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Principles of correlation counting

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Among the basic restrictions on the measurement of source disintegration rates by the coincidence method are the condition that the decay is prompt and that the radiations emitted in the two stages can be registered in two detectors (and usually distinguished experimentally). These limitations, however, are not inherent in the physical process to be measured, but are due to the method by which is established the genetic connection between those pairs of pulses which result from the same nuclear transformation.

As a matter of fact, such an interdependence can also result in other observable effects than synchronism of the emissions, and these might permit one to determine the frequency of "coincident" events without actually counting them. Among the various quantities which can serve as a measure for the degree of internal relationship or "correlation" between two series of events, the covariance and the correlation function might be good candidates for the applications aimed at.

1. Introduction

Although some earlier applications of similar methods may be traced, the feasibility of correlation techniques for application in experimental nuclear physics was first clearly recognized around 1955 by a Russian group [1]. Practical applications began with the measurement of half-lives [2] which are too long for an efficient application of the delayed-coincidence method, and this has continued to be of major interest [3]. Examples of measurements are the 394 keV level in ^{88}Y or the ground state of ^{114}In. The possibility of determining absolute disintegration rates by correlation counting, although clearly described in [1], has not met much interest first. It was only after an article by Friedländer [4] had crossed the linguistic barrier that metrologists in the western hemisphere became aware of the interesting features of correlation counting as an alternative to the coincidence method. Thus, apart from some preliminary attempts [5] which passed largely unnoticed, the first detailed experiments were performed only in 1968 [6]. They showed that for a case like ^{60}Co the results for absolute disintegration rates, as determined by the correlation technique, were in excellent agreement with

those obtained by the conventional coincidence method, although the dead-time corrections were somewhat larger and the precision slightly lower (for equal measuring times). As the experimental set-up was more involved too, these early applications did not show a real advantage of this new approach. In the course of time [7], however, the following unique features of the correlation method were clearly recognized:

- as no coincidence circuit is used, all corrections involving its finite resolving time disappear,
- the absolute measurement of disintegration rates can be extended to two-stage decays with a relatively long-lived intermediate state,
- the method can also be applied if the two correlated radiations cannot be distinguished experimentally.

Finally, even in those cases where both techniques can be used, a comparison of their results is most interesting since the critical corrections applied are sufficiently different in both cases for the measurements to be considered as practically independent. Therefore systematic errors, if present, could then be recognizable. It should be clearly realized that all the additional problems encountered in absolute $4\pi\beta$-γ coincidence counting (e.g. decay-scheme dependent effects) which are not directly related to the way the coincidences are measured, remain unchanged.

Apart from its usefulness in determining half-lives and disintegration rates, a variation of the correlation technique has also been used for the quantitative measurement of afterpulses [8].

2. Application of covariance-variance techniques

We confine ourselves to give a simple sketch of the method as applied to two specific experimental situations. While this may be sufficient to explain the basic principle, anybody considering a real application will have to resort to the original papers, in particular to [7], as all the more subtle points will not be mentioned here.

a) Arrangement with two counters

If the two radiations, which will be described by the stationary stochastic series in time $X(t)$ and $Y(t)$, can be measured separately, we determine their covariance

$$\begin{aligned} \mathrm{Cov}\,(X,Y) &\equiv E\left\{(X-\hat{X})\cdot(Y-\hat{Y})\right\} \\ &= E\,(X\cdot Y) - \hat{X}\cdot\hat{Y}\,, \end{aligned} \tag{1}$$

where $\hat{X} \equiv E(X)$ is the expectation of X (in a measuring time interval T), and likewise for Y.

In our case, X and Y are the number of observed particles in T, thus

$$X = B + C \qquad \text{and} \qquad Y = G + C\,, \tag{2}$$

where B and G denote the number of uncorrelated pulses in the beta and gamma channels, respectively, whereas C corresponds to the true coincidences*. For the sake of simplicity, dead-time corrections will be ignored in what follows. Then B, G and C are independent and one obtains

$$\begin{aligned} \mathrm{Cov}\,(X,Y) &= E\left\{(B + C)\cdot(G + C)\right\} - (\hat{B} + \hat{C})\cdot(\hat{G} + \hat{C}) \\ &= E\,(C^2) - (\hat{C})^2 = \sigma^2(C). \end{aligned} \tag{3}$$

If the coincidences follow Poisson statistics, we have

$$\mathrm{Cov}\,(X,Y) = \hat{C} = \varepsilon_\beta\, \varepsilon_\gamma\, N_o\, T\,, \tag{4}$$

where N_o stands for the absolute disintegration rate and ε_β, ε_γ are the detection efficiencies of the counters. After n intervals, the experimental value for the covariance is obtained from the readings (x_i, y_i) by forming

$$\mathrm{Cov}\,(X,Y) = \frac{1}{n-1}\left[\sum_{i=1}^{n} x_i \cdot y_i - \frac{1}{n}\left(\sum_{1}^{n} x_i\right)\cdot\left(\sum_{1}^{n} y_i\right)\right]. \tag{5}$$

Neglecting dead-time effects again, the variance of C can be shown to be [7]

$$\sigma^2(C) \cong \frac{1}{n}\left[\hat{X}\cdot\hat{Y} + \hat{C}\,(\hat{C} + 1)\right]. \tag{6}$$

The individual measuring times are usually quite small ($T \cong 10^{-4}$ s). The number of intervals has to be very high ($n \cong 10^6$) as well as the total number of events counted to assure sufficient statistical precision.

The main field of application of the correlation method is not for prompt decays, but rather for those disintegrations which pass through a delayed state. Here, the conventional method falls seriously short since the corrections for missed (true) coincidences as well as for the accidental ones become too uncertain for large resolving times, even in the variants of anticoincidence counting or of delayed coincidences.

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* For more general situations, the terms parent and daughter events would be appropriate for X and Y.

The correlation method is more flexible here as the effect of finite lifetimes can be readily taken into account. Thus, for the case of two measurable radiations, the expression (4) for the covariance is now

$$\text{Cov}\,(X,Y) = \varepsilon_\beta\, \varepsilon_\gamma\, N_o\, T\, (1 - \frac{1 - e^{-\lambda T}}{\lambda T}) , \tag{7}$$

where λ is the decay constant of the intermediate level.

b) Arrangement with one counter

It is perhaps somewhat surprising that, even in the case where the two radiations X and Y cannot be distinguished experimentally, it may be possible to determine their coincidence rate. This is essentially achieved by an analysis of the variance of the superimposed process Z. We therefore form the variance

$$\text{Var}\,(Z) = E\{(Z - \hat{Z})^2\} = E(Z^2) - (\hat{Z})^2 , \tag{8}$$

where now $Z \equiv X + Y = B + G + 2\,C$, always for a given time interval T.

The mean number of events - still neglecting dead-time effects, although this would be less obvious to justify here - is clearly

$$E(Z) = \hat{Z} = \hat{B} + \hat{G} + 2\,\hat{C} . \tag{9}$$

If again B, G and C are Poisson distributed, we get

$$\text{Var}\,(Z) = \hat{B} + \hat{G} + 4\,\hat{C} = \hat{Z} + 2\,\hat{C} . \tag{10}$$

Hence, coincident pulses increase the variance more than the mean, and the coincidence rate can be deduced from their difference since

$$\hat{C} = \frac{1}{2}\left[\text{Var}\,(Z) - \hat{Z}\right] . \tag{11}$$

The statistical uncertainty of C, based on n measurements of Z, can be shown to be [9]

$$\sigma^2(C) \cong \frac{\hat{C}}{n} + \frac{(\hat{Z} + 2\,\hat{C})^2}{2\,n} . \tag{12}$$

For delayed intermediate states, the number of "coincident" pulses C, still measured by (11), is now given by

$$\hat{C} = \varepsilon_\beta\, \varepsilon_\gamma\, N_o\, T\, (1 - \frac{1 - e^{-\lambda T}}{\lambda T}) \tag{13}$$

and has to be interpreted as twice the number of parent-daughter pairs in T which originate from the same disintegration. The equations (7) and (13) are valid for $\lambda\tau \ll 1$ only, that is as long as the dead-time corrections can be neglected (τ is the dead time).

In all practical applications of the correlation method for absolute measurements of disintegration rates which should be of metrological interest, the simple formulae given above have to be modified to account for the losses produced by dead times. The evaluation of these corrections is often far from trivial, and first order approximations (in $N_o\tau$, $\lambda\tau$ or related quantities) are occasionally all one can do for the moment. A considerable amount of work has already been done and the best presently available corrections are summarized in [7], a paper which is fundamental for every serious application of correlation counting. Here details are also given on how this method has been applied to the absolute standardization of the isotopes ^{75}Se, ^{67}Ga, ^{67}Cu and ^{85}Sr and to the estimation of afterpulses in liquid scintillation counting.

3. Application of correlation functions

Another quantity currently used in statistics for characterizing the degree of similarity or interdependence which exists between random sequences of events is the correlation function. For two stationary and real processes, it may be defined by

$$R_{xy}(\delta) \equiv E\{X(t) \cdot Y(t+\delta)\}, \qquad (14)$$

where δ, the new variable, can be interpreted as a time delay inserted artificially between the two processes X and Y. We should mention, however, that for reasons of convergences X(t) and Y(t) are in general not directly the counting processes (number of events in t), but some conveniently chosen functions which are closely related to the sequences under study. To our knowledge, the cross-correlation function (14) has not yet been exploited for correlation counting, although attempts are under way and quite a number of applications have been made in related fields (see [10], with an abundant bibliography).

On the other hand, the corresponding one-channel version, where X and Y are identical, that is

$$R(\delta) = E\{X(t) \cdot X(t+\delta)\}, \qquad (15)$$

has already found several interesting applications in the fields which are of interest in this context. Here, too, an appropriate choice of the form of X(t) is a problem on its own. Probably the simplest possible choice is a two-valued function (for instance with the values +1 and -1) which changes state on each arrival of a pulse. In this case, X(t) is essentially what has been

known for long as a "random telegraph signal" (compare [11]). Its use for the study of nuclear disintegrations was probably first suggested by a French group in 1960 [12]. This choice for X(t) has also the effect that the (auto-) correlation function can be written as

$$R(\delta) = \sum_{k=0}^{\infty} (-1)^k \cdot W(k) \qquad (16)$$

$$= 1 - 2 \cdot \text{Prob (k odd)} ,$$

where W(k) is the probability for measuring exactly k counts in a time interval δ (with random start). For a simple Poisson process with mean rate N_o, we then obtain

$$R(\delta) = e^{-2 N_o |\delta|} , \qquad (17)$$

and the modifications due to the presence of a dead time are exactly known for this case [13]. Here again, the first applications have been made with the aim of measuring half-lives [14]. Examples are the 615 keV level in ^{181}Ta and the 304 keV level in ^{75}As. For an intermediate state with decay constant λ, (17) must be generalized to

$$R(\delta) = \exp\left\{-2(\alpha - \beta)|\delta| - \frac{4\beta}{\lambda}(1 - e^{-\lambda|\delta|})\right\} , \qquad (18)$$

where $\alpha \equiv N_o \cdot (\varepsilon_\beta + \varepsilon_\gamma - \varepsilon_\beta \varepsilon_\gamma)$ and

$$\beta \equiv N_o \cdot \varepsilon_\beta \varepsilon_\gamma ,$$

neglecting dead-time effects.

From the experimental point of view, the modulo-two version of correlation counting is attractive by its simplicity. The states of X(t) correspond to the two positions of a gate, and the same is true for $X(t+\delta)$, where the delay δ can be introduced by a shift register. The mean value of the product is obtained by a periodic sampling which checks the states of the two gates. Both the test pulses and the shifting of the registers are synchronized to the same crystal oscillator. Thus, in contrast with the covariance-analysis method [7], the outcomes of the individual runs have not to be stored or processed on line.

Although the limitation of X(t) to two values has been made primarily for the sake of simplicity* - other shapes, like rectangular with fixed width have occasionally been tried and are even used in some commercially available cross-correlators -, this choice turns out to be a very fortunate one

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* The loss of information is small as long as $N_o \delta$ is not too far from unity.

when viewed from a somewhat different angle. As can be seen from (16), $R(\delta)$ essentially indicates whether the number of counts in δ is even or odd. It is thus a modulo-two counter. Hence all pairs of pulses arriving in δ pass unnoticed, and it is this simple peculiarity which allows one to measure separately the count rate of singles [15], since out of the (average) total incoming events

$$(\varrho_{sing} + 2 \cdot \varrho_{pair}) \cdot \delta ,$$

this device registers only $\varrho_{sing}\,\delta$ (apart from edge effects due to the finite length of δ). The method, which has been developed for measuring afterpulses, can be generalized also to multiplets of higher order and has a sensitivity for $\varrho_{pair}/\varrho_{sing}$ approaching 10^{-3}. In this general form, the correlation technique provides a simple and versatile tool for analyzing a stationary random process.

4. Conclusion

Although previsions are dangerous, one would not expect, in the field of absolute disintegration rate measurements, the traditional coincidence method to be replaced by correlation techniques. In some experimental cases (delayed states), however, the situation is different since here the new methods are unrivalled. A few special applications (as for afterpulses) may also continue to deserve some interest. For the majority of the nuclides measured traditionally by the coincidence method, however, correlation counting will be applied only occasionally for control purposes.

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