by one means or another, i.e., in fact the correlation function is determined indirectly. However, with a further increase of  $\bar{m}$  when  $\bar{m}$

$\gtrsim 1/\sqrt{g_l g_k}$ , i.e.,  $S_{lk} \gtrsim 1$ , the method of coincidences can no longer be used because of the large number of counts being lost. In the correlation method no limitations exist in principle for large values of  $\bar{m}$ . This means that for a given intensity of the primary radiation  $a$  one may increase the length of the interval  $t$  during which measurements are made, i.e., one can investigate related processes separated by long time intervals. The possibility of such investigations is limited only by the total length of the experiment and by the desired accuracy of results, which increases with increasing number of intervals in accordance with Eq. (46).

As has already been shown, the minimal error of the correlation method for a given duration of the whole experiment [retaining condition (14)], is attained for  $\bar{m} \rightarrow 0$  and  $N \rightarrow \infty$ , and is determined by (46''). As the number of intervals is decreased

(and, correspondingly,  $\bar{m}$  is increased), the relative error of the correlation method for  $g_l g_k \ll 1$  increases in accordance with

$$\delta = \delta_{\min} \sqrt{1 + \bar{m}}. \quad (48)$$

Thus, for a given duration of the experiment, the accuracy of the method begins to diminish appreciably only for  $\bar{m} \gg 1$ , when the method of coincidences begins to become completely unusable. By increasing the duration of the experiment, one can also achieve in this region any desired degree of accuracy in the determination of the correlation function. However, it should be noted (this was not emphasized in our short preliminary communication<sup>2</sup>) that if the intensity of radiation  $a$  can be decreased so that, for a given large  $t$ , the condition  $\bar{m} \ll [g_l g_k]^{1/2}$  is satisfied, then measurements by means of coincidence circuits become possible and, for the same duration of the experiment, give the same accuracy as the correlation method. Let us now turn to a comparison of the effect of background in both methods.

#### 4. THE ROLE OF BACKGROUND IN THE CORRELATION METHOD. COMPARISON WITH THE ROLE OF BACKGROUND IN THE METHOD OF COINCIDENCES

In the presence of background the total number of counts recorded by each counter in a certain time interval will be given by

$$r_l = r'_l + r''_l, \quad (49)$$

$$r_k = r'_k + r''_k, \quad (49')$$

where  $r'_l$  and  $r'_k$  are the counts associated with the process being investigated, and  $r''_l$  and  $r''_k$  are the background counts statistically independent of the former. It follows from Eq. (49) that

$$\bar{r}_l = \bar{r}'_l + \bar{r}''_l, \quad (50)$$

$$\bar{r}_k = \bar{r}'_k + \bar{r}''_k, \quad (50')$$

and also that

$$D(r_l) = D(r'_l) + D(r''_l) \quad (51)$$

and

$$D(r_k) = D(r'_k) + D(r''_k). \quad (51')$$

Insofar as  $\bar{r}'_l$  and  $\bar{r}'_k$  may usually be measured separately, the quantities  $r'_l$  and  $r'_k$  which we require may be determined easily. The correlation function is given by

$$\begin{aligned} \Phi_{lk} &= \bar{r}_l \bar{r}_k - \bar{r}_l \bar{r}_k \\ &= (\bar{r}'_l \bar{r}'_k - \bar{r}'_l \bar{r}'_k) + (\bar{r}''_l \bar{r}''_k - \bar{r}''_l \bar{r}''_k). \end{aligned} \quad (52)$$

If the background counts of the two counters are statistically independent, as is most frequently the case, then the second term in Eq. (52) is equal to zero, and the correlation function in which we are interested is identical with the one measured directly. In the opposite case (if, for example, both counters may be set off by the same particle) it is necessary to subtract from the measured correlation function the correlation function of background counts which is obtained by a separate experiment.

Thus the presence of background does not introduce any noticeable complications with respect to the speed of counting or the mean value of the correlation function. The presence of background, however, may increase the fluctuations of all the measured quantities, thus reducing the accuracy of measurement. The effect of background on the accuracy of measuring  $r'_l$  and  $r'_k$  is easily taken into account. Let us examine the more complicated question of the effect of background on the accuracy of measurement of the correlation function assuming that the background counts of the two counters are mutually independent. Using equations analogous to Eqs. (26)-(32), we obtain for the dispersion of the correlation function in the presence of background the expression

$$\begin{aligned} D(\Phi_{lk}) &= \frac{N-1}{N^2} \left[ 1 + g_l g_k + \frac{N-1}{Nm} \right. \\ &\quad \left. + \alpha_k + \alpha_l + \alpha_k \alpha_l \right] g_l g_k (\bar{m})^2, \end{aligned} \quad (53)$$

where  $\alpha_l$  and  $\alpha_k$  are the ratios of the background to the desired counts of the  $l$ th and  $k$ th counters. The relative error in the determination of  $\Phi_{lk}$  is given by

$$\delta(\Phi_{lk}) = \sqrt{\frac{1}{N-1} + \frac{1}{g_l g_k Nm} + \frac{1}{(N-1)g_l g_k} [1 + \alpha_l + \alpha_k + \alpha_l \alpha_k]}. \quad (54)$$

<sup>2</sup> V. I. Gol'danskii and M. I. Podgoretskii, Dokl. Akad. Nauk SSSR 100, 237 (1955).

While in the method of correlations the background merely increases the fluctuations without changing the average value of the correlation function, in the method of coincidences the background also affects the mean value of  $S_{lk}$  which, in accordance with Eq. (47), becomes equal to

$$S_{lk} = g_l g_k \bar{m} + g_l g_k (\bar{m})^2 \quad (55)$$

$$\times [1 + \alpha_l + \alpha_k + \alpha_l \alpha_k].$$

Thus in the presence of background for  $\bar{m} \geq (1 + \alpha_l + \alpha_k + \alpha_l \alpha_k)^{-1}$  chance coincidences have already become important, while for  $\bar{m} > [g_l g_k \times (1 + \alpha_l + \alpha_k + \alpha_l \alpha_k)]^{-1/2}$  the method of coincidences becomes inapplicable in practice because of missed counts. It is evident that in the correlation method, as before,  $\delta_{\min}(\Phi_{lk})$  for  $\bar{m} \rightarrow 0$ ,  $N \rightarrow \infty$  is determined by Eq. (46''), and for arbitrary  $\bar{m}$ ,

$$\delta(\Phi_{lk}) = \delta_{\min}(\Phi_{lk}) \quad (56)$$

$$\times \sqrt{1 + \bar{m} (1 + \alpha_l + \alpha_k + \alpha_l \alpha_k)}.$$

Consequently, in the presence of background, and for  $\bar{m} [1 + \alpha_l + \alpha_k + \alpha_l \alpha_k] \gg 1$ , i.e., in the region in which the method of coincidences is no longer applicable, the error of the correlation method increases not faster than  $\sqrt{\bar{m}}$ .

For a very high background count ( $\alpha_l \gg 1$ ,  $\alpha_k \gg 1$ ) we obtain from Eqs. (46'') and (56),

$$\delta^2(\Phi_{lk}) \approx \alpha_l \alpha_k / g_l g_k N. \quad (57)$$

Consequently, for the required number of intervals of measurement, which is determined by the condition  $\delta(\Phi_{lk}) \ll 1$ , we have

$$\sqrt{N} \gg \sqrt{\alpha_l \alpha_k / g_l g_k}, \quad (58)$$

and if we are dealing with the region in which  $\bar{m} \gtrsim 1/\alpha_l \alpha_k$ , then we obtain for the required total number of disintegrations in the course of the whole experiment

$$\sqrt{N\bar{m}} = \sqrt{M} \gg 1/\sqrt{g_l g_k}. \quad (59)$$

Let us now turn to an examination of concrete examples of possible applications of the correlation method.

## 5. EXAMPLES OF POSSIBLE APPLICATIONS OF THE CORRELATION METHOD

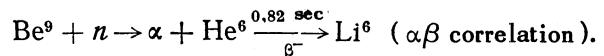
It has already been mentioned above that, in principle, the correlation method can be used to solve all those problems which are solved by the method of coincidences. Therefore, we shall not examine in detail those examples for which both methods are applicable. Let us just note briefly that above we were everywhere comparing the correlation method with that variant of the method of coincidences in which  $g_l$  and  $g_k$  are unknown, and for the determination of  $M$  it is necessary to record not only the number of coincidences but also the number of counts in the individual channels.

Frequently, however, this is not necessary, and only the number of coincidences is recorded. In such cases the method of correlations may become inapplicable, because in this method the counts in individual channels must be determined, and because of the presence of background they may be so large as to become difficult to record. Therefore, it may become necessary to reduce the intensity of radiation  $a$  when the correlation method is used, i.e., to increase the length of time of taking measurements in using the correlation method in comparison with the method of coincidences, and the correlation method may turn out to be the less advantageous one.

Let us now turn to typical problems, the solution of which at high intensities is possible by means of the correlation method, but impossible by the method of coincidences.

a) *Determination of cross sections of nuclear reactions.*

If, when the target  $A$  is bombarded by particles  $a$ , a nuclear reaction  $c$  takes place with the emission of particles  $b$  and the formation of a radioactive isotope  $B$ :  $A + a \rightarrow b + B$ , then, from observing the correlation between the emission of particles  $b$  at the instant of formation of  $B$  and the subsequent radioactive disintegration of  $B$ , one can determine the rate of formation of  $B$  under equilibrium conditions, and consequently, for a known flux of particles which leads to the formation of  $B$  (one can of course also formulate this question in the opposite direction). Example:



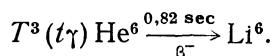
It is clear that at high intensities the method of coincidences is not applicable to the solution of

this and many similar problems. Indeed, for long resolving times the circuit will be missing counts almost continuously, and for short resolving times  $\Delta t$ , the efficiency of recording would be reduced by a factor of order  $\Delta t / \tau$ , where  $\tau$  is the mean lifetime of the isotope formed.

In such cases the only way of obtaining by the coincidence method the same accuracy and the same duration of the experiment as in the correlation method is to reduce the intensity of radiation (without increasing the original ratio of background to desired counts). However, this does not turn out to be possible in all cases, and coincidence circuits with resolving times greater than  $10^{-4} - 10^{-3}$  sec usually cannot be used because of missing counts.

The correlation method permits one to determine the cross section of the indicated reaction on the basis of the determination of the correlation function for the  $\alpha$  and  $\beta$  counters for some one arbitrary relative orientation of the two counters. For processes separated by large time intervals angular correlation is generally absent. Therefore, the cross section of the reaction under consideration is determined independently of the angular distribution of the  $\alpha$ -particles, and it therefore becomes unnecessary to measure the angular distribution in this problem. Any competing reactions accompanied by the emission of  $\alpha$ -particles [for example,  $\text{Be}^9(n, 2n) 2 \text{He}^4$ ] are of no importance for the determination of the cross section.

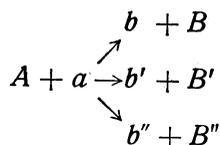
Another example of the same kind is the reaction



By studying the  $\gamma\beta$  correlation for one arbitrary relative orientation of the two counters it is possible to determine the cross section of this reaction without knowing either the efficiency of the  $\gamma$ -counter or the cross sections of competing reactions.

b) *Identification of the products of nuclear reactions.*

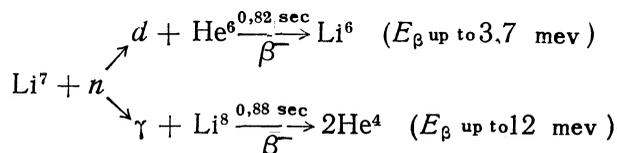
If when the target  $A$  is bombarded by some given particles several nuclear reactions can take place:



with the formation of different radioactive isotopes then by a suitable choice of various operating conditions of the counters one can determine with

which particular radioactive decay one or another type of secondary particles  $b$  is correlated, i.e., one can ascribe definite radioactive properties to definite isotopes.

Example:



It is evident that, for example, by varying the thickness of filters in front of the counters or the characteristics of the counters (we do not examine these possibilities in detail, because the same is done also in the method of coincidences, as is described, for example, in reference 1, one may select the more penetrating  $\beta$ -particles or distinguish between  $\gamma$  and  $d$  and to identify the resulting isotopes by this means. Still another possibility of separating effects produced by different isotopes is connected with the variation of the length  $t$  of the intervals of measurement which leads to a violation of condition (14).

c) *Determination of half-lives of decay.*

The correlation function for disintegrations of the  $l$ th and  $k$ th kind in an equilibrium radioactive mixture is defined by the general relation (20). Up till now we used assumption (14) because for intervals of length much greater than the half-lives of decay of all the substances formed between the  $l$ th and  $k$ th disintegrations the correlation is the most marked. However, assumption (14) is by no means compulsory. In the general case, for example, if the  $l$ th and  $k$ th decays are adjacent we obtain from Eq. (20) and from the laws of radioactive decay

$$\Phi_{lk} = g_l g_k \left[ at - \frac{a}{\lambda_k} (1 - e^{-\lambda_k t}) \right] = \Phi_{lk}(t), \quad (60)$$

where  $at = \bar{m}$ , and  $\lambda_k$  is the decay constant for the disintegration of the  $k$ th type.

It is evident that if we know  $\Phi(t)$  for at least two values of interval length we can determine the half-life of the  $k$ th member of the radioactive family without knowing the quantity of the  $k$ th or of the other substances, or the half-lives of the other substances, and independently of  $a$ ,  $g_l$  and  $g_k$ .

Knowing  $\lambda_k$ , we can then determine in a way analogous to what was done before the quantities  $a$ ,  $g_l$  and  $g_k$ , and also the equilibrium number of atoms of the  $k$ th type. Indeed

$$\frac{\Phi_{lk}(t_1)}{\Phi_{lk}(t_2)} = (\lambda_k t_1 - 1 + e^{-\lambda_k t_1}) / (\lambda_k t_2 - 1 + e^{-\lambda_k t_2}). \quad (61)$$

It is evident from (61) that the determination of  $\lambda_k$  must be carried out in such a way that at least one of the values of  $t$  utilized is of order  $1/\lambda_k$ , since for  $\lambda_k t \ll 1$  the correlation functions approach zero, while for  $\lambda_k t \gg 1$  they approach the constant value utilized in all the above calculations. The determination of the half-lives of decay by the correlation method may be carried out, naturally, not only for adjacent decays, but for arbitrarily related decays (including those separated by branchings of the chain), but the corresponding relations will be more complicated than (61).

The number of examples could be easily multiplied, but we have set ourselves for the present merely the problem of the general illustration of the possibilities of the method. For the same reason we shall not here dwell on the different

variants of the correlation method which can differ appreciably from the one described above.

## 6. CONCLUSIONS

1. The determination of the correlation function between the recorded number of decays of the  $l$ th and the  $k$ th type:

$$\Phi_{lk} = \overline{r_l r_k} - \overline{r_l} \cdot \overline{r_k}$$

permits one to find the absolute yield of nuclear transformations.

2. The mean value of the correlation function of consecutive related disintegrations in the absence of branching in between and with the length of the interval during which measurements are taken considerably larger than the mean time interval between these disintegrations is

equal to  $\overline{\Phi_{lk}} = \frac{N-1}{N} g_l g_k \bar{m}$  and does not depend on the background. The relative error in the determination of  $\overline{\Phi_{lk}}$  in the general case (in the presence of background) is equal to

$$\delta(\Phi_{lk}) = \sqrt{\frac{1}{N-1} + \frac{1}{g_l g_k N \bar{m}} + \frac{1}{(N-1)g_l g_k} (1 + \alpha_l)(1 + \alpha_k)}.$$

3. For a given duration of the measurement interval  $t$  the accuracy of the correlation method is the same as the accuracy of the method of coincidences. However, for  $\bar{m} > [g_l g_k (1 + \alpha_l) \times (1 + \alpha_k)]^{-1/2}$  the method of coincidences can no longer be used because of missed counts, while the correlation method continues to be usable.

4. The applicability of the correlation method for large  $\bar{m}$  is particularly important for the study of nuclear transformations the individual stages of which are separated by relatively great time-intervals (of the order of  $10^{-3} - 1$  sec).

Translated by G. M. Volkoff  
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