Appl. Phys. B 41. 95–102 (1986)

Applied Physics B Photophysics and Laser Chemistry

Dead Time Correction of Photon Correlation Functions

K. Schätzel

Institut für Angewandte Physik, Universität, D-2300 Kiel, Fed. Rep. Germany

Received 25 November 1986/Accepted 2 June 1986

Abstract. The dynamic range of single photon counting measurements in quasi elastic light scattering is restricted by detector and counter dead time effects. While distortions of single interval statistics have been treated at great length, only lowest order corrections or very special cases of dead time effects on temporal correlation functions were computed in the past.

Dead times result in a direct distortion of correlograms on time scales comparable to the dead time. This effect exists even at low count-rates. It is independent of the count rate for paralyzable systems. Nonparalyzable systems show a count rate dependence with increasing correlation times at high count rates.

Furthermore, counting saturation produces additional distortions extending to all lag times. These distortions are computed for the rather general case of Γ-distributed intensities with arbitrary shape of the photon correlation function. Such signals are commonly found in multiparticle homodyne experiments with a finite size detector, i.e. arbitrary value of the intercept or contrast of the correlogram. Exact results are provided for the paralyzable system including the effect of fluctuating dead times. The latter case is then used to compute a useful approximation for nonparalyzable systems as well.

PACS: 07.65.Eh, 82.80.Di, 85.10.Pe

Introduction

The great success of single photon counting techniques in modern light scattering may largely be attributed to two facts: First, single photon counting offers high sensitivity with almost ideal, quantum limited noise performance. Second, the digital character of a sequence of count pulses is well suited for digital processing, free of drifts and distortion problems. For these reasons, photon correlation [1–3] has become the standard technique for spectral analysis of quasi elastic light scattering data and found many applications in physics, chemistry and biology – the most prominent one being submicron particle sizing and the measurement of diffusion constants.

However, compared to analog semiconductor detectors, photomultipliers and amplifier/discriminator systems used for single photon counting are restricted to a rather limited dynamic range only. Dark count rates, e.g. due to thermal electrons and nuclear decay events [4] set a lower limit of some 10 Hz, which may be somewhat reduced by cooling of the detector tube. The upper limit is set by detector or processor dead times, typically between 10 and 100 ns. The easy way to avoid appreciable dead time distortions is to keep mean count rates below say 1% of the maximum peak count rates, and indeed many experimenters commonly restrict light intensities to avoid count rates exceeding 100 kHz. The price they pay is reduced statistical accuracy or prolonged measurement times.

Although there exists an extensive amount of literature about distortions of single interval counting distributions [5–18], only a few results are available for dead time effects on temporal correlation functions [19, 20]. It is the purpose of this paper to provide more detailed knowledge of correlation distortions for Gaussian, i.e. multi-particle, scattering signals with arbitrary temporal correlation including the important effect of fintie detector size. This type of signal is typically found in homodyne or self-beat experiments

[3, 17], the most common optical setup for dynamic light scattering.

A summary of fundamental properties (Sect. 1) will be followed by an exact calculation for paralyzable detector systems (Sect. 2) and an approximation for nonparalyzable systems (Sect. 3).

1. Notation and Fundamental Properties

Most work on dead time distortion in photon statistics refers to the pioneering work of DeLotto et al. [5], who considered counting distributions for counters which are unblocked or blocked by an event just at the beginning of the sampling interval of length t. Both cases are not typical for photon correlation. Modern real-time correlators use sophisticated derandomizers to achieve continuous counting with perfectly adjacent sampling intervals (width t). Their single interval counting statistics are characterized by the so called equilibrium distributions, where nothing is known about the initial state of the counter. Hence we will use equilibrium distributions throughout this paper.

The dead-time characteristics of counting systems depend on the fact whether events falling into a "dead" period extend this period ("paralyzable counter") or are completely ignored ("nonparalyzable counter") [5]. A Schmitt-trigger type discriminator circuit approximates the paralyzable system – overlapping photoelectron pulses result in one extended output pulse. Non retriggerable one-shots commonly used for pulse shaping correspond to the nonparalyzable system. Although most authors restricted their calculations to the latter case, we will consider both cases here.

Photon correlation generally relies on the simple linear relationship between the measured digital correlogram (more precisely: its expectation) and the intensity autocorrelation function,

$$\langle n_0 n_\tau \rangle = \alpha^2 t^2 \langle I_0 I_\tau \rangle , \tag{1}$$

where n_0 and n_τ denote the photoelectron counts obtained during sampling intervals of width t centered at times 0 and τ, I_0 and I_τ are the corresponding intensities, and α stands for the quantum efficiency of the detector system [3]. For sampling times t well below coherence times – the typical situation in quasi elastic light scattering – the photon count n is Poisson distributed about αI in the absence of dead times [17], hence

$$\langle n \rangle = \alpha I t . \tag{2}$$

For non-overlapping counting intervals, statistical independence of individual photoelectrons implies statistical independence of n_0 and n_τ, i.e. for each particular realization of the intensity as a function of time we obtain

$$\langle n_0 n_\tau \rangle = \langle n_0 \rangle \langle n_\tau \rangle . \tag{3}$$

Averaging over intensity statistics and using (2) we arrive at (1).

Unfortunately, both conditions are violated in the presence of a finite dead time T. With increasing count rate more and more events fall into dead times and nonlinear saturation replaces (2). We will calculate this nonlinearity and its consequences in Sects. 2.2 and 3.2. At small lag times τ, dead time introduces a direct statistical dependence between adjacent pulses – an immediate consequence of the existence of a minimum pulse separation T. This direct correlation is present even at low count rates! The resulting distortion of correlograms at small lag times will be the subject of Sects. 2.1 and 3.1.

As dead-time distortions generally depend on details of the intensity statistics [14, 15] as well as on mean count rates, we will have to use a particular stochastic process for many of our calculations. Fortunately, most quasi-elastic light scattering experiments for particle sizing applications are performed in the homodyne setup [3] with a large number of scatterers present in the measurement volume. The central limit theorem implies Gaussian amplitude statistics or an exponential intensity distribution [17], if the detector is small enough to ensure perfectly coherent detection. In practice, larger apertures are used to improve detector sensitivity, as indicated by intercepts of measured correlograms

$$\beta = \left(\lim_{\tau \to 0} \langle n_0 n_\tau \rangle - \langle n \rangle^2 \right) \Big/ \langle n \rangle^2 \tag{4}$$

well below the Gaussian value 1. The finite size detector covers more than one coherence area and the resulting intensity may be described by a Γ-distribution

$$p(I) = \frac{1}{\Gamma(1/\beta)} (\beta \langle I \rangle)^{-1/\beta} I^{1/\beta - 1} \mathrm{e}^{-I/\beta\langle I \rangle} , \tag{5}$$

where $\langle I \rangle$ denotes the mean intensity and $1/\beta$ is a measure of the number of coherence areas covered [17]. For second order correlations we need the two-point distribution of the intensity which may be obtained by inverse Laplace transformation of its generating function

$$\langle \mathrm{e}^{-s_0 I_0 - s_\tau I_\tau} \rangle = [(1 + \beta\langle I \rangle s_0)(1 + \beta \langle I \rangle s_\tau) - \beta^2 \langle I \rangle^2 \varrho^2 s_0 s_\tau]^{-1/\beta} \tag{6}$$

as

$$p(I_0, I_\tau) = \frac{\exp[-(I_0 + I_\tau)/\beta\langle I \rangle (1 - \varrho^2)]}{\beta^2 \langle I \rangle^2 (1 - \varrho^2) \Gamma(1/\beta)} \times \left(\frac{\sqrt{I_0 I_\tau}}{\varrho \beta \langle I \rangle} \right)^{1/\beta - 1} I_{1/\beta - 1} \left(\frac{2\varrho \sqrt{I_0 I_\tau}}{\beta \langle I \rangle (1 - \varrho^2)} \right) , \tag{7}$$

where ϱ is defined by

$$\langle I_0 I_\tau \rangle = \langle I \rangle^2 (1 + \beta \varrho^2) \tag{8}$$

and denotes the temporal correlation coefficient of a single mode field amplitude. It should be noted that our model distribution is general enough to include typical heterodyne experiments as well. The corresponding very small intercepts β imply a large number of degrees of freedom, where the Γ-distributions approach the desired Gaussian distributions because of the central limit theorem.

2. Paralyzable System

2.1. Direct Dead Time Distortion

A paralyzable system by definition "kills" all events which occur within less than the dead time T of another event of the "input" process, i.e. the process idealized to zero dead time. For Poisson input statistics with a mean given by (2) we obtain the probability $q \cdot dt$ of obtaining an event within a certain time interval of width dt as a product of the probability to find no input event in the dead time interval T prior to dt, $\exp(-\alpha I T)$, and the probability of an input event within dt, $\alpha I dt$, or

$$q = \alpha I \exp(-\alpha I T). \tag{9}$$

The only restriction implied in (9) is the reasonable assumption of dead times well below coherence times of the intensity process. Whenever we refer to q at a specific time, we will use this time as an index just like we did for intensities in (6) and (7).

Integration of (9) over the sampling interval dt yields the dead time distorted mean count number

$$\langle n \rangle = \alpha I t \exp(-\alpha I T). \tag{10}$$

Direct statistical dependence between events does not extend beyond one dead time T. Input pulses occuring earlier than T before the considered interval dt have no influence on q. Since a dead time T, on the other hand, ensures a minimum separation of events by T, we obtain the conditional probability density for detection of an event within dt centered at τ past another (output-) event as

$$q^c(\tau) = \begin{cases} 0 & \text{for} \quad 0 < \tau < T \\ q & \text{for} \quad \tau > T \end{cases}. \tag{11}$$

Multiplication with the probability density of the initial event and with the widths of both intervals yields the two-point distribution

$$q_0 q_\tau^c(\tau) dt^2 = \begin{cases} 0 & \text{for} \quad 0 < \tau < T \\ q_0 q_\tau dt^2 & \text{for} \quad \tau > T \end{cases}, \tag{12}$$

which must be integrated over the finite lengths of the sampling intervals to produce the photon correlation function for a given realization of the intensity:

$$\begin{aligned} &\langle n_0 n_\tau | I_0, I_\tau \rangle \\ &= \int_0^t dt_1 \int_\tau^{\tau+t} dt_2 q_0 q_\tau^c(t_2 - t_1) \\ &= q_0 \int_{\tau-t}^{\tau+t} (t - |\tau' - \tau|) q_\tau^c(\tau') d\tau' \\ &= \begin{cases} q_0 q_\tau t^2 & \text{for } \tau > T + t \\ q_0 q_\tau [t^2 - \frac{1}{2}(t + T - \tau)^2] & \text{for } T + t > \tau > T \\ q_0 q_\tau \frac{1}{2}(\tau - T + t)^2 & \text{for } T > \tau > T - t \\ 0 & \text{for } T - t > \tau . \end{cases} \end{aligned} \tag{13}$$

Since the usual digital correlators compute correlograms at lag times which are integer multiples of the sampling time t,

$$\tau_k = k \cdot t, \tag{14}$$

the results of (13) are more appropriately expressed in Fig. 1, where the dead time correction factors

$$f_k = \frac{\langle n_0 n_{kt} \rangle}{q_0 q_{kt} t^2} \tag{15}$$

are graphed as a function of the ratio of dead time T and sample time t.

For dead times smaller than the sample time, only the first channel of the correlogram will be distorted by

$$f_1 = 1 - T^2/2t^2. \tag{16}$$

It should be noted that (13) does really not depend on the intensities I_0 and I_τ. The conditional expectation equals the expectation, and this fact was used in the definition of (15). Independence of the direct dead time correction from count rate and intensity statistics yields a nice and simple general correction formula. But it also implies that these distortions of the correlogram at small lag times cannot be avoided by resorting to small mean count rates. Instead the

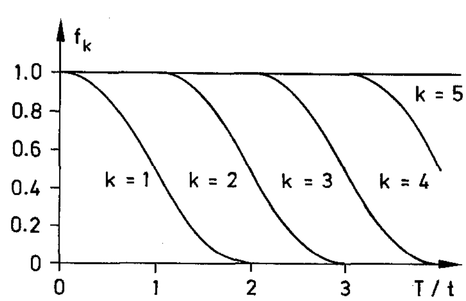

Fig. 1. Direct dead time correction factors for paralyzable systems as a function of dead time over sample time

corrections of Fig. 1 or (13) should be used to put an end to the common practice of ignoring the fist one or two channels during the evaluation of measured correlograms – at least, if afterpulsing remains negligible on the short time-scales considered.

2.2. Nonlinearity Effects

For a complete dead time correction of correlograms measured with paralyzable systems we still have to compute the distortion of $\langle q_0 q_\tau \rangle$ due to counting saturation. This distortion may be obtained by direct integration over the two-point intensity density, as given in (7) for the finite detector, many particle signal. A simpler approach makes use of the generating function, (6), and requires the calculation of a derivative only. For maximum generality we allow variable dead times T_0 and T_τ for the two sampling intervals considered:

$$\begin{aligned}
&\langle q_0 q_\tau | T_0, T_\tau \rangle \\
&= \langle \alpha I_0 e^{-\alpha I_0 T_0} \alpha I_\tau e^{-\alpha I_\tau T_\tau} \rangle \\
&= \alpha^2 \left\langle \frac{\partial}{\partial s_0} \frac{\partial}{\partial s_\tau} e^{-I_0 s_0 - I_\tau s_\tau} \Big|_{s_0 = \alpha T_0, s_\tau = \alpha T_\tau} \right\rangle \\
&= \alpha^2 \frac{\partial}{\partial s_0} \frac{\partial}{\partial s_\tau} [(1+\beta\langle I\rangle s_0)(1+\beta\langle I\rangle s_\tau) \\
&\quad - \beta^2 \langle I\rangle^2 \varrho^2 s_0 s_\tau]^{-1/\beta} \Big|_{s_0=\alpha T_0, s_\tau = \alpha T_\tau} \\
&= \alpha^2 \langle I\rangle^2 \{1 + \alpha\langle I\rangle \beta (T_0 + T_\tau) \\
&\quad + \alpha^2 \langle I\rangle^2 \beta^2 T_0 T_\tau + \beta[1 - \alpha\langle I\rangle (T_0 + T_\tau) \\
&\quad - 2\alpha^2 \langle I\rangle^2 \beta T_0 T_\tau] \varrho^2 + \alpha^2 \langle I\rangle^2 \beta^2 T_0 T_\tau \varrho^4\} \\
&\quad \times [1 + \alpha\langle I\rangle \beta (T_0 + T_\tau) \\
&\quad + \alpha^2 \langle I\rangle^2 \beta^2 (1 - \varrho^2) T_0 T_\tau]^{-2-1/\beta}\,. \qquad (17)
\end{aligned}$$

Simplifying to a single dead time $T = T_0 = T_\tau$, dividing by the square of the mean count rate,

$$\begin{aligned}
\langle q \rangle &= \langle \alpha I e^{-\alpha I T} \rangle = \alpha \frac{\partial}{\partial s} \langle e^{-Is} \rangle |_{s=\alpha T} \\
&= \alpha \frac{\partial}{\partial s} (1 + \beta \langle I \rangle s)^{-1/\beta} |_{s=\alpha T} \\
&= \alpha \langle I \rangle (1 + \alpha \langle I \rangle \beta T)^{-1-1/\beta}\,, \qquad (18)
\end{aligned}$$

and using the abbreviation

$$\varepsilon = \alpha \langle I \rangle T \qquad (19)$$

we obtain the dead time corrected normalized photon correlation function

$$\frac{\langle q_0 q_\tau \rangle}{\langle q \rangle^2} = \frac{1 + \beta(1 - 2\varepsilon - 2\beta\varepsilon^2)(1+\beta\varepsilon)^{-2} \varrho^2 + \beta^2 \varepsilon^2 (1+\beta\varepsilon)^{-2} \varrho^4}{[1 - \beta^2 \varepsilon^2 (1+\beta\varepsilon)^{-2} \varrho^2]^{2+1/\beta}}\,. \qquad (20)$$

Dead time enters into (20) as the product of input count rate $\alpha\langle I \rangle$ in the absence of dead time and the dead time T. This product, ε, equals the mean number of input counts which fall into one dead time interval. At low count rates ($\varepsilon \ll 1$), (20) may be approximated up to second order in ε:

$$\begin{aligned}
\frac{\langle q_0 q_\tau \rangle}{\langle q \rangle^2} &\approx 1 + \beta[1 - 2\varepsilon(1+\beta) + \varepsilon^2 (1+\beta)(1+3\beta)] \varrho^2 \\
&\quad + \beta^2 \varepsilon^2 (1 + 3\beta) \varrho^4\,. \qquad (21)
\end{aligned}$$

The first order term agrees with earlier calculations [19] and causes a decrease of intercept only, i.e. the shape of the correlogram remains identical. More important are the terms of second order in ε, which include the square of the undistorted correlation as well. This term is the lowest order nonlinear distortion of the correlogram and produces artificial polydispersity and bias towards smaller hydrodynamic radii or larger diffusion constants in particle sizing applications.

Expansions of (20) to higher orders in ε are of little practical value, due to the asymptotic character of these representations which limits their use to very small ε. The next higher power of ϱ, a ϱ^6-term, does not occur until fourth order in ε.

2.3. Fluctuating Dead Times

The Schmitt-Trigger type discriminator mentioned in Sect. 1 as a system which should provide paralyzable dead time effects, will generally show a certain spread of dead times associated with a spread in photomultiplier pulse amplitudes. If this dead time spread can be determined experimentally, e.g. in the form of a histogram, we can use this information to average dead time corrections over fluctuations in T.

For the direct distortion discussed in Sect. 2.1 this implies further smoothing of the correction factors f_k. If all dead times remain below the sample time t, direct distortion will still be limited to the first channel. We obtain

$$f_1 = 1 - \langle T^2 \rangle / 2t^2 = 1 - \frac{\langle T \rangle^2}{2t^2} - \frac{\mathrm{Var}(T)}{2t^2}\,, \qquad (22)$$

an increasing distortion for increasing variance of the dead time.

Averaging the mean count rate expression (18) is just as straightforward, if dead time distribution information is available as a histogram. More care has to be taken for the two-interval expressions, which involve two independent dead times. We have to use (17), the conditional expectation, and average over both dead times, T_0 and T_τ, independently. Only then may we compute the quotient for the normalized corrected correlogram, similar to (20):

$$\frac{\langle q_0 q_\tau\rangle}{\langle q\rangle^2}=\frac{\langle\!\langle q_0 q_\tau | T_0, T_\tau\rangle\!\rangle_{T_0,T_\tau}}{[\langle\!\langle q|T\rangle\!\rangle_T]^2}, \tag{23}$$

where $\langle\,\rangle_T$ denotes the dead time averages.

2.4. *Iterative Dead Time Correction*

As a first stage of dead time correction, the one (or more) correction coefficients f_k (13 and 15), should be divided out to correct the first channel(s) of a measured correlogram. This step is necessary even at low count rates.

The second stage, correction of nonlinearity distortions, is necessary, if ε^2 terms cannot be neglected in (21), more precisely if

$$\varepsilon^2\beta(1+3\beta)\ll 1 \tag{24}$$

is violated. At first glance, (24) seems to indicate less dead time problems at small intercepts β. However, smaller intercepts obtained by larger apertures of the detector imply larger intensities and hence larger dead time parameters $\varepsilon=\alpha\langle I\rangle T$. In fact, ε rises like $1/\beta$ or more, and the dead time effects increase with decreasing intercept.

If nonlinearity distortions are to be corrected, this correction should preferably be based upon (20). The asymptotic character of (21) makes it difficult to estimate the quality of this approximation at larger ε. Clearly, (19) cannot be solved for β and $\varrho^2(\tau)$ in a closed form. These solutions must be approached by iterative techniques instead.

As a first problem we extrapolate the measured correlogram towards zero lag time in order to obtain

$$\langle q\rangle^{-2}\lim_{\tau\to 0}\langle q_0 q_\tau\rangle =(1+\beta)(1+\beta\varepsilon)^{2+2/\beta}(1+2\beta\varepsilon)^{2-1/\beta}. \tag{25}$$

Together with (18) or

$$\langle q\rangle=\varepsilon\frac{t}{T}(1+\beta\varepsilon)^{-1-1/\beta} \tag{26}$$

we may compute ε and β for a given dead time T. Starting from an initial value

$$\varepsilon=\langle q\rangle T/t \tag{27}$$

we solve (25) for β. With this estimate of β we can use (26) to improve our ε value, and this iteration may be continued until stable results are obtained. Only a few cycles should be required for moderate ε.

The second problem yields the final dead time correction. With known ε and β, (20) has to be solved at all lag times in the correlogram to obtain $\varrho^2(\tau)$. Like for the solution of (25), the complicated form of the equation suggests to use an iteration procedure that does not require the knowledge of derivatives. The uncorrected channels of the correlogram may be used as initial values for ϱ^2, or we may compute the solution of the ε-expansion (21),

$$\varrho^2(\tau)\approx\frac{1-a}{2b\sqrt{\beta}}\left\{\left[1+\frac{4b}{(1-a)^2}\times\left(\frac{\langle q_0 q_\tau\rangle}{\langle q\rangle^2}-1\right)\right]^{1/2}-1\right\} \tag{28}$$

with

$$a=2\varepsilon(1+\beta)-\varepsilon^2(1+\beta)(1+3\beta),$$
$$b=\varepsilon^2(1+3\beta).$$

The correction technique may be generalized to include fluctuating dead times by replacing (20) with (23) and changing (25) and (26) accordingly. The computational effort rises essentially linearly with the number of bins used in the dead time histogram.

3. Nonparalyzable System

3.1. *Direct Dead Time Distortion*

Again we restrict our calculations to dead times T well below coherence times. Hence we may apply the known results for Poisson input signal [5] to determine the probability of finding an output event in a certain time interval dt,

$$q=\frac{\alpha I}{1+\alpha IT}, \tag{29}$$

and the conditional probability to find an output event in dt centered at a time τ past a given output event,

$$q^c(\tau)=\alpha I\sum_{k=1}^{K}\frac{(\alpha I)^{k-1}}{(k-1)!}(\tau-kT)^{k-1}\times\exp[-\alpha I(\tau-kT)], \tag{30}$$

where K is the largest integer such that $KT<\tau$. Equation (29) approaches the simple step function obtained for the paralyzable system, (11), at low count rates. There is, however, a distinct count rate dependence at high rates which is illustrated in Fig. 2. The systematic antibunching in non-paralyzable systems leads to an increasing direct correlation effect for increasing count rates with the extreme case of periodically spaced events with period T at infinite input count rates.

In order to obtain $\langle n_0 n_\tau\rangle$, we have to integrate $q_0\cdot q_\tau^c(t_2-t_1)$ over both sampling intervals like in (13)

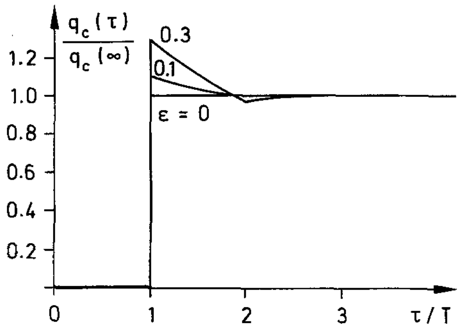

Fig. 2. Relative conditional probability of an output pulse following another pulse after a time τ as a function of τ over dead time T for three input count rates, 0, $0.1/T$, and $0.3/T$

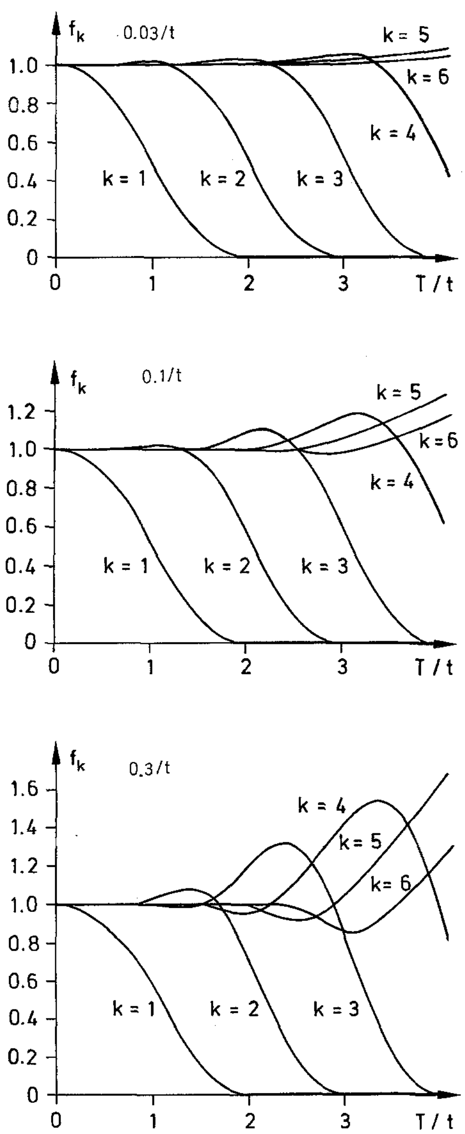

Fig. 3. Direct dead time correction factors for nonparalyzable systems as a function of dead time T over sample time t for three input count rates, $0.03/t$, $0.1/t$, $0.3/t$

and obtain

$$\langle n_0 n_\tau \rangle = q_0 \int_{\tau-t}^{\tau+t} q_\tau^c(\tau')(t - |\tau' - \tau|)d\tau' . \quad (31)$$

The integration is straightforward but leads to complicated expressions due to the τ'-dependent upper summation limit in (26). Numeric integration of (30) over the triangular weight function in (31) provides a more practical solution and was used to compute the correction factors $\langle n_0 n_{it} \rangle / q_0 q_{it} t^2$ as a function of T for various count rates in Fig. 3. For large lag times we obtain – of course – a limit equal to 1 just as for paralyzable systems.

3.2. Nonlinearity Effects

Averaging $q_0 q_\tau$ over the finite detector, multi particle statistics (7),

$$\langle q_0 q_\tau \rangle = \left\langle \frac{\alpha I_0}{1+\alpha I_0 T} \frac{\alpha I_\tau}{1+\alpha I_\tau T} \right\rangle , \quad (32)$$

involves a two-dimensional integration over the joint Γ-density of (7) or (32) may be expressed as an integral over the associated generating function, (6). Both approaches yield a rather complicated integral involving incomplete Γ-functions.

For a first estimate of nonlinearity distortions we may expand (32) as a series in $\varepsilon = \alpha\langle I \rangle T$ and compute the mixed intensity moments as derivatives of the joint characteristic function, (6):

$$\begin{aligned}
&\langle q_0 q_\tau \rangle \\
&= \alpha^2 \langle I \rangle^2 \left[\frac{\langle I_0 I_\tau \rangle}{\langle I \rangle^2} - 2\varepsilon \frac{\langle I_0^2 I_\tau \rangle}{\langle I \rangle^3} + 2\varepsilon^2 \frac{\langle I_0^3 I_\tau \rangle}{\langle I \rangle^4} \right. \\
&\quad \left. + \varepsilon^2 \frac{\langle I_0^2 I_\tau^2 \rangle}{\langle I \rangle^4} + O(\varepsilon^3) \right] \\
&= \alpha^2 \langle I \rangle^2 \{1 + \beta\varrho^2 - 2\varepsilon(1+\beta)(1+2\beta\varrho^2) \\
&\quad + 2\varepsilon^2(1+\beta)(1+2\beta)(1+3\beta\varrho^2) \\
&\quad + \varepsilon^2(1+\beta) \\
&\quad \times [1 + \beta + 4\beta(1+\beta)\varrho^2 + 2\beta^2\varrho^4] + O(\varepsilon^3)\} \\
&= \alpha^2 \langle I \rangle^2 \{1 - 2\varepsilon(1+\beta) + \beta[1 - 4\varepsilon(1+\beta) \\
&\quad + 2\varepsilon^2(1+\beta)(5+8\beta)]\varrho^2 \\
&\quad + 2\varepsilon^2\beta^2(1+\beta)\varrho^4 + O(\varepsilon^3)\} . \qquad (33)
\end{aligned}$$

Expansion of $\langle q \rangle$ up to ε^2 provides the normalized correlation function:

$$\begin{aligned}
\langle q \rangle &= \alpha \langle I \rangle [1 - \varepsilon(1+\beta) \\
&\quad + \varepsilon^2(1+\beta)(1+2\beta) + O(\varepsilon^3)] , \qquad (34)
\end{aligned}$$

$$\begin{aligned}
\frac{\langle q_0 q_\tau \rangle}{\langle q \rangle^2} &= 1 + \beta[1 - 2\varepsilon(1+\beta) + \varepsilon^2(1+\beta) \\
&\quad \times (3+7\beta)]\varrho^2 + 2\varepsilon^2\beta^2(1+\beta)\varrho^4 + O(\varepsilon^3) . \quad (35)
\end{aligned}$$

As compared to the paralyzable system (21), (35) indicates the same linear decrease of the intercept with increasing count rate. The ε^2-terms are both larger, resulting in a less rapid decrease of the intercept and a stronger (nonlinear!) ϱ^4-term, particularly for β well below 1.

The larger ε^2-coefficients also indicate even worse convergence of the asymptotic expansion (33). Growth of the coefficients like $\Gamma(1+k\beta)$ for the order ε^k limits the use of (33–35) to very small ε, where dead time corrections are not very necessary. Better convergence may be obtained by an expansion of the mean dead time corrected count rate q into a sum of negative exponentials. For practical calculations the approximation

$$\frac{1}{1+\alpha IT} \approx \frac{2+\sqrt{2}}{4}\exp[-(2-\sqrt{2})\alpha IT] + \frac{2-\sqrt{2}}{4}\exp[-(2+\sqrt{2})\alpha IT] \quad (36)$$

provides a close fit (error $<0.1\%$ up to $\alpha IT=0.2$, $<1\%$ up to $\alpha IT=0.5$) and is well suited for exact calculations for $\langle q_0 q_\tau \rangle$. Equation (36) was obtained by fitting up to the third derivative at $T=0$.

Equation (36) approximates the nonparalyzable count rate saturation by a paralyzable system with a dead time distributed with weights $(2\pm\sqrt{2})/4$ at the dead times $T(2\mp\sqrt{2})$. Equation (23) provides the solution of this problem as

$$\frac{\langle q_0 q_\tau \rangle}{\langle q \rangle^2} \approx \frac{(6+4\sqrt{2})\langle q_0 q_\tau | T_1, T_1 \rangle + 4\langle q_0 q_\tau | T_1, T_2 \rangle + (6-4\sqrt{2})\langle q_0 q_\tau | T_2, T_2 \rangle}{[(2+\sqrt{2})(1+\alpha\langle I \rangle \beta T_1)^{-1-1/\beta} + (2-\sqrt{2})(1+\alpha\langle I \rangle \beta T_2)^{-1-1/\beta}]^2 \alpha^2 \langle I \rangle^2} \quad (37)$$

with $T_{1,2}=(2\mp\sqrt{2})/2T$ and the conditional correlations given by (17). Like (36), this approximation for dead time distorted correlograms of a nonparalyzable system is correct up to order ε^2 and provides a highly improved fit even for ε-values not very much smaller than 1, as compared to the polynomial expansion. The closed form of the result is well suited for an iterative correction procedure paralleling that for paralyzable systems given in Sect. 2.4.

5. Conclusions

Dead times affect photon correlation functions in two separate ways. A direct (anti-)correlation exists for lag times comparable to the dead time. This effect exists even at small count rates and should be corrected whenever sample times are not very large compared to the dead time. The correction is particularly simple for paralyzable systems, because it shows no dependences upon count rate.

The second way in which dead times distort correlograms is through saturation nonlinearities of the count rate. The resulting distortion may be computed exactly for paralyzable systems, if we restrict our attention to sample times well below coherence times – the typical situation in most photon correlation experiments. Results are given for Γ-distributed intensities with arbitrary degrees of freedom and arbitrary temporal correlation. This very general process covers all homodyne (and approximates well most heterodyne) experiments with a large number of scattering particles, i.e. Gaussian amplitude statistics. The results are generalized to fluctuating dead times described by a histogram.

Equivalent calculations for nonparalyzable systems are more complicated. Instead we suggest an approximation of the nonparalyzable system by a paralyzable one with a simple, two-peak dead time distribution. This approximation provides much better estimates of first moments and the correlation than a power series expansion, while still being simple enough to serve for a practical correction procedure.

Such a dead time correction procedure involves iterative routines to first solve for the time independent distribution parameters by extrapolation to lag time zero – obviously including direct distortion correction – and then so solve for the undistorted correlation at all lag times of the experiment. The procedure requires precise knowledge of the dead time or dead time statistics and the design of suitable measurement setups as well as computer simulations are now under way in our laboratory.

Of the two dead time effects on photon correlation functions, only the direct anticorrelation may be avoided by performing a two detector cross correlation experiment – a useful scheme to avoid afterpulsing problems [21]. The second type of distortions, due to count rate saturation occurs in cross correlation just as in autocorrelation. The same type of intensity statistics applies and the only further complication in the calculations of Sect. 2.2 is the introduction of two possibly different quantum efficiencies α_0 and α_τ of the two detectors.

Acknowledgements: Thanks go to E. O. Schulz-DuBois and M. Drewel for continuing discussion and to B. Stampa for first experimental verifications.

References

1. H.Z. Cummins, E.R. Pike (eds.): *Photon Correlation and Light Beating Spectroscopy* (Plenum, New York 1974)
2. H.Z. Cummins, E.R. Pike (eds.): *Photon Correlation Spectroscopy and Velocimetry* (Plenum, New York 1977)
3. E.O. Schulz-DuBois (ed.): *Photon Correlation in Fluid Mechanics*, Springer Ser. Opt. Sci. **38** (Springer, Berlin, Heidelberg 1983)
4. An Introduction to the Photomultiplier, EMI Electronics Ltd.
5. I. DeLotto, P.F. Manfredi, P. Principio: Energia Nucl. **11**, 557 (1964)
6. F.A. Johnson, R. Jones, T.P. McLean, E.R. Pike: Phys. Rev. Lett. **16**, 589 (1966)
7. G. Bédard: Proc. Phys. Soc. (London) **90**, 131 (1967)
8. R.F. Chang, V. Korenman, C.O. Alley, R.W. Detenbeck: Phys. Rev. **178**, 612 (1969)
9. A. Kikkawa, K. Ohkubo, H. Sato, N. Suzuki: Opt. Commun. **12**, 227 (1974)
10. E.E. Serralach, M. Zulauf: J. Appl. Math. Phys. (ZAMP) **26**, 669 (1975)
11. M.C. Teich, W.J. McGill: Phys. Rev. Lett. **36**, 754 (1976)
12. S.K. Srinivasan: Phys. Lett. **50** A, 277 (1974), J. Phys. A (Math. Gen.) **11**, 2333 (1978)
13. B.I. Cantor, L. Matin, M.C. Teich: Appl. Opt. **14**, 2819 (1975)
14. G. Vannucci, M.C. Teich: Opt. Commun. **25**, 267 (1978), J. Opt. Soc. Am. **71**, 164 (1981)
15. M.C. Teich, G. Vannucci: J. Opt. Soc. Am. **68**, 1338 (1978)
16. L. Mandel: J. Opt. Soc. Am. **70**, 873 (1979)
17. B. Saleh: *Photoelectron Statistics* Springer Ser. Opt. Sci. **6** (Springer, Berlin, Heidelberg, New York 1978)
18. A.S. Arutyunov: Opt. Spectrosc. (USSR) **53**, 179 (1982)
19. E. Jakeman, C.J. Oliver, E.R. Pike: J. Phys. A **4**, 827 (1971)
20. S.K. Srinivasan, M. Singh: Phys. Lett. **8**, 409 (1981)
21. F.T. Arecchi, M. Corti, V. Degiorgio, S. Donati: Opt. Commun. **3**, 284 (1971)