Clipped correlation of integrated intensity fluctuations of thermal light with an arbitrary spectrum

1975 J. Phys. A: Math. Gen. 8 1134

(http://iopscience.iop.org/0305-4470/8/7/017)

J. Phys. A: Math. Gen., Vol. 8, No. 7, 1975. Printed in Great Britain. © 1975

Clipped correlation of integrated intensity fluctuations of thermal light with an arbitrary spectrum

Bahaa E A Saleh and Jules Hendrix
Max Planck Institut für Biophysikalische Chemie, Göttingen, Germany

Received 26 November 1974, in final form 19 February 1975

Abstract. An approximate formula is obtained for the single clipped correlation function of integrated intensity fluctuations of thermal light with an arbitrary spectrum. This formula is compared to the exact function known for light with a Lorentzian spectrum.

1. Introduction

Since the advent of the techniques of photon correlation spectroscopy, special attention has been given to thermal light with a Lorentzian spectrum. Indeed all aspects of the performance of the photon digital correlator have been examined for this light (for a recent review see Jakeman 1973). Apart from the simplifications, due to its Markovian nature, which make theoretical investigations possible, it occurs in fact in many interesting situations. The light scattered from fluctuations of molecules in solutions due to Brownian motion and from many other targets of light scattering experiments has Lorentzian spectra (Cummins 1973).

With the establishment of photon correlation spectroscopy as an efficient laboratory technique for the study of the hydrodynamic properties of macromolecules in dilute solutions, other spectra became of interest. For example, the multi-Lorentzian spectrum (sum of Lorentzian components) is of particular importance in experiments where the rotational diffusion or the internal motion of flexible molecules adds to the usual light scattering effect due to translational diffusion (see the review by Cummins 1973 and references therein). Therefore it becomes necessary to investigate further those aspects of the performance of a digital photon correlator which depend on the spectral shape.

In this paper an approximate formula is obtained for the clipped correlation function of integrated intensity fluctuations (which is the quantity measured by a clipping digital correlator) for thermal light with an arbitrary spectral shape. No limitation is set on the sampling time or on the area of the detector. However, the dead-time effects of the photodetector are assumed negligible.

The clipped correlation function is normally obtained from the joint moment generating function (MGF) of integrated intensities (Jakeman and Pike 1969). Since an exact expression for this MGF for an arbitrary spectrum is not available, an approximate expression recently developed by Saleh (1975) is used. Using this approximation we show that the single clipped correlation function is equal to a constant (independent of time delay) plus a term proportional to the unclipped (full) correlation function. The temporal and spatial integration effects determine these constants and, of course, they determine the relation between the full correlation function and the spectrum.

2. The clipped correlation function

Let $Q(s_1, s_2) = \langle \exp(-s_1 w_1 - s_2 w_2) \rangle$ be the joint MGF of the integrated intensities w_1 and w_2 (measured in units of number of counts) in time intervals T separated by a time delay τ. Then as discussed by Saleh (1975), a good approximation for Q which is valid for arbitrary T is

$$Q(s_1, s_2) \simeq \left(1 + (s_1 + s_2)\frac{\bar{n}}{N} + s_1 s_2 \frac{\bar{n}^2}{M^2(\tau)}(1 - |g^{(1)}(\tau)|^2)\right)^{-N} \tag{1}$$

where $\bar{n} = \langle w \rangle$ is the average number of counts per sampling time, $g^{(1)}(\tau)$ is the normalized coherence function and N and $M(\tau)$ are parameters to be defined below.

From (1), the one-fold MGF

$$Q(s) = \langle \exp(-sw) \rangle \simeq (1 + s\bar{n}/N)^{-N} \tag{2}$$

results and is recognized as the expression which leads to the approximate photon counting statistics of Bédard *et al* (1967). Equations (1) and (2) generate the following moments:

$$g^{(2)}(0) = \frac{\langle w^2 \rangle}{\langle w \rangle^2} = \left.\frac{\partial^2 Q(s)}{\partial s^2}\right|_{s=0} = 1 + \frac{1}{N} \tag{3}$$

and

$$g^{(2)}(\tau) = \frac{\langle w_1 w_2 \rangle}{\langle w \rangle^2} = \left.\frac{\partial}{\partial s_1}\frac{\partial}{\partial s_2} Q(s_1, s_2)\right|_{s_1 = s_2 = 0}$$

$$= 1 + \frac{1}{N} - \frac{N}{M^2(\tau)}(1 - |g^{(1)}(\tau)|^2) \qquad \tau > 0. \tag{4}$$

Now N and $M(\tau)$ can be adjusted such that (1) generates the exact $g^{(2)}(\tau)$ for all τ. This can be attained by taking

$$N = (g^{(2)}(0) - 1)^{-1} \tag{5}$$

and

$$M^2(\tau) = N(1 - |g^{(1)}(\tau)|^2)/(g^{(2)}(0) - g^{(2)}(\tau)) \qquad \tau \neq 0 \tag{6}$$

which when substituted in (1) gives

$$Q(s_1, s_2) \simeq \left[\left(1 + s_1\frac{\bar{n}}{N}\right)\left(1 + s_2\frac{\bar{n}}{N}\right) - s_1 s_2 \frac{\bar{n}^2}{N}(g^{(2)}(\tau) - 1)\right]^{-N}. \tag{7}$$

Not only does (7) generate exact $\langle w^2 \rangle$ and $\langle w_1 w_2 \rangle$ but it also generates good approximations for higher moments ($\langle w_1^2 w_2 \rangle$ and $\langle w_1^2 w_2^2 \rangle$) as has been shown by comparison with the exact formulae of a Lorentzian spectrum (Saleh 1975). In the limit of short sampling time and small detector area, $g^{(2)}(0) \simeq 2$, $N \simeq 1$ and (7) reproduces the exact expression (Jakeman 1970). Note that (7) is different from the approximate formula used by Jakeman *et al* (1971) to account for the spatial effects and which does not generate the correct $g^{(2)}(\tau)$ for $\tau \neq 0$ (if N is chosen to satisfy (5)) in the presence of temporal integration.

Now we proceed to determine the normalized clipped correlation function at a clipping level K by using the formula (Jakeman and Pike 1969)

$$g_K^{(2)}(\tau) = \frac{1}{\bar{n}_K \bar{n}} \left(\bar{n} + \sum_{m=0}^{K} \frac{(-1)^m}{m!} \frac{\partial^m}{\partial s_2^m} \frac{\partial}{\partial s_1} Q(s_1, s_2) \Big|_{\substack{s_1=0 \\ s_2=1}} \right) \tag{8}$$

where $\bar{n}_K$ is the average number of clipped counts given by

$$\bar{n}_K = 1 - \sum_{m=0}^{K} \frac{(-1)^m}{m!} \frac{\partial^m}{\partial s^m} Q(s) \Big|_{s=1}. \tag{9}$$

Substituting from (7) and (2) in (8) and (9) it is straightforward to show that

$$g_K^{(2)}(\tau) = 1 + c(\bar{n}, N, K)(g^{(2)}(\tau) - 1) \tag{10}$$

where

$$c(\bar{n}, N, K) = \frac{\bar{n}}{\bar{n}_K} \binom{N+K}{K} \left(\frac{N}{\bar{n}+N} \right)^{N+1} \left(\frac{\bar{n}}{\bar{n}+N} \right)^K \tag{11}$$

and

$$\bar{n}_K = 1 - \left(\frac{N}{\bar{n}+N} \right)^N \sum_{m=0}^{K} \binom{N+m-1}{m} \left(\frac{\bar{n}}{\bar{n}+N} \right)^m \tag{12}$$

in which

$$\binom{N}{m} = \frac{\Gamma(N+1)}{m!\,\Gamma(N-m+1)}$$

and $\Gamma(\cdots)$ is the gamma function. Equation (10) is the desired approximate relation. The sampling time T enters this equation through $g^{(2)}(\tau)$ and N.

In the limit of short sampling time and small detector area ($g^{(2)}(0) = 2$, $N = 1$ and $g^{(2)}(\tau) = 1 + |g^{(1)}(\tau)|^2$), equations (10), (11) and (12) reproduce, as expected, the known results (Jakeman and Pike 1969)

$$g_K^{(2)}(\tau) = 1 + \frac{1+K}{1+\bar{n}} |g^{(1)}(\tau)|^2$$

and

$$\bar{n}_K = \left(\frac{\bar{n}}{\bar{n}+1} \right)^{K+1}.$$

The implications of (10) are rather interesting (and perhaps unexpected). The temporal dependence of the (full) digital correlation function is not (in this approximation) distorted by single clipping. Exact formulae for a Lorentzian spectrum show that this indeed holds in the case of a small detector area but arbitrary sampling time (Jakeman 1973). The approximate analysis of Jakeman *et al* (1971) in the case of short coherence time but arbitrary area and spectral shape reaches the same conclusion. Koppel (1971) has shown that for a sum-of-Lorentzians spectrum and arbitrary sampling time, $g_K^{(2)}(\tau)$ is an undistorted linear function of $|g^{(1)}(\tau)|^2$ in the restricted range of τ much greater than any coherence time.

It should be noted that equation (10) does not imply that the spectral shape or the spatial coherence distribution at the detector has minor importance in determining $g_K^{(2)}(\tau)$ (see the comments of Mandel 1972 on the work of Lachs 1971). These factors determine the constant c by determining N and more importantly by determining the relationship between $g^{(2)}(\tau)$ and $|g^{(1)}(\tau)|$. The example given below makes this point clear.

2.1. *Application to Gaussian cross-spectrally pure light with multi-Lorentzian spectrum*

Let the temporal part of the normalized field correlation function be a sum of exponentials, ie

$$g^{(1)}(\tau) = \sum_j \alpha_j \,\mathrm{e}^{-\Gamma_j|\tau|} \qquad \sum_j \alpha_j = 1, \tag{13}$$

or the spectrum a sum of Lorentzians. Then by direct double integration or by solving a differential equation analogous to that used by Jakeman (1970), we show that

$$g^{(2)}(\tau) = 1 + f(A) \sum_{i,j} \alpha_i\alpha_j \frac{\sinh^2\gamma_{ij}}{\gamma_{ij}^2} \mathrm{e}^{-(\Gamma_i+\Gamma_j)|\tau|} \qquad \tau \neq 0 \tag{14}$$

$$g^{(2)}(0) = 1 + f(A) \sum_{i,j} \alpha_i\alpha_j(\mathrm{e}^{-2\gamma_{ij}} + 2\gamma_{ij} - 1)/2\gamma_{ij}^2 \tag{15}$$

where $\gamma_{ij} = \frac{1}{2}(\Gamma_i + \Gamma_j)T$ and $f(A)$ is the spatial integration factor found by Jakeman *et al* (1970). Using (5), N is determined by

$$N^{-1} = f(A) \sum_{i,j} \alpha_i\alpha_j(\mathrm{e}^{-2\gamma_{ij}} + 2\gamma_{ij} - 1)/2\gamma_{ij}^2. \tag{16}$$

Also from (10) and (14)

$$g_K^{(2)}(\tau) = 1 + c(\bar{n}, N, K) \sum \alpha_i\alpha_j \frac{\sinh^2\gamma_{ij}}{\gamma_{ij}^2} \mathrm{e}^{-(\Gamma_i+\Gamma_j)\tau}, \tag{17}$$

indicating that the spatial integration has only a multiplicative effect and that temporal integration changes the amplitudes of the different exponential components without affecting the time constants. Hence a fitting procedure can recover the time constants (which are usually the quantities of most interest in a light scattering experiment). If the relative amplitudes are also to be estimated then a correction should be made for the factors $\sinh(\gamma_{ij})/\gamma_{ij}$.

3. Comparison with exact theory for a single Lorentzian and zero clipping

As a check on the approximate formula we compare it with the exact formula in the special case of a single Lorentzian spectrum and small detector area. This exact formula (Jakeman 1973) is in general very complicated because it contains multiple derivatives of order K. Therefore we restrict ourselves to the case of $K = 0$ in which exact results are given by

$$g_0^{(2)}(\tau) = 1 + c|g^{(1)}(\tau)|^2, \tag{18}$$

$$c = -\frac{\sinh\gamma}{\gamma} \frac{Q(1)}{1 - Q(1)} P(1) \tag{19}$$

and

$$\bar{n}_0 = 1 - Q(1) \tag{20}$$

where

$$Q(s) = e^{\gamma}[\cosh y + \tfrac{1}{2}(\gamma/y + y/\gamma)\sinh y]^{-1}, \tag{21}$$

$$P(s) = \tfrac{1}{2}Q(s)(\gamma/y - y/\gamma)\sinh y \tag{22}$$

and

$$y = (\gamma^2 + 2\gamma s\bar{n})^{1/2}. \tag{23}$$

On the other hand the approximate formula for $K = 0$ predicts

$$c = \frac{\bar{n}}{\bar{n}_0}\left(\frac{N}{\bar{n}+N}\right)^{N+1}\frac{\sinh^2\gamma}{\gamma^2} \tag{24}$$

and

$$\bar{n}_0 = 1 - \left(\frac{N}{\bar{n}+N}\right)^{N} \tag{25}$$

where, as seen from (16),

$$N = 2\gamma^2/(e^{-2\gamma} + 2\gamma - 1). \tag{26}$$

Numerical values for c and $\bar{n}_0$ have been computed for a range of values of γ and $\bar{n}$. The results are plotted in figures 1 and 2 from which it is apparent that for $\bar{n} < 1$ the

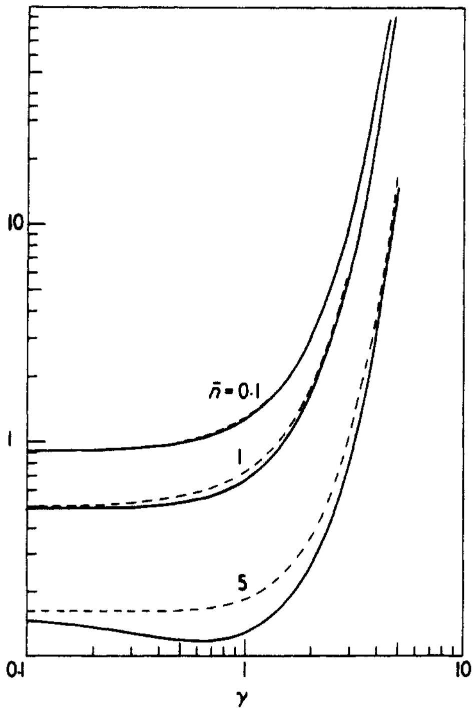

Figure 1. The factor c relating the zero clipped correlation function $g_0^{(2)}(\tau)$ to $|g^{(1)}(\tau)|^2$ as a function of $\gamma = \Gamma T$ for several values of $\bar{n}$, the average counting rate per sampling time.

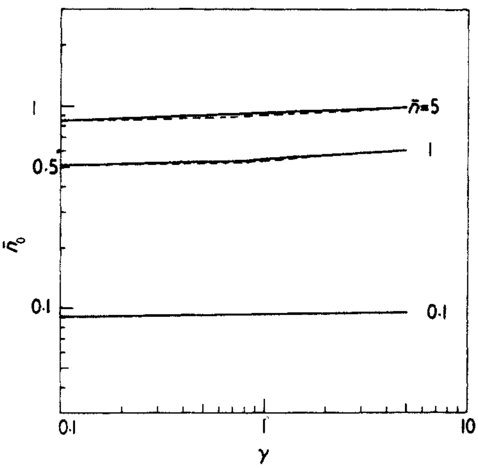

Figure 2. The average number of zero clipped counts per sample as a function of γ for several values of $\bar{n}$.

accuracy is excellent. In general the accuracy is better for small and for large γ. For $\bar{n} = 1$ a maximum error of about 9 % in c and 1 % in $\bar{n}_0$ occurs around $\gamma = 1$. For large $\bar{n}$ the error is larger and unless γ is very small or very large the approximation is not satisfactory.

References

Bédard G, Chang J C and Mandel L 1967 *Phys. Rev.* **160** 1496–500
Cummins H Z 1973 *Photon Correlation and Light Beating Spectroscopy* eds H Z Cummins and E R Pike (New York and London: Plenum Press) pp 285–330
Jakeman E 1970 *J. Phys.* A: *Gen. Phys.* **3** 201–15
—— 1973 *Photon Correlation and Light Beating Spectroscopy* eds H Z Cummins and E R Pike (New York and London: Plenum Press) pp 75–149
Jakeman E, Oliver C J and Pike E R 1970 *J. Phys.* A: *Gen. Phys.* **3** L45–8
—— 1971 *J. Phys.* A: *Gen. Phys.* **4** 827–35
Jakeman E and Pike E R 1969 *J. Phys.* A: *Gen. Phys.* **2** 411–2
Koppel D E 1971 *J. Appl. Phys.* **42** 3216–25
Lachs G 1971 *J. Appl. Phys.* **42** 602–9
Mandel L 1972 *J. Appl. Phys.* **43** 258
Saleh B E A 1975 *J. Appl. Phys.* **46** 943–5