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Correction methods for photon pile-up in lifetime determination by single-photon counting

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Abstract. The distortion of photon-count distributions from single photon counting at high count rates is discussed with consideration of its use in excited-state lifetime measurements. An *inhibit* function technique is described which facilitates collection of data at high count rates.

1. Introduction

Single photon counting is often necessary in measurements on light sources of low intensity and, in particular, in the determination of excited-state lifetimes from detection of weak emission (Bennett *et al.* 1965, Bridgett and King 1967). In this type of experiment an exciting source, for instance an electron beam, is rapidly cut off and the time distribution of the emission of single photons recorded as a histogram on a multi-channel analyser. In the study of an excited state of energy E above the ground state, complications arising from cascading are minimized by exciting with controlled energy electrons of energy $E+\Delta E$, where ΔE is kept as small as possible. In the form of the experiment using a start–stop time-to-amplitude converter, at the instant of the excitation cut-off a trigger pulse is produced which acts as a start or time origin pulse. A stop pulse is obtained from the detection of a single photon by a fast photomultiplier and the start–stop time interval converted into a proportional voltage pulse and recorded. The essential features of a typical apparatus are shown in figure 1, in which a new inhibit function is included which is discussed below. This

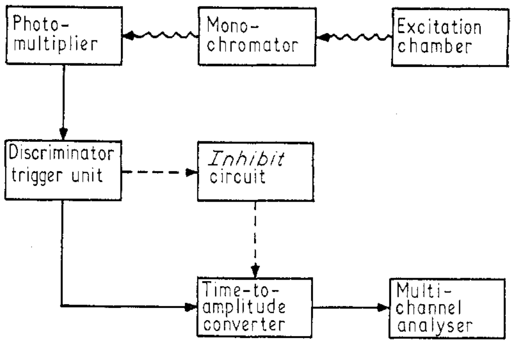

Figure 1. Schematic diagram of a typical single-photon counting apparatus for decay time measurements with an inhibit function incorporated.

experiment has the disadvantage that on the average only less than one count is stored in the analyser for each excitation cycle. Increase and optimization of the data collection rate is useful in reducing the time required to record the complete decay curve.

For excitation cycles in which more than one photon is detected only time information of the first photon is recorded which leads to distortion of the decay information.

This paper describes methods in which the experiment can be run at high count rates and in which curve distortion is reduced to a negligible value or corrected. An alternative is to use a photon detection rate sufficiently low for the probability of more than one photon being detected in each observation time range to be below an acceptable maximum value. This, however, greatly increases the necessary observation time for an adequate signal-to-noise ratio of the decay curve.

2. Single-photon counting in lifetime measurement

When operating at high count rates the true (undistorted) number of counts in the ith channel is $C\bar{n}_i$, where C is the number of excitation cycles and $\bar{n}_i$ is the true mean number of counts in channel i in one cycle. We need to obtain $C\bar{n}_i$ from the observed count distribution N_i.

The quantity N_i is related to the probability P_i of an event (one or more photons detected) in channel i by (Coates 1968)

$$N_i = CP_i \prod_{j=1}^{i-1} (1-P_j) \tag{1}$$

from which

$$P_i = \frac{N_i}{C - \sum_{1}^{i-1} N_j}. \tag{2}$$

The probability function P_i is given by (Mandel and Wolf 1965)

$$P_i = \sum_{n_i=1}^{\infty} p(n_i, T, (i-1)T) \tag{3}$$

where $p(n_i, T, (i-1)T)$ depends on the statistics of the emitting source and is the probability of n_i counts in channel i covering the time interval $(i-1)T$ to iT and is of the form (Mandel and Wolf 1965)

$$p(n_i, T, (i-1)T) = \frac{1}{n_i!} \langle \{qU(T, (i-1)T)\}^{n_i} \exp\{-qU(T, (i-1)T)\} \rangle \tag{4}$$

with

$$U(T, (i-1)T) = \int_{(i-1)T}^{iT} I(t')\, \mathrm{d}t'.$$

Here $I(t')$ is the intensity at time t' in the ith channel in one cycle and q is the photocathode quantum efficiency.

The way in which P_i is related to $\bar{n}_i$ depends on several factors, but principally on the ratio $T : T_c$, where T_c is the coherence time of the source, and on its state of polarization.

For the thermal (Gaussian) light source considered here we have (Mandel and Wolf 1965)

$$p(n_i, T, (i-1)T) = \int \frac{(qU)^{n_i}}{n_i!} \exp(-qU) p(U)\, \mathrm{d}U$$

and we obtain the relation

$$\bar{n}_i = P_i + \alpha P_i^2 + \beta P_i^3 + (\text{higher power terms}) \tag{5}$$

in which

$$\begin{aligned} \tfrac{1}{2}(T \gg T_c{}^{a}) \leqslant \alpha \leqslant 1(T \ll T_c{}^{b}) \\ \tfrac{1}{3}(T \gg T_c{}^{a}) \leqslant \beta \leqslant 1(T \ll T_c{}^{b}) \end{aligned} \tag{6}$$

where the superscript a refers to the paper by McLean and Pike (1965) and b refers to the paper by Mandel and Wolf (1965).

The exact correction needed to generate the true decay curve from the observed curve using equations (2) and (5) is difficult, particularly when $T \sim T_c$ because of the uncertainty in the relevant values of α and β in equation (5), which are also affected by the state of polarization of the emission.

Consequently it is undesirable to run the experiment at such high count rates that P_i^2 becomes significant in comparison with P_i, and the maximum acceptable count rate is set by the condition $P_i^2/P_i < \epsilon$ where ϵ is the maximum relative error which can be tolerated. However, if we make the assumption $\bar{n}_i \simeq P_i$ we obtain an equation for $\bar{n}_i$ of the form previously obtained by Coates

$$\bar{n}_i = P_i = \frac{N_i}{C - \sum_{1}^{i-1} N_j}. \tag{7}$$

3. The inhibit function technique

A convenient way of running the experiment at high count rate such that P_i is much higher but the N_i's form an undistorted true decay curve is to incorporate an *inhibit* function as shown in figure 1. This can be done in a number of ways but, in general, the standardized photoelectron pulses are split into two. With the use of suitable delays one pulse acts as the stop pulse and the other activates the inhibit circuit if more than one photoelectron pulse occurs over the observation time. As seen in figure 1 the discriminator-trigger unit which standardizes the photoelectron pulses provides two simultaneous outputs, one of which feeds directly into the inhibit circuit, the other goes via a delay, equal to or greater than the observation time range, to the stop input of the time-to-amplitude converter. The start pulses pass through a similar delay on their way to the time-to-amplitude converter to preserve the time relationship between start and stop pulses. If more than one photoelectron pulse occurs during the cycle, the arrival of the second photoelectron pulse causes the inhibit circuit to generate an output which disables the time-to-amplitude converter before the delayed pulse, which would otherwise have stopped it and recorded a count in the multi-channel analyser, has arrived. Suitable inhibit circuits include the use of fast scalers, coincidence units, pile-up gates or mixers.

An alternative and convenient method of incorporating the inhibit function is to feed the photoelectron pulses into both the start and stop inputs of the time-to-amplitude converter, via suitable delays so that a single photoelectron pulse will just fail to both start and stop the time-to-amplitude converter. The time-to-amplitude converter requires a certain minimum delay of a few nanoseconds between pulses occurring at the start and stop inputs in order for it to produce an output. Any additional photoelectron pulse during the cycle will stop the time-to-amplitude converter and a count will be recorded which represents the time interval between the first two photoelectron counts occurring during a cycle in which more than one photon is detected. Repetitive pulses produced at the initiation of each experimental cycle are also fed into the stop input of the time-to-amplitude converter via a delay equal to the observation time range. The multi-channel analyser is slowly ($\sim 0{\cdot}1$ Hz) cycled repetitively between add and subtract while the repetitive stop pulses are disconnected during subtract cycles. Consequently those counts which represent the time intervals between pairs of photoelectron counts recorded during multi-photon cycles add and subtract out. The data remaining derives from experiment cycles where only one photoelectron count appeared and the time interval is defined as between that count and the repetitive stop pulse. To prevent random starting of the time-to-amplitude converter it is maintained in an inhibit mode until the beginning of each cycle.

With the inhibit function the recorded decay curve in most practical cases is equivalent to the true decay curve with negligible distortion. The curve is made up of counts recorded during single-photon cycles. As the mean number of events per cycle increases from very low count rates, the number of single-photon cycles increases up to a maximum and then falls as multi-photon cycles begin to predominate. In most cases for count rates up to this optimum value, $P_i^2 \ll P_i$. It is undesirable to operate an experiment, either with or without the inhibit function incorporated, at such high count rates as to make $P_i^2/P_i > \epsilon$, as any attempted theoretical correction to the decay curve is subject to the errors arising from uncertainties in the constants in equation (5).

In the experimental inhibit methods, if the first photoelectron in a cycle occurs at a time corresponding to channel i, then a count is recorded only if no further counts occur after channel $i+r$. Counts occurring after the first count in the cycle, but within the channels i to $i+r$, are not observed because of the finite resolving time of the inhibit circuit for pairs of photoelectron pulses. This is taken to be rT, where r is the integer and T is the time interval corresponding to one channel width.

The probability of an event in channel i is, as before, P_i. This event is only recorded if the event in channel i is the first in the cycle. In this case

$$N_i = CP_i \prod_{1}^{i-1} (1-P_j) \prod_{i+r+1}^{m} (1-P_k^i) \tag{8}$$

where m is the number of channels of the multi-channel analyser and P_k^i is the conditional probability of an event in channel k given that there has been an event in channel i and possible events in channels $i+1$ to $i+r$. We can then write

$$N_i = \frac{CP_i}{1-P_i} \prod_{1}^{i} (1-P_j) \prod_{i+r+1}^{m} (1-P_k) \prod_{i+r+1}^{m} \left(\frac{1-P_k^i}{1-P_k}\right). \tag{9}$$

The factor

$$\prod_{i+r+1}^{m} \left(\frac{1-P_k^i}{1-P_k}\right) = P_c$$

alters the value of N_i because of correlations between photons arriving in different channels. We then obtain

$$N_i = \frac{CP_iP_c}{(1-P_i)} \frac{\prod_{1}^{m} (1-P_j)}{\prod_{i+1}^{i+r} (1-P_j)}. \tag{10}$$

Now,

$$C\prod_{1}^{m} (1-P_j)$$

is just the total number of experiment cycles where no event occurs and equals $C-N_E$. Here N_E is the total number of event cycles, the number of cycles where one or more photons are detected. Also

$$\prod_{i+1}^{i+r} (1-P_j)$$

is the probability of there being no photoelectron counts within the resolving time. For each channel covered by the resolving time $\bar{n}_j = P_j + \alpha P_j^2 + \beta P_j^3 + \ldots$ from equation (5).

In general, the resolving time will cover only very few channels (or, often in practice, fractions of a channel) and we can take P_j $(i+1 \leqslant j \leqslant i+r)$ as constant and equal to P_i. Then

$$\prod_{i+1}^{i+r}(1-P_j) \simeq \prod_{i+1}^{i+r}(1-P_i) \simeq 1-r\bar{n}_i$$

to first order. Hence for the experimental inhibit method the total number of counts in channel i is given by

$$N_i = P_i P_c (C-N_E)\{(1-P_i)(1-r\bar{n}_i)\}^{-1}. \tag{11}$$

When $P_k^2 \ll P_k$ one can neglect coherence and polarization effects and $P_c \to 1$, and for high inhibit circuit time resolution $r \to 0$, such that we obtain

$$\bar{n}_i = N_i(C-N_E+N_i)^{-1}. \tag{12}$$

Then, if, as is usually the case, $C-N_E \gg N_i$,

$$\bar{n}_i = \frac{N_i}{C-N_E} = \frac{N_i}{\text{constant}} \tag{13}$$

so that the recorded decay curve is undistorted.

During the experiment we record the following parameters:

$$C, N_i, N_D \left(= \sum_{i=1}^{m} N_i\right)$$

and, if required, N_E by use of an additional scaler. The relationship of N_D to N_E is shown in figure 2 for steady and decaying light sources. Here $N_D = Cp(1, mT)$,

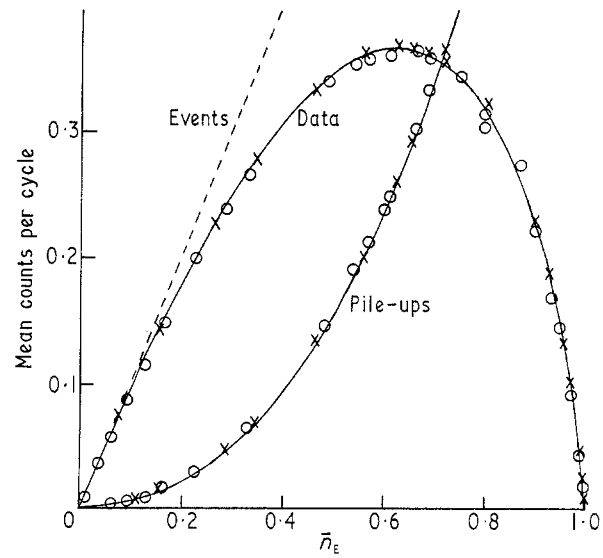

Figure 2. Mean photoelectron count rate distributions of $\bar{n}_E$, $\bar{n}_D$ and $\bar{n}_p$ for steady and decaying light sources (collected over 80 channels), $\bar{n}_E = N_E/C$, $\bar{n}_D = N_D/C$ and $\bar{n}_p$ = mean number of pile-ups per cycle = $(N_E-N_D)/C$. ○ steady light source; × decaying light source with s = 20.

in which $p(n, mT)$ is the probability of n photoelectron counts in a cycle defined by the time mT. If we assume that $p(n, mT)$ is Poisson distributed within a cycle ($mT \gg T_c$),

$$N_D = C\bar{n}_z \exp(-\bar{n}_z) \tag{14}$$

$$N_E = C\{1-\exp(-\bar{n}_z)\} \tag{15}$$

where $\bar{n}_z$ is the mean number of photoelectron counts per cycle. Then

$$N_D = \ln\{C(C-N_E)^{-1}\}(C-N_E). \tag{16}$$

N_D has a maximum value at $N_D = C/e$.

Equation (16) is strictly accurate only for measurements on steady light sources where the time of arrival of a photoelectron count within the cycle is random. However, experimental measurements show that N_D and N_E follow equation (16) closely, even when the light source decays during the cycle, as shown in figure 2.

5. Error considerations

We can compare the errors involved in using the inhibit method and the theoretical correction method of equation (7) with the errors occurring in an uncorrected curve. In each case we assume $\bar{n}_i \simeq P_i + \alpha P_i^2$. For an uncorrected curve the error in $\bar{n}_i$ arises from the assumption that $\bar{n}_i = N_i/C$. The relative error in $\bar{n}_i$ is

$$\frac{\Delta\bar{n}_i}{\bar{n}_i} = \left[\frac{N_i+\Delta N_i}{C-\sum_1^{i-1}(N_j+\Delta N_j)} + \frac{\alpha(N_i+\Delta N_i)^2}{\left\{C-\sum_1^{i-1}(N_j+\Delta N_j)\right\}^2} - \frac{N_i}{C}\right]\left(\frac{N_i}{C-\sum_1^{i-1}N_j}\right)^{-1}$$

$$\simeq \frac{\Delta N_i}{N_i} + \frac{2\alpha\Delta N_i}{\left(C-\sum_1^{i-1}N_j\right)} + \frac{\sum_1^{i-1}\Delta N_j}{C} + \frac{\sum_1^{i-1}N_j}{C} + \frac{\alpha N_i}{\left(C-\sum_1^{i-1}N_j\right)} \tag{17}$$

$$\simeq \text{random errors} + \frac{\sum_1^{i-1}N_j}{C} + \alpha\bar{n}_i. \tag{18}$$

The relative error in $\bar{n}_i$ arising from the theoretical correction is

$$\frac{\Delta\bar{n}_i}{\bar{n}_i} = \left[\frac{N_i+\Delta N_i}{C-\sum_1^{i-1}(N_j+\Delta N_j)} + \frac{\alpha(N_i+\Delta N_i)^2}{\left\{C-\sum_1^{i-1}(N_j+\Delta N_j)\right\}^2} - \frac{N_i}{C-\sum_1^{i-1}N_j}\right]\left(\frac{N_i}{C-\sum_1^{i-1}N_j}\right)^{-1}$$

$$\simeq \frac{\Delta N_i}{N_i} + \frac{\sum_1^{i-1}\Delta N_j}{\left(C-\sum_1^{i-1}N_j\right)} + \frac{2\alpha\Delta N_i}{\left(C-\sum_1^{i-1}N_j\right)} + \frac{\alpha N_i}{\left(C-\sum_1^{i-1}N_j\right)} \tag{19}$$

$$\simeq \text{random errors} + \alpha\bar{n}_i. \tag{20}$$

The relative error in $\bar{n}_i$ arising from the use of the inhibit function is

$$\frac{\Delta\bar{n}_i}{\bar{n}_i} = \left\{\frac{N_i+\Delta N_i}{C+N_i+\Delta N_i-N_\mathrm{E}-\Delta N_\mathrm{E}}+\frac{\alpha(N_i+\Delta N_i)^2}{(C+N_i+\Delta N_i-N_\mathrm{E}-\Delta N_\mathrm{E})^2}-\frac{N_i}{C-N_\mathrm{E}}\right\}$$

$$\times\left(\frac{N_i}{C+N_i-N_\mathrm{E}}\right)^{-1}$$

$$\simeq \frac{\Delta N_i}{N_i}+\frac{\Delta N_\mathrm{E}}{C-N_\mathrm{E}}+\frac{(2\alpha-1)\Delta N_i}{C-N_\mathrm{E}}-\frac{(1-\alpha)N_i}{C-N_\mathrm{E}}-\frac{\alpha N_i}{(C+N_i-N_\mathrm{E})(C-N_\mathrm{E})} \tag{21}$$

$$\simeq \text{random errors}-(1-\alpha)\bar{n}_i-\alpha\bar{n}_i^2. \tag{22}$$

The largest term in the random errors is in most cases the term $\Delta N_i/N_i$. However in most experiments the data obtained is fitted to a decay curve to obtain a value of lifetime and here the non-random errors are most important. For an uncorrected curve at moderate and high count rates this term becomes very large, particularly for the last few channels where it approaches the value N_E/C. The non-random error from the use of the theoretical correction arises from the uncertainty in the value of α, this error is small provided that $\bar{n}_i \ll 1$. This is also the case for the non-random error with the inhibit function. In both cases it is undesirable to run the experiment at such a high count rate that P_i^2/P_i becomes significant. In most practical cases when the inhibit function technique is used, provided the data collection rate is kept below or near the optimum, P_i^2 is still negligible in comparison with P_i.

The small value of curve distortion can be illustrated by considering the two special cases of a steady light source and an exponentially decaying source. For the steady light source

$$\bar{n}_i = \frac{\bar{n}_z}{m} = \text{constant}.$$

Without the inhibit

$$N_i = C\bar{n}_i(1-\bar{n}_i)^{i-1}$$

and the main error term is

$$\frac{\sum_1^{i-1} N_j}{C} = 1-(1-\bar{n}_i)^{i-1} = 1-\left(1-\frac{\bar{n}_z}{m}\right)^{i-1}.$$

With the inhibit the main error term is

$$\frac{(1-\alpha)N_i}{C-N_\mathrm{E}} = (1-\alpha)\bar{n}_i = \text{constant}$$

(from equation (10)), so that the data is undistorted. At the optimum data collection point $\bar{n}_z = 1$ and $\bar{n}_i = 1/m$. Since m is almost always very much greater than 1 (typically 100 to 500 channels), the condition $\bar{n}_i < \epsilon$ is fulfilled. An experimental count distribution of this form is shown in figure 3.

For an exponential decay of lifetime τ of the form $\bar{n}_i = A\exp(-iT/\tau)$, if we put $T = \tau/s$, where s is the number of channels per exponential decay period, $\bar{n}_i = A\exp(-i/s)$. For accuracy s must be reasonably large, and in an experiment where $\bar{n}_i$ is observed over two decades on the analyser display $s \approx m/5$. With

$$A = \bar{n}_z\left\{\exp\left(\frac{1}{s}\right)-1\right\}\left\{1-\exp\left(-\frac{m}{s}\right)\right\}^{-1}$$

then

$$(\bar{n}_i)_{\max} = \text{maximum error term} = A \exp\left(-\frac{1}{s}\right)$$

and from equation (12)

$$(\bar{n}_i)_{\max} = \bar{n}_z\left\{1-\exp\left(-\frac{1}{s}\right)\right\}\left\{1-\exp\left(-\frac{m}{s}\right)\right\}^{-1} \simeq \frac{\bar{n}_z}{s} \simeq \frac{N_{\mathrm{E}}}{sC}.$$

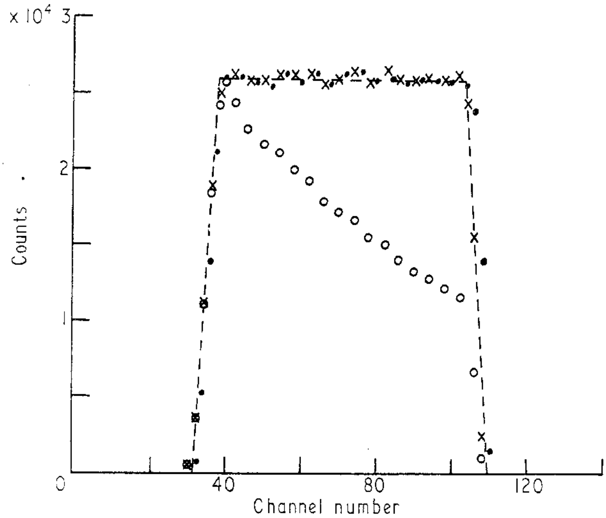

Figure 3. High count rate intensity distribution for gated steady light source with uncorrected and corrected data and data collected using inhibit function, channel width $T = 2{\cdot}5$ ns. ○ uncorrected data, $\bar{n}_{\mathrm{E}} \simeq 0{\cdot}58$; × corrected data; ● data collected using inhibit, $\bar{n}_{\mathrm{D}} \simeq 0{\cdot}37$; ---- true form of data.

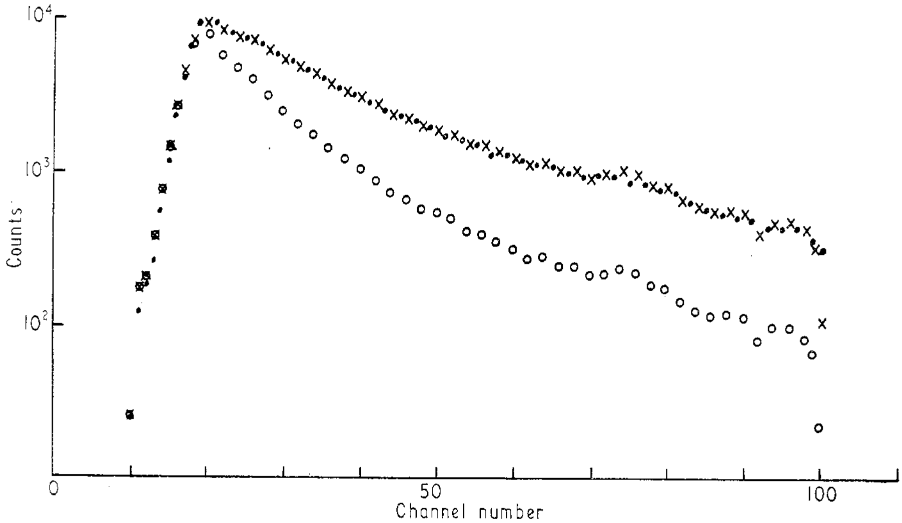

Figure 4. A gated decay curve measured at high count rate with and without the use of the inhibit function compared with data corrected by equation (7). ○ uncorrected data, $\bar{n}_{\mathrm{E}} \simeq 0{\cdot}8$; × corrected data; ● data collected using inhibit, $\bar{n}_{\mathrm{D}} \simeq 0{\cdot}32$.

For a count rate as high as 0·2 events per cycle and with s as small as 20 the maximum error in a channel is only about 1% and the data distortion is lower than this.

Figure 4 shows an experimental decay curve obtained with and without inhibit and on which the theoretical correction has been carried out.

6. Discussion

Although the theoretical correction gives an improvement in accuracy in all experimental cases, it is often difficult to carry out, particularly because of uncertainties in the values of N_i in the first few channels. Then gating of the photoelectron pulses is desirable to allow data to accummulate in well-defined regions of the analyser.

It can be seen from figures 3 and 4 that there is good agreement between the true data obtained at low count rate and the data obtained at high count rate using the inhibit function.

It is apparent that in most experimental situations the inhibit techniques allows the collections of accurate data at much higher count rates than is otherwise possible and without correction of the recorded data being necessary. The technique is also particularly useful when used in experiments running at moderate count rates (10^{-1}–10^{-2} events per cycle) where moderate curve distortion would otherwise result and where the need for data correction may not be appreciated.

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