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Clipped correlation of integrated intensity fluctuations of gaussian light

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Abstract. Experimental confirmation is presented of recent theoretical results of Jakeman on the time averaging of photon-counting fluctuations when utilizing clipping techniques for autocorrelation. The effects of finite electronic resolving time (dead-time correction) and finite receiving apertures (spatial-coherence correction) are included.

1. Introduction

Measurements of the intensity fluctuations of laser light scattered by protein molecules undergoing Brownian motion in solution have been used for the determination of their diffusion coefficients. Dubin *et al.* (1967) determined the diffusion coefficients of a number of biological macromolecules using a wave analyser to study the intensity-fluctuation spectra. More recently Foord *et al.* (1969, 1970) and Pike (1969) have described the use of a clipping digital correlator to analyse the photon-counting fluctuations of scattered laser light. This technique offers the advantages of parallel processing of the information, compared with the single-channel operation of the wave analyser.

The 'single-clipped' correlation function given by Foord *et al.* (1970) for gaussian–lorentzian light required a factor to allow for the experimental effects of finite time and space averaging. Both these effects give rise to loss of correlation and hence, other things being equal, to loss of measurement accuracy. For unclipped autocorrelation functions this term could be factorized into spatial and temporal parts. The spatial part has been investigated by Scarl (1968) and by Jakeman *et al.* (1970). The temporal part has been calculated by Jakeman and Pike (see Pike 1969). For clipped correlation functions this factorization does not occur in general, but in the absence of any spatial-coherence correction the temporal effects have been evaluated by Jakeman (1970) for single- and double-clipped cases at clipping levels of zero. In this paper we present an approximate analysis of the combined effects of spatial coherence and clipping level as a function of count rate taking into account also the presence of a dead-time in the apparatus. The formulae are used firstly to correct experimental results showing the dependence of the single-clipped autocorrelation function on clipping level in the limit of small sample time, and secondly in the experimental verification of the dependence of the single-clipped and double-clipped correlation functions on sample time when clipping is carried out at zero. A preliminary result of this work was given by Pike (1970), the aim is to obtain a complete understanding of experimental results obtained using these new techniques.

2. Theory

In this section we present formulae which can be used to correct the experimental measurements for dead-time effects and for the effect of finite aperture sizes and thus to enable a direct comparison with the theory of the effects of integration to be made.

In an earlier paper (Jakeman 1970) it was shown that when dead-time and finite-detector-area effects are neglected, the autocorrelation function of photon-counting fluctuations of gaussian-lorentzian light is determined by the generating function $Q_L(s, s')$ of the joint integrated intensity fluctuation distribution

$$Q_L(s, s') = \frac{P(s)P(s')Q_L(s)Q_L(s')}{P(s)P(s')-Q_L(s)Q_L(s')|g_L^{(1)}(\tau)|^2}. \tag{1}$$

Here $Q_L(s)$ is the generating function for the single integrated intensity fluctuation distribution of gaussian–lorentzian light (Bédard 1966, Jakeman and Pike 1968)

$$Q_L(s) = \exp(\gamma)\left\{\cosh y+\tfrac{1}{2}\left(\frac{\gamma}{y}+\frac{y}{\gamma}\right)\sinh y\right\}^{-1} \tag{2}$$

where

$$y^2 = \gamma^2+2\gamma s\langle E\rangle \tag{3}$$

and

$$\gamma = \Gamma T. \tag{4}$$

$\langle E\rangle$ is the mean intensity integrated over the sample time T and Γ the reciprocal coherence time characterizing the field or first-order optical correlation function

$$g_L^{(1)}(\tau) = \exp(-\Gamma|\tau|+i\omega_0\tau) \tag{5}$$

ω_0 being the centre frequency of the lorentzian spectrum. $P(s)$ is defined in terms of equations (3) and (4) by

$$P(s) = \left\{\frac{1}{2}\left(\frac{\gamma}{y}-\frac{y}{\gamma}\right)\sinh y\right\}^{-1}. \tag{6}$$

If $n(t)$ is the number of counts arriving in the interval T centred at time t, and the clipped photon count is defined by

$$\begin{aligned} n_k(t) &= 1 \qquad \text{if } n(t) > k \\ &= 0 \qquad \text{if } n(t) \leqslant k \end{aligned} \tag{7}$$

then the normalized autocorrelation function of photon-counting fluctuations with arbitrary generating functions $Q(s)$, $Q(s, s')$ is given by

$$g^{(2)}(\tau) = \frac{\langle n(0)n(\tau)\rangle}{\bar{n}^2} = \left.\frac{\partial^2 Q(s, s')}{\partial s\partial s'}\right|_{s=s'=0}. \tag{8}$$

The corresponding quantity when clipping is carried out in one channel at an arbitrary level k is (Jakeman and Pike 1969)

$$g_k^{(2)}(\tau) = \frac{\langle n_k(0)n(\tau)\rangle}{\bar{n}_k\bar{n}} = \frac{1}{\bar{n}\bar{n}_k}\left\{\bar{n}+\sum_{m=0}^{k}\frac{(-1)^m}{m!}\left(\frac{\partial}{\partial s}\frac{\partial^m}{\partial s'^m}\right)Q(s, s')\bigg|_{\substack{s=0\\s'=\alpha}}\right\} \tag{9}$$

where $\bar{n} = \alpha\langle E\rangle$ is the mean count per sample and α is the quantum efficiency of the detector. This we call the 'single-clipped' correlation function. A further quantity of interest is the 'double-clipped' correlation function. When clipping is carried out at zero in both channels we obtain (Jakeman and Pike 1969)

$$g_{00}^{(2)}(\tau) = \frac{\langle n_0(0)n_0(\tau)\rangle}{\bar{n}_0^{\,2}} = \frac{1-2Q(\alpha)+Q(\alpha,\alpha)}{\bar{n}_0^{\,2}}. \tag{10}$$

The mean clipped count per sample $\bar{n}_k$ is given by

$$\bar{n}_k = 1 - \sum_{m=0}^{k} \frac{(-1)^m}{m!} \frac{\partial^m}{\partial s^m} Q(s)\big|_{s=\alpha}. \tag{11}$$

The formulae (8)–(11) apply only in the absence of dead-time effects and were evaluated for gaussian–lorentzian light using relations (1)–(6) in the earlier paper (Jakeman 1970). In figures 2 and 3 $g_0^{(2)}(\tau)$ and $g_{00}^{(2)}(\tau)$ are plotted as functions of T for the case $\tau = T$ (ie the first channel). These curves do not, of course, contain the effects of dead times in the electronic instrumentation or of finite aperture sizes in the optical system, and the corrections due to these factors which must be applied to the experimental results before comparison with figures 2 and 3 will now be considered.

2.1. *Aperture correction*

The effect of finite apertures on autocorrelation measurements has been considered recently, for a system with cylindrical symmetry, by Jakeman *et al.* (1970) who showed

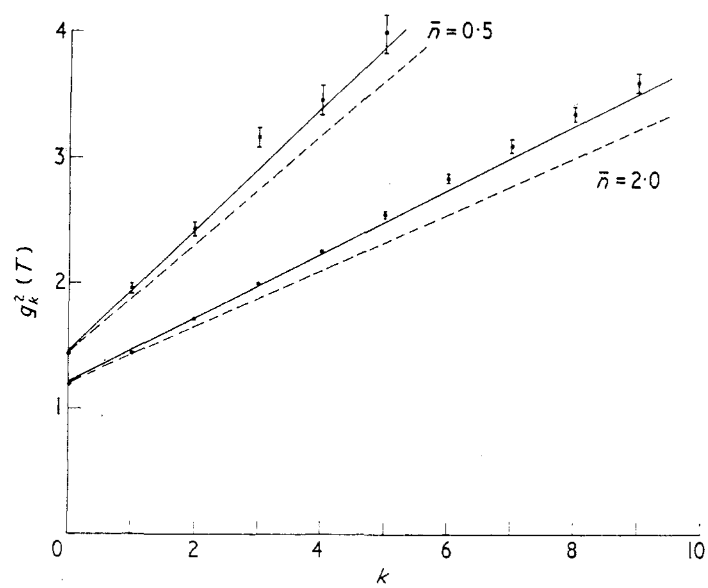

Figure 1. The dependence of the correlation coefficient $g_k^{(2)}(T)$ for clipping at a level k in one channel, as a function of k. Two values of the mean count rate (0·5 and 2·0) are considered. For comparison the theoretically predicted values are also given after (i) a simple correction for loss of spatial coherence due to finite apertures (broken line) and (ii) after a more accurate correction for these effects (full line). These data have also been corrected for small dead-time effects.

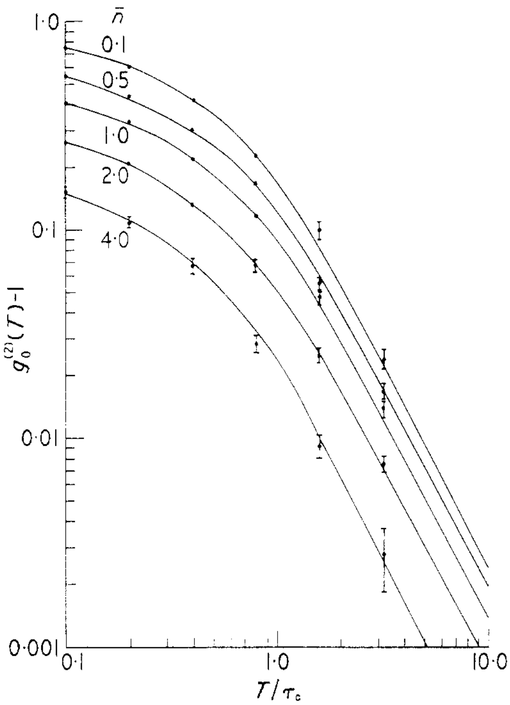

Figure 2. The dependence of the correlation coefficient $g_o^{(2)}(T)-1$, for clipping at zero in one channel, as a function of the ratio of the sample time T to the coherence time τ_c for various mean count rates. The data are corrected for spatial-coherence and dead-time effects.

that the correlation function defined by equation (8) above takes the form

$$g^{(2)}(\tau) = 1+f(A)\left(\frac{\sinh\gamma}{\gamma}\right)^2 |g^{(1)}(\tau)|^2 \tag{12}$$

where

$$f(A) = \sum_{s=0}^{\infty}\left(\frac{(2s+2)!}{\{(s+1)!\}^2(s+2)!}\right)^2(-1)^s(\tfrac{1}{2}\kappa R)^{2s}. \tag{13}$$

Here $\kappa = k_0S/2Z$, k_0 being the wavevector of the light, S and R the radii of the scattering volume and the photocathode or receiver aperture respectively and Z the mean distance of the scattering volume from the detector. Extending the calculation of the earlier paper to evaluate clipped autocorrelation functions is difficult owing to the form of equations (9)–(11). However, it is possible to devise an approximation analogous to that used by Bédard *et al.* (1967) in connection with the sample-time dependence of photon-counting distributions.

If the detector area is so large that it can be resolved into a large number N of coherence areas (defined roughly by $\kappa R \simeq 1$ in the above notation) then the distributions of photons falling on the subareas will be virtually independent and the generating functions characterizing the entire system will be simple products over those characterizing the subareas (see for example Lachs 1965, Perina 1967). Thus the generating function of the joint integrated intensity distribution of gaussian–lorentzian light falling on a photocathode of N coherence areas is $\{\tilde{Q}_L(s, s')\}^N$, where $\tilde{Q}_L(s, s')$

is given by equations (1)–(6) with $\langle E\rangle$ replaced by $\langle E\rangle/N$. Although this result is exact only in the limit $N\to\infty$, it can be used, following the procedure of Bédard *et al.* (1967), as an approximation to the true generating function for all detector areas if N is replaced by a nonintegral parameter η which is adjusted to give the correct lowest order autocorrelation function (12). It is not difficult to show that

$$\eta^{-1} = f(A). \tag{14}$$

Since the resulting generating function

$$\{\tilde{Q}_{\mathrm{L}}(s, s')\}^{\eta} \tag{15}$$

where $\tilde{Q}_{\mathrm{L}}(s, s')$ is now given by equations (1)–(6) with $\langle E\rangle$ replaced by $\langle E\rangle/\eta$ and η is defined by equation (14), is exact in the limits $\eta\to 1$, $\eta\to\infty$ and yields the correct intensity autocorrelation function for all η, it might be expected to generate at least the lowest moments of the joint photon-counting distribution with a reasonable degree of accuracy.

In the case of clipping at zero the appropriate correction can now be derived straightforwardly from equations (9) to (11) and (14) and (15). For present purposes it is sufficient to calculate the aperture correction for the general case of single clipping at k in the limit of small sample time ($\gamma\to 0$); equation (15) then reduces to

$$\{(1+sx)(1+s'x)-ss'x^2|g^{(1)}(\tau)|^2\}^{-\eta} \tag{16}$$

where $x=\bar{n}/\eta$, η is defined by equation (14) and $g^{(1)}(\tau)$ is given by equation (5) for a lorentzian spectrum. Substituting into equations (9), (10) and (11) leads to

$$g_k^{(2)}(\tau) = 1+\frac{1}{\eta}\frac{\Sigma_{m=0}^{k}(\bar{n}-m)\{(\eta)_m/m!\}x^m(1+x)^{-(\eta+m+1)}}{1-\Sigma_{m=0}^{k}\{(\eta)_m/m!\}x^m(1+x)^{-(\eta+m)}}|g^{(1)}(\tau)|^2 \tag{17}$$

and

$$g_{00}^{(2)}(\tau) = \frac{1-2(1+x)^{-\eta}+\{(1+x)^2-x^2|g^{(1)}(\tau)|^2\}^{-\eta}}{\{1-(1+x)^{-\eta}\}^2} \tag{18}$$

where

$$(\eta)_m = \eta(\eta+1)\ldots(\eta+m-1).$$

2.2. *Dead-time corrections*

It has been shown (De Lotto *et al.* 1964) that the Poisson emission probability of photoelectrons due to an integrated intensity E falling on the photocathode of a detector is modified by the presence of dead-time effects as follows†:

$$\frac{(\alpha E)^n}{n!}\exp(-\alpha E)\to\frac{(\alpha E)^n}{n!}\exp(-\alpha E)\left\{1+n(\alpha E-n+1)\frac{\tau_{\mathrm{D}}}{T}+\mathrm{O}\left(\frac{\tau_{\mathrm{D}}}{T}\right)^2\right\} \tag{19}$$

where $\tau_{\mathrm{D}}\ll T$ is the dead time after each registration of the arrival of a photon. The joint photon counting and integrated intensity fluctuation distributions are related through the equation (retaining terms up to first order in τ_{D}/T only)

$$p(n,m) = \int_0^\infty \mathrm{d}E\int_0^\infty \mathrm{d}E'\,\frac{(\alpha E)^n}{n!}\frac{(\alpha E')^m}{m!}\exp\{-\alpha(E+E')\}P(E,E')$$
$$\times\left(1+\frac{\tau_{\mathrm{D}}}{T}\{n(\alpha E-n+1)+m(\alpha E'-m+1)\}\right) \tag{20}$$

† Assuming that the electronic processing time is larger than τ_{D} so that dead times do not overlap samples.

where

$$Q(s, s') = \langle \exp\{-(sE+s'E')\}\rangle = \int_0^\infty \mathrm{d}E \int_0^\infty \mathrm{d}E' \exp\{-(sE+s'E')\}P(E, E'). \quad (21)$$

It is both intuitively obvious and evident from equation (20) that the quantity

$$g_{00}^{(2)}(\tau) = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} p(n, m) = (1-2p(0)+p(0,0))/\bar{n}_0^2 \quad (22)$$

is unaffected by the presence of a dead time. The normalized, single-clipped autocorrelation defined by

$$g_k^{(2)}(\tau) = \frac{1}{\bar{n}_k \bar{n}} \sum_{k=0}^{\infty} \sum_{m=k+1}^{\infty} np(n, m) \quad (23)$$

is changed, however. The right hand side of equation (23) may be written, using equations (20) and (21), in the form

$$\frac{1}{\bar{n}_k \bar{n}} \Bigg(\bar{n} - \sum_{m=0}^{k} \Bigg[-\frac{\partial}{\partial s}\left(\frac{\partial}{\partial s'}\right)^m \frac{Q(s,s')}{m!} + \frac{\tau_\mathrm{D}}{T}\Bigg\{ -\frac{\partial^2}{\partial s^2}\left(-\frac{\partial}{\partial s'}\right)^m \frac{Q(s,s')}{m!}$$
$$- \frac{m}{m!}\frac{\partial}{\partial s}\left(-\frac{\partial}{\partial s'}\right)^{m+1} Q(s,s') + \frac{m(m-1)}{m!}\frac{\partial}{\partial s}\left(-\frac{\partial}{\partial s'}\right)^m Q(s,s')\Bigg\}\Bigg]_{\substack{s=0\\ s'=\alpha}}\Bigg) \quad (24)$$

which replaces the right hand side of equation (9). This can be evaluated in principle using equation (15) but we consider here only the case $\gamma \to 0$, $\eta = 1$. Substituting equation (16) with $\eta = 1$ into equation (24) leads to

$$g_k^{(2)}(\tau) = 1 + \frac{1+k}{1+\bar{n}}|g^{(1)}(\tau)|^2 - \frac{\tau_\mathrm{D}}{T}\frac{\bar{n}(1+k)}{(1+\bar{n})^2}|g^{(1)}(\tau)|^2$$
$$\times\{2+2\bar{n}-k+|g^{(1)}(\tau)|^2(k-2\bar{n})\}. \quad (25)$$

For small values of τ the fractional dead-time correction to the $|g^{(1)}(\tau)|^2$ term is $-\tau_\mathrm{D}2\bar{n}/T(1+\bar{n})$. This suggests that the validity of the approximation should be independent of k. However, examination of the intermediate stages of the calculation casts doubt on this conclusion. For arbitrary τ the fractional correction appearing in equation (25) becomes large and positive for $k \gg 2\bar{n}+2/(1-|g^{(1)}(\tau)|^2)$ and becomes large and negative for $\bar{n} \gg \frac{1}{2}k - 1/(1-|g^{(1)}(\tau)|^2)$ so that the approximation is not good in these regions.

3. Apparatus and methods

The experimental arrangement used in this work was essentially the same as that used in earlier publications (Foord *et al.* 1969, 1970, Pike 1969). As previously, light from a Spectra Physics 125 He–Ne laser was scattered from a solution of protein molecules undergoing Brownian motion. The protein used was haemocyanin (murex trunculus) which can be prepared in a monodisperse form and has been shown to have a lorentzian spectrum with gaussian statistics as required by the theory (Foord *et al.* 1970). The concentration of the solution was made sufficiently high (5 mg ml^{-1}) for the scattered intensity to be adequate but without concentration effects becoming apparent, which might modify the statistics (Foord *et al.* 1970). The light scattered from the focussed laser beam was collimated by apertures at the source and detector, the latter being an ITT FW130 selected for its good photon-counting performance and low dark-count rate. The fraction of a coherence area subtended by the detector was obtained by measurement of the photon-counting distribution. Calculation of the second factorial moment of this distribution enabled the coherence area correction to be obtained since, in this case, temporal and spatial corrections factorize (Jakeman *et al.* 1970).

For each experiment it was necessary to consider the effects discussed above, of spatial coherence (aperture) correction and dead-time correction before studying the effects of time averaging. As far as possible these corrections were investigated separately and various experimental limits were used to satisfy these requirements.

4. Results and discussion

In order to investigate the correlation coefficients at different clipping levels it was necessary to use an aperture of size appreciable with respect to a coherence area so that high enough count rates could be attained. The dead-time correction could be minimized by choosing a sample time (300 μs) much greater than the system dead time (1 μs). Even with high count rates ($\bar{n} = 2$) and high clipping levels ($k = 8$) the clipped counts were then only affected by a few percent. The theory was first tested for unaveraged values ($T \to 0$). To remove the integration effect of finite sample times the scattering angle was reduced to 22° 10′ giving a coherence time by 3·8 ms so that $\gamma = 0{\cdot}079$. Under these conditions the temporal correction factor is very close to unity.

The first correlation coefficient (delay equal to sample time) of the single-clipped autocorrelation function $g_k^{(2)}(T)-1$ was now measured as a function of clipping level k for count rates of $\bar{n} = 0{\cdot}5$ and 2·0. The data are plotted in figure 1 after correction for dead-time effects. The error bars shown correspond to the statistical spread in the results over several runs. This accuracy is determined by the clipping level and the experiment duration which was 30 s for $k < 8$ and 300 s for $k = 8$ or 9. If the correction factor $f(A, T, \bar{n}, k)$ factorized so that the correlation coefficient for single-clipping at k could be written as

$$g_k^{(2)}(T)-1 = f(A)\frac{1+k}{1-\bar{n}}\exp(-2K^2D_TT) \tag{26}$$

with $f(A)$ independent of k, $\bar{n}$ and T, then the predicted coefficients would be as shown by the broken lines on figure 1. For high k this approximation is manifestly not correct. If one uses the more accurate equation (17) then the full line in figure 1 is obtained. This is in much closer agreement with the observed data, though for higher k and $\bar{n}$ the prediction still lies slightly below the observed data.

The effects of integration were next investigated. The scattering angle was changed to 90° giving a coherence time of 270 μs for the scattered light. The first correlation coefficient with single- and double-clipping at zero was measured over a range of sample times from 27 μs to 864 μs and for count rates of 0·1, 0·5, 2·0 and 4·0 counts per sample time. Comparison of experiment with theory was chiefly affected by the uncertainty of $\bar{n}$ for high $\bar{n}$ and short sample time due to the increasingly large dead-time correction. This correction only applies strictly in the limit of $\gamma \to 0$ which is where the dead time has most effect. As γ increases the correction becomes very small ($\simeq$ 1% at $\gamma = 3$ and $\bar{n} = 4$) and the inaccuracy introduced by using the same approximation becomes negligible. Next the data were corrected for the coherence-area effect using the approximation that has already been verified. This too only applies in the limit of $\gamma \to 0$ but, since the correction is only small for high γ, by including the temporal correction in the spatial term a reasonable approximation could be made.

The statistical accuracy of the data points was governed by the length of experiment ($\simeq$ 100 s) and varied between 1% for low $\bar{n}$ and γ to 20% for high $\bar{n}$ and γ. In the high $\bar{n}$, high γ region the statistical uncertainties became greater than the dead-time and spatial-coherence corrections so no further refinements of these latter corrections were necessary. The statistical errors were obtained by making several readings of each quantity.

In figure 2 we compare the corrected data for the first correlation coefficient, with clipping at zero in one channel, as a function of count rate and sample time. The full curves are those calculated by Jakeman (1970). The agreement is everywhere within the statistical errors justifying the use of the corrections made and indicating the validity of the theory. In figure 3 a similar result is shown for the double clipped correlation coefficient $g_{00}^{(2)}(T)-1$. In this case also the agreement with the theory is seen to be good within experimental errors.

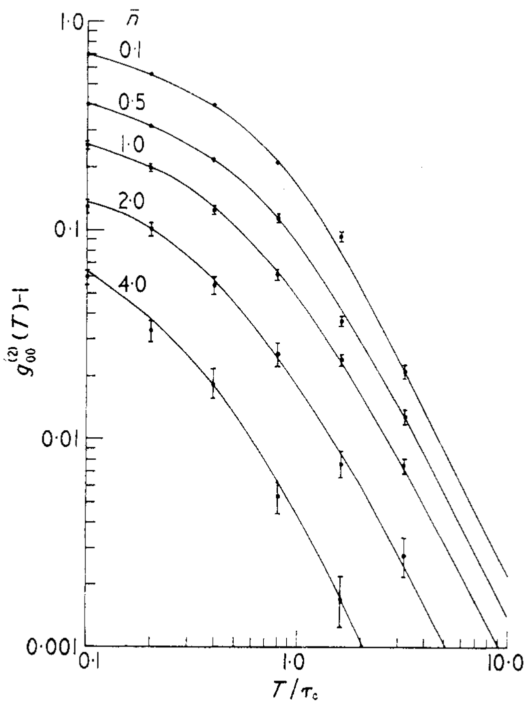

Figure 3. The dependence of the correlation coefficient $g_{00}^{(2)}(T)-1$, for clipping at zero in both channels, as a function of the ratio of the sample time T to the coherence time τ_c for various mean count rates. The data are corrected for spatial-coherence effects.

5. Conclusions

By applying corrections for dead-time and for spatial-coherence effects an experimental confirmation of the recent results of Jakeman (1970) on the time averaging of photon-counting fluctuations in clipped autocorrelation is obtained. Thus theoretical results have now been given which enable a satisfactory complete interpretation of the experimental clipped autocorrelation functions to be made for gaussian–lorentzian light sources.

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