



Effects of Temporal Averaging on Digital Correlation Functions

by

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Introduction

It is a well known fact from signal theory that any temporal averaging of a signal for further signal processing introduces certain distortions compared to the theoretical expectations for the case of "instantaneous" signals being processed. Still, such averaging is an unavoidable property of any measuring apparatus being used, either due to the inability to measure "instantaneous" signals and therefore the required use of non-zero "sampling times" for digital data, or due to the required, non-zero, integration times in an analog pre-processer (e.g. an A/D-converter, for example).

Though this effect is not necessarily identical with the well known aliasing problems occuring due to insufficiently fast sampling of a signal with a given maximum frequency (with the well known results of the famous Shannon-Theorem for this particular case), it does indeed show at least some similarity to this effect.

In this text, a detailed mathematical approach is used to outline the effects of temporal averaging for the case of photon correlation functions (although it could be extended to A/D-converted input signals from other sources easily), a comparison of the influence of the "Triangular Averaging Distortions" on different Multiple Tau Correlation schemes, their noise performance, as well as a detailed description on how to avoid that "Triangular Averaging Distortions" have <u>any</u> noticable effect on the results obtained from data analysis of the correlation functions measured.

1.0 Sampling

Consider a stream of digital pulses in time, each identifying, for example, the successful conversion of an incoming (randomly distributed over time, though with non-uniform conditional probability of occurrence with time difference – or, in simple words, they "correlate") photons into a binary digital information by an appropriate single photon detector with infinitely good temporal resolution. If each of this pulses was tagged with a time-tag, again with infinitely good temporal resolution, this pulse stream could as well be seen as a continuous stream of "arrival times" of photons on this (just to mention this, of course non-existing in reality) detector. Thus we would define a series of pulses at certain, distinct arrival times

$$\partial_{t_0}, \partial_{t_1}, \partial_{t_2} \dots \partial_{t_N} \quad \text{with } t_0 < t_1 \dots < t_N \tag{1.0}$$

or, more handy for further computations, a series of time differences

$$t_N - t_{N-1}, t_N - t_{N-2}, t_N - t_0, t_{N-1} - t_{N-2}, t_{N-1} - t_{N-3} \dots t_1 - t_0$$
(1.1)

Clearly, the probability that any $(t_k - t_j) = (t_l - t_m) \rightarrow zero$, simply due to the random nature of arrival times on the detector and the (presumed) infinite temporal resolution of the detector.

The next task would be to compute a temporal auto correlation function (it should be noted that this holds true for cross correlations as well) from this stream of data.

Using the usual definition of an (unnormalized) temporal auto correlation function of a process u(t)

$$G(T) = \int_{0}^{\infty} u(t)u(t-T)dt$$
 (1.2)

or, the more practical approach of using an approximation to infinite upper bound integral above and replacing the upper bound by a finite measurement time

$$G^{*}(T) = \int_{0}^{t_{\text{max}}} u(t) u(t-T) dt$$
(1.3)

the above time series would now have to be converted in a rather simple "signal", namely

$$u(t)u(t-T) = 1\Big|_{T=t_k-t_{ki}}, 0\Big|_{else}$$
(1.4)

or, it becomes one whenever the time difference between two pulses of the above time series corresponds <u>exactly</u> to T and zero for every other case.

Actually, the result of performing the upper bounded integration of (1.3) and thus using all N events in the time series under consideration would be rather disappointing - the correlation function would show non-zero results for at maximum N^2 lag times and zero for all others. Not much to work with.

Clearly this would be different if the integration was performed over an infinite measurement time – in this case, the above scheme would indeed recover the correct auto correlation function for every lag time T. The cost is still prohibitive – infinity is a pretty long time to wait for.

Still, and this is the good news, a careful inspection of the resulting correlation function approximation (1.3) would yield to the fact that the number of non-zero lag times in any given lag time interval would still be proportional to the average correlation function to be expected in this lag time interval from (1.2).

The reason is, that although the probability of two pulse pairs having <u>exactly</u> the same time difference is zero, the probability that the time difference of two pulse pairs fall within an certain, non-zero, interval centred around a given time difference is not at all zero (obviously, if it was, no pulse pair would ever appear). Thus, any further approach should bin events falling into certain time intervals and compute correlation functions from these time intervals, rather than from the exact times.

Using time intervals of length τ , or in technical terms a "sampling time", leads to a simple integration all over a certain "sampling time" the instantaneous signal u(t) resp. a simple counting of events within this sampling time for binary input signals and a resulting time integrated μ_i , can be defined as

$$\mu_{i} = \frac{1}{\tau} \int_{(i-1)\tau}^{i\tau} u(t) dt$$
(1.5)

With this, and using (1.2), the resulting time averaged correlation function reads

$$G_{\mu}(i) = \int_{0}^{\tau} \int_{0}^{\tau} G(i\tau + t - t') dt dt'$$

$$= \int_{-\tau}^{\tau} G(i\tau + t) (\tau - |t|) dt$$
(1.6)

if the integration over t' is performed. For the normalized time integrated correlation function, the above integral reads

$$g_{\mu}(i) = \frac{1}{\tau^2} \int_{-\tau}^{\tau} g(i\tau + t')(\tau - |t'|) dt'$$
(1.7)

Obviously, the resulting correlation of the time integrated process is itself the temporal average of the correlation function of the original process u(t) over the sampling time τ and centred at lag time $i\tau$, however with a triangular weight imposed. It is noteworthy, that is was for this triangular shaped weight in the above integration, the terms "Triangular Averaging Error", resp. "Triangular Averaging Distortion" were introduced in the literature.

2.0 Triangular Averaging Errors

The triangular weight in (1.6) introduces a certain error in $G_{\mu}(i)$ compared to G(T) for every correlation function that is non-constant, resp. non-linear, or, more precisely (and this will become important later on) has a non-zero second derivative.

2.1 Exponential Correlation Functions

In the special case of fully normalized exponential correlation functions, the pre-factor can quickly be computed as

$$g_{2}(t) = \beta e^{-2\Gamma t}$$

$$g_{\mu}(k) = \frac{\beta}{\tau^{2}} \int_{-\tau}^{\tau} e^{(-2\Gamma k \tau - 2\Gamma t)} (\tau - |t|) dt$$
(2.1)

And, after performing the integration accordingly, this yields

$$g_{\mu}(k) = \beta f(2\Gamma\tau) e^{(-2\Gamma k\tau)}$$
$$= f(2\Gamma\tau) g_{2}(t)$$
(2.2)

with

$$f(x) = \frac{1}{x^2} \left(2\cosh(x) - 2 \right) \sim 1 + \frac{x^2}{12} \quad (for small x, <5\% \ error \ for \ x = 2)$$

Obviously, the "Triangular Averaging Error", represented here as f(x) is a constant pre-factor on the resulting time averaged correlation function and hence does not alter the shape of the correlation function, but acts as a simple scale transformation only. As a result, for the case of a constant sampling time being used over the complete range of lag times there are no "Triangular Averaging Distortions" for a single exponential correlation function at all.

It should be noted that the above equations hold true in the case of multi-exponential correlation functions, because we can express both, the fully normalized intensity correlation function, as well as the field correlation function as a sum of exponentials.

However, as can be seen, the pre-factor will no longer stay constant in this case and "Triangular Averaging Distortions" (though at very small magnitude) will follow :

$$g_{2}(t) = \beta \left(\sum_{j} c_{j}^{(2)} e^{-\Gamma_{j}t} \right)^{2} = \beta \left(c_{1}^{(2)} c_{1}^{(2)} e^{-2\Gamma_{1}t} + c_{1}^{(2)} c_{2}^{(2)} e^{-(\Gamma_{1} + \Gamma_{2})t} + \dots + c_{1}^{(2)} c_{N}^{(2)} e^{-(\Gamma_{1} + \Gamma_{N})t} + \dots + c_{N}^{(2)} c_{N}^{(2)} e^{-2\Gamma_{N}t} + c_{N}^{(2)} c_{N}^{(2)} e^{-2\Gamma_{N}t} \right)$$
$$= \beta \sum_{l} c_{l}^{(1)} e^{-2\Gamma_{l}t}$$

In this case

$$g_{\mu}(k) = \frac{\beta}{\tau^{2}} \int_{-\tau}^{\tau} \sum_{l} c_{l}^{(1)} e^{-2\Gamma_{l}k\tau - 2\Gamma_{l}t} (\tau - |t|) dt$$

$$= \frac{\beta}{\tau^{2}} \sum_{l} c_{l}^{(1)} e^{-2\Gamma_{l}k\tau} \int_{-\tau}^{\tau} e^{-2\Gamma_{l}t} (\tau - |t|) dt$$

$$\sim \beta \sum_{l} c_{k}^{(1)} e^{-2\Gamma_{l}k\tau} (1 + \frac{(2\Gamma_{l}\tau)^{2}}{12})$$

(2.4)

As already mentioned, in the case of multi-exponential correlation functions, the prefactor is no longer a constant (though very small in variation), because in detail

(2.5)

$$\sum_{l} c_{k}^{(1)} e^{-2\Gamma_{l}k\tau} \left(1 + \frac{(2\Gamma_{l}\tau)^{2}}{12}\right) = c_{1}^{(1)} e^{-2\Gamma_{l}k\tau} \left(1 + \frac{(2\Gamma_{1}\tau)^{2}}{12}\right) + c_{2}^{(1)} e^{-2\Gamma_{2}k\tau} \left(1 + \frac{(2\Gamma_{2}\tau)^{2}}{12}\right) \dots$$

$$= \sum_{l} c_{k}^{(1)} e^{-2\Gamma_{l}k\tau} + \left(c_{1}^{(1)} e^{-2\Gamma_{l}k\tau} \frac{(2\Gamma_{1}\tau)^{2}}{12} + c_{2}^{(1)} e^{-2\Gamma_{2}k\tau} \frac{(2\Gamma_{2}\tau)^{2}}{12} + \dots\right)$$

and

$$\sum_{l} c_{k}^{(1)} e^{-2\Gamma_{l}k\tau} \text{ is not proportional to } (c_{1}^{(1)} e^{-2\Gamma_{l}k\tau} \frac{(2\Gamma_{1}\tau)^{2}}{12} + c_{2}^{(1)} e^{-2\Gamma_{2}k\tau} \frac{(2\Gamma_{2}\tau)^{2}}{12} + \dots)$$

2.2 Hyperbolic Correlation Functions

For the case of hyperbolic correlation functions, the pre-factor can be computed using

$$g_{2}(t) = \beta \frac{1}{\omega + t}$$

$$g_{\mu}(k) = \frac{\beta}{\tau^{2}} \int_{-\tau}^{\tau} \frac{1}{k\tau + \omega + t} (\tau - |t|) dt$$
(2.6)

which can be re-written as

$$g_{\mu}(k) = g_{2}(t) \frac{1}{\tau^{2}} \int_{-\tau}^{\tau} \frac{1}{1 + \frac{t}{k\tau + \omega}} (\tau - |t|) dt$$
(2.7)

and, after performing the integration accordingly and expanding the resulting $\ln(1+\frac{\tau}{\omega+t})$ and $\ln(1-\frac{\tau}{\omega+t})$ terms up to the fourth order this yields

$$g_{\mu}(k) \sim g_{2}(t) \left(1 + \frac{1}{6} \frac{\tau^{2}}{(t+\omega)^{2}}\right) = g_{2}(t) \left(1 + \frac{1}{6\beta^{2}} (\tau g_{2}(t))^{2}\right)$$
 (2.8)

Different from the case of a single exponential correlation function, the pre-factor is non-constant for the resulting time averaged correlation function of a hyperbolic, even for a constant sampling time τ .

In the definition of this text, an exponential correlation function only shows "Triangular Averaging Errors" as long as a constant sampling time is used, whereas a hyperbolic correlation function not only shows "Triangular Averaging Errors", but "Triangular Averaging Distortions" as well, because the pre-factor itself always is a function of the lag time. It should be mentioned, however, that the variation is negligible whenever $\tau^2 \ll \omega^2$, since $f(0) - f(\infty) = \frac{1}{6} \frac{\tau^2}{\omega^2}$. For $\tau^2 = (\frac{1}{10}\omega)^2$ for example, the pre-factor varies from ~1.0017 to 1 over the complete lag time range from $[0...\infty]$.

2.3 Undamped and Damped Cosine Correlation Functions

For the case of undamped cosine correlation functions, the pre-factor can be computed using

$$g_{2}(t) = \beta \cos(\omega t)$$

$$g_{\mu}(k) = \frac{\beta}{\tau^{2}} \int_{-\tau}^{\tau} \cos(\omega k \tau + \omega t) (\tau - |t|) dt$$
(2.9)

After performing the integration accordingly, the result reads

$$g_{\mu}(k) \sim g_{2}(t) \left(\frac{2 - 2\cos(w\tau)}{\tau^{2}\omega^{2}}\right)$$
 (2.10)

which can again be expanded to second order terms

$$g_{\mu}(k) \sim g_{2}(t) \left(1 + \frac{(w\tau)^{2}}{12}\right)$$
 (2.11)

However, such expansion requires that $(\tau \omega) \le 1$ due to the "non-decaying" nature of the undamped cosine function. Of course, this condition will be quickly violated for Multiple Tau Correlation schemes using increasing sampling times with lag time (see chapter 3.0), and for these, either higher order expansions than just second order, or (2.10) should be used instead.

For the case of damped cosine functions,

$$g_{2}(t) = \beta e^{-\Gamma t} \cos(\omega t)$$

$$g_{\mu}(k) = \frac{\beta}{\tau^{2}} \int_{-\tau}^{\tau} e^{-\Gamma k \tau - \Gamma t} \cos(\omega k \tau + \omega t) (\tau - |t|) dt$$
(2.12)

The usual approach of expanding the integration result to second order terms is a sufficiently accurate approximation, as long as $\omega < 2\Gamma$, independent of the sampling times used.

In either case, no matter if single-exponential, multi-exponential or hyperbolic correlation functions, the simple picture of a "Triangular Averaging Errors" fails anyway whenever non-constant sampling times are used along the lag time axis.

Such use of (many) non-constant sampling times is absolutely desired for statistical reasons though and exactly followed by the implementation of the "Multiple Tau Correlation" scheme. In this scheme, blocks of *k* correlation estimators are computed using a constant sampling time τ_k and the sampling time is increased (usually doubled) for the next block of *l* correlation estimators, now using a sampling time of τ_l etc. pp.

Typical implementations use blocks of 16 or 32 estimators for the first block and 8 or 16 estimators for all preceding blocks with the sampling time being doubled from block to block, with the "classical" Multiple Tau Correlation implementation first realised by K. Schätzel and ALV in the early 80's of last century already used a 16/8 channel per block scheme (for good reason).

In this case, the "Triangular Averaging Errors" still act as a constant pre-factor on the time averaged correlation function within each such block, but will of course be different from block to block due to the change (doubling) of the sampling time used. Here we are indeed faced with "Triangular Averaging Distortions", because the resulting time averaged correlation function is no longer a scale transformed equivalent of the correlation function of the instantaneous process.

The absolute deviations between the two resulting correlation functions can easily be computed using either the exact form or the approximation given in (2.4) for the case of a single exponential (again, this holds true for multi-exponential correlation functions in just the same way), in detail

$$g_{\mu}(i) - g_{2}(i\tau) = \frac{(2\Gamma\tau)^{2}}{12}\beta e^{(-2\Gamma i\tau)}$$
(3.0)

which shows a maximum of

$$\frac{\beta}{3e^2k^2} \sim \frac{4.5 \cdot 10^{-3}\beta}{k^2}$$
(3.1)

The below graph illustrates the "Triangular Averaging Distortions" for the case of a Multiple Tau Correlation scheme with an initial block of 16 estimators and preceding blocks of 8 estimators with doubled sampling time (MTC-16/8) and a Multiple Tau Correlation scheme with an initial block of 32 estimators and preceding blocks of eight estimators with doubled sampling time (MTC-32/16), both for $\beta = 1$ and a 2 Γ of 0.001.



Clearly, in neither case the "Triangular Averaging Distortions" are of an important magnitude for real-world correlation functions. Most experiments obtaining photon correlation experiments yielding exponential or correlation functions that can be reasonably well approximated by a sum of weighted exponential functions with potentially different amplitudes (such data from DLS, DWS, FCS etc.) would require substantial measurement times to reach absolute noise levels in the order of << 10⁻³, which would anyway be required to show significant, yet even just visible, influence of the "Triangular Averaging Distortions", no matter if a 16/8 Multiple Tau Correlation scheme or a 32/16 Multiple Tau Correlation scheme is used.

The use of even more channels with constant sampling time within a sampling time block, such as a 64/32 Multiple Tau Correlation scheme, can not at all be motivated on the basis of "Triangular Average Distortions" alone, keeping in mind that noise levels below << 10^{-4} are, to say the least, difficult to reach within still reasonable measurement times.

Even less a problem the "Triangular Averaging Distortions" get with a more realistic presumption of an experimental correlation function - if a multi-exponential correlation function with three components at equal amplitude and decay rates 0.00025, 0.001 and 0.004 (for the field correlation function) is presumed, the "Triangular Averaging Distortions" reduce even further (because the resulting correlation function is more "smooth" or, more mathematical, has a smaller second derivative), see the below graph



Quite obviously, while a noise level low enough to have significant influences of the "Triangular Averaging Distortions" become visible is difficult to reach in the case of a single exponential function already, this becomes even more difficult in most practical application for both, MTC-16/8 and MTC-32/16 in terms of the required measurement time. For this reason, it is hard to see why there should be any advantage of using a MTC-64/32 implementation just for the sake of reducing the "Triangular Averaging Distortions" to below 3×10^{-5} .

For hyperbolic correlation functions, such as in FCS experiments (2D-model), the "Triangular Averaging Distortions" are shown in the next graph:



And, for the sake of completeness, for the case of a double-hyperbolic correlation function (with a Γ -ratio of 1 : 5) the "Triangular Averaging Distortions" are shown in the next graph:



which, not too surprisingly, does no longer show much difference in the peak value of the "Triangular Averaging Distortions" compared to it's single-hyperbolic sibling. The same holds true for FCS using 3D-models, and those including triplet-state correlations.

Keeping in mind, that particularly FCS correlation data usually suffers from rather short measurement times compared to DLS or DWS data and thus significantly higher noise levels, "Triangular Averaging Distortions" can hardly be accounted as being anything near to "problematic" for these applications.

Drawbacks "High-Resolution" Multiple Tau Correlation Schemes

Obviously with some marketing strategy in mind, some other producers of correlator hardware refer to their implementations of 32/16 or 64/32 Multiple Tau Correlation Schemes as being of "high resolution" type. Unfortunately, this argument does not really hold on a closer investigation of the matter.

"High resolution", in a strict sense and not as a slightly exaggerated description of the plain fact that simply more correlation channels are used within the same lag time range, would require that the ability of resolving certain features of the correlation function, or even more realistically in the results of the data reduction used on the correlation function, was increased compared to the classical 16/8 Multiple Tau Correlation scheme. Actually, this is not the case, but just the opposite is true - the "resolution" of a 32/16 Multiple Tau Correlation scheme will at best be close to a 16/8 implementation, but for sure not better, and a 64/32 Multiple Tau Correlation scheme will perform even worse than a 32/16 Multiple Tau Correlation scheme.

From a data analysis point of view, there as well is hardly any need for more than 16/8 channels per "octave" (or doubling of the lag time), because these still ensure eight possible degrees of freedom per octave for the fit procedures used. As a matter of fact, the mathematical condition of the underlying fit problems (DLS/DWS/FCS ...) will not allow to reliably extract more than eight ... ten degrees of freedom even in a lag time range as large as 1 : 1000 without having practically noiseless data.

Thus, the "resolution" is mainly a matter of the statistical accuracy of the correlation estimators computed, less, but more precise correlation estimators are clearly the better choice to more, but significantly less accurate correlation estimators – and here a 32/16 or 64/32 Multiple Tau Correlation scheme falls short compared to the classical 16/8 Multiple Tau Correlation scheme – more, but significantly less accurate estimators will always be the result for identical measurement times.

But why is this so ? The route to the answer will directly lead us to the next topic ...

4.0 Noise on Correlation Functions

Systematic distortions, such as the "Triangular Averaging Distortions" form only one part of the total contribute to the total uncertainty in a correlation estimator at a certain sampling / lag time combination and for a given measurement time. As could be seen above, in most experimental situations, they do not even contribute significantly, not to speak about the fact that fully systematic distortions are the scientists dream anyway, because the can easily be corrected due to the systematic nature (see *Removing "Triangular Averaging Distortions" from Correlation Functions*).

Keeping in mind the fact that photon correlation experiments usually try to gather information on stochastic processes rather than deterministic signals and in addition are to be "measured" using detectors which as well show stochastic photon-to-pulse conversions, these two noise sources must be considered as well, if the significance of "Triangular Averaging Distortions" is to be analysed.

The statistics of the photon-to-pulse conversion is merely detector dependent and rather independent of the underlying statistics of the photon pulse stream itself. This is a must condition obviously – the detector has to convert the stream of photon into a stream of pulses such that the statistical properties of the photon stream is not altered other than in the absolute magnitude of first, second and/or higher moments.

If this was not the case, any following moment analysis, such as computing correlation functions, for example, would completely fail. For most single photon detectors, this holds true in the "linear response range", although some complications arise due the inherent self-correlations practically all single photon detectors show, such as dead-times and after-pulsing effects.

The usual approach is to only use selected such detectors which show smallest selfcorrelation and at small lag times only or to operate two detectors in a cross or pseudo-cross correlation scheme, hoping for the statistical independency of these self-correlations from one to another detector.

While true in most cases, even this presumption fails for very small lag times and certain classes of single photon detectors (e.g. Avalanche Photo Diode detectors due to their inherent light emission [ref 3]).

Nevertheless, even a completely noise-less, self-correlation-free, 100% efficient and instantaneously converting detector would still convert a stochastic signal, namely the signal under investigation, and no matter how many photons such a detector would be able to detect and convert in a given sampling time, it would still at best yield a noiseless conversion of the underlying stochastic process. In simple words : no matter how many photons are detected, the statistical accuracy of the resulting correlation function (and/or other types of moment analysis) can never be better than the underlying statistical process of the signal allows.

For this reason, the literature typically refers to two distinct sources of noise,

- "Photon Noise" or "Shot Noise", which is introduced by the finite number of photons available to sample the instantaneous (or time averaged) underlying signal
- "Diffusion Noise" or "Signal Noise", which describes the fact that the stochastic nature of the underlying process under investigation gives rise to another distinct "uncertainty" in the resulting correlation function for finite measurement times. Interestingly, the term "Diffusion Noise" was introduced because most photon correlation experiments are used to probe the diffusion of either molecules in a solvent (via light be scattered by these in DLS, or fluorescent light being emitted by these in FCS experiments) or the "diffusion" of light within a highly dense molecule/solvent combination (via multiple-scattering of light in DWS experiments). In a more general context, the use of "Signal Noise" instead of "Diffusion Noise" would be more precise.

Without further lengthy and detailed algebra (see [ref. 1] or [ref. 2] for all details), the contribution of each of the above noise sources to the variance of a (normalized) intensity correlation function for complex-gaussian amplitude statistics (such as for DLS, sometimes in FCS and most probably for DWS experiments) can be expressed as the expectation of the correlation function itself as follows

$$Var(g_{2}(k) = \frac{1}{M}\beta^{2}\left\{\sum_{i}|\chi_{i}|^{4} + \sum_{i}|\chi_{i}|^{2}|\chi_{i+2k}|^{2} + 2\beta|\chi_{k}|^{2}\sum_{i}|\chi_{i}|^{2} + 2\beta\chi_{k}^{2}\sum_{i}\chi_{i}\chi_{i+2k} + 2\beta\sum_{i}\chi_{i}\chi_{i+2k}^{2} + 4\beta|\chi_{k}|^{4}\sum_{i}|\chi_{i}|^{2} - 8\beta|\chi_{k}|^{2}\sum_{i}\chi_{i}\chi_{i+k} + \frac{2}{\beta n}[|\chi_{0}|^{2} + |\chi_{2k}|^{2} + 2\beta|\chi_{k}|^{2} + 2\beta\chi_{k}^{2}\chi_{2k} - 2\beta|\chi_{k}|^{4}] + \frac{1}{\beta^{2}n^{2}}[1 + \beta|\chi_{k}|^{2}]\right\}$$

$$(4.1)$$

(see footnote for comments on FCS)¹

With *M* being the total number of samples taken (and thus $M\tau_{\kappa}$ being the total measurement time for the sampling time/sampling time group τ_k), *n* being the number of detected photons per sampling time and $1+\beta |\chi_k|^2$ being the expectation of the normalized intensity correlation function at lag time point *k*. All above terms are strongly covariant signal noise terms, with the exception of the last term, which is the random photon noise contribution (and thus only present on the diagonal terms of the co-variance matrix) [ref 2].

Numerical computations can now be performed straightforward (alternatively, a closed form of (4.1) can be given presuming a certain functionality of $1+\beta |\chi_k|^2$ such as an exponential, for example [ref 1]. However, the above numerical approach is much less complicated whenever a more complex functionality is presumed, such as a sum of exponential, for example) by in addition equating that:

¹ For FCS, in particular for FCS on single molecules, severe complications arise due to the generally non-gaussian statistics for a very small average number of molecules in the illuminated beam, as well as secondary effects such as triplet states, photo-bleaching and "fluctuation of the average number of particles in the volume" effects (the "number fluctuations" inherent variance, so to say). The model shown herein is thus not directly applicable for the case of single molecule FCS experiments, yet not even easily adoptable for the FCS case.

Generally, correlation functions from FCS experiments show worse noise behaviour compared to DLS data, simply because the "long lag time tails" are much more pronounced for the hyperbolic decay behaviour of FCS correlation functions compared to the exponentials obtained in DLS experiments.

This further supports the strategy of quickly increasing the sampling time with the lag time, such as performed by the 16/8-channel Multiple Tau Correlation scheme, because successful estimator noise reduction, in particular of the covariant contribution of the diffusion noise, is strongly dependent on the sampling time used at a given lag time point and can not be reached with a larger number of constant sampling time channels working at smaller sampling times !

$$\chi_{k} = \sqrt{g_{2}(2k)}$$

$$|\chi_{k}|^{2} = g_{2}(2k)$$

$$|\chi_{0}|^{2} = \frac{1}{\tau_{k}^{2}} \int_{-\tau}^{\tau} (|\chi(t)|^{2} (\tau - |t|) dt$$
(4.2)

It is noteworthy, that whenever the measured normalized intensity correlation function is of sufficient accuracy and thus already a reasonable representation of the expectation, the estimators $g_2^{ex}(k)$ can safely be used instead of the $g_2(k)$ enabling the computation of the variances of an arbitrary normalized intensity correlation function, as long as the presumption of amplitudes with complex gaussian statistics of the underlying process holds true.

Equipped with these formulae, the to be expected variances (or Standard Deviations, simply by taking the square-root of the variances) for several, typical experimental situations can quickly be computed for any particular implementation of Multiple Tau Correlation schemes and compared to each other and the additional influence of "Triangular Averaging Distortions" can be discussed in comparison to the effective noise after a certain, finite, total measurement time.

To cover a rather broad range of experimental situations, the following types of correlation functions were considered :

- Single-exponential, fast decay time, smaller average count rate in the 25.000 counts/s regime this would typically occur in DLS experiments on small particles, such as proteins-monomers, for example.
- Multi-exponential, fast decay time to slow decay time, normal average count rate in the 250.000 counts/s regime – this would typically occur in most DLS experiments on multi-modal particles/molecules
- Massively multi-exponential, fast to slow decay times, count rates in the 250.000 counts/s regime – this would be a rather "limiting" approximation to "general" DLS experiments and a reasonable approximation for DWS backscattering experiments as well.

In the below graphs, the correlation function variance estimators are plotted for the above experimental conditions, each for a total measurement time of 100 s and 1000 s, for MT-16/8 and MT32/16 respectively (MT-64/32 implementation data is not shown – the Std.Dev behaviour is always worse than that of MT-32/16 anyway), along with the correlation function and the "Triangular Averaging Distortions" for the MT-16/8 scheme for comparison.



It clearly becomes visible, that for both cases of total measurement time plotted (100 s and 1000 s), the average Std.Dev (red and black graph) is considerable larger than the "Triangular Averaging Distortion" for the Multiple Tau 16/8 implementation.

Since the average Std.Dev scales with the total measurement time as $\sqrt{\frac{1}{\tau}}$ the

required total measurement time for obtaining Std.Dev. of the same magnitude as the "Triangular Averaging Distortion's" maximum (~ $7 \cdot 10^{-4}$), a total measurement time of approximately 3000 s would be required.

Additionally, it becomes visible that the MT-32/16 Multiple Tau implementation (black graphs) falls short in total Std.Dev. for practically the complete lag time range by a factor of $\sqrt{2}$ compared to the MT-16/8 Multiple Tau implementation (red graphs). While this effect can be compensated for random noise sources, such as the photon noise, by the doubled number of correlation estimators per lag time range, it can not for the signal noise sources, because this is not a random noise source, but shows strong covariance. For covariant noise, the increased number of correlation estimators is, however, of no further help [ref 2]. It should be noted, that the MT-64/32 implementation shows even worse average Std.Dev. compared to either MT-16/8 or MT-32/16.

The situation does not at all change for a strongly multi-exponential correlation function at 10 times higher count rate. In the next plot, the same information as above is plotted for a correlation function composed of four exponential at significantly different decay rate (0.1/ms, 1/ms, 10/ms and 100/ms) and 1:4:4:1 amplitudes.



While the noise performance of both implementations are about the same (with slight advantages for the MT-16/8 implementation) for smaller lag times – which is easy to explain by the fact that in the above presumed experimental condition the photon noise is extremely small due to the comparably high count rate presumed and the strongly covariant signal noise is the dominant noise source , MT-32/16 again falls short for lag times in the > 1 ms regime. Notably, the small contribution of the $\Gamma = 0.1/ms$ decay rate fluctuation suffers most from this fact.

As was to be expected, the "Triangular Average Distortions" are much smaller than the average Std.Dev., even for 1000 s measurement time and yet again, a total measurement time of more than 1 hour must be used to have the average Std.Dev. hitting the maximum contribution of the "Triangular Average Distortions".

As a last example, the "massively multi exponential" case is plotted, here using a hyperbolic correlation function at fast decay rate of $\Gamma = 100$ /ms. As already outlined, this is a good approximation to many DLS/DWS experiments and gives, noting the comments made above on FCS correlation functions and their estimator variances - at least some general insight into a typical noise distribution for FCS experiments as well (typically FCS experiments will show significantly lower decay rates though).



Two things become very obvious, at first that MT-32/16 again falls short in noise performance compared to MT-16/8, second that this is a first example were at least a few correlation estimators of the MT-16/8 Multiple Tau Correlation scheme could be visibly influenced by the "Triangular Averaging Distortion" within still reasonable total experiment durations (here 1000 s). This gives at least a motivation for the correction schemes developed later in this text.

In conclusion, a MT-32/16 or MT-64/32 Multiple Tau implementation obviously does not offer "resolution increase" compared to the MT-16/8 scheme. Increased is solely the number of correlation estimators, however at the cost of decreased statistical accuracy per estimator. While this effect compensates for the photon noise contribution due to its truly random nature, it can not for the covariant signal noise contribution. For this, an increased number of correlation estimators with higher average Std.Dev. does not have any advantage. As was shown above, however, even for rather low count rates presumed, the photon noise is quickly reduced by either Multiple Tau scheme implementation along the lag time axis and practically photon noise never is the dominant noise source anyway.

An exception form this would be the measurement of very fast fluctuations at extremely small count rates (say Γ > 100/ms at a count rate << 5 kcps), as it could appear for rotational diffusion measurements in DLS or FCS or DWS transmission measurements. In these cases, all Multiple Tau implementations would give about the same overall statistical accuracy.

Yet again, even in these cases there would not be any "resolution increase" due to the use of a higher number of linear channels per lag time interval.

5.0 <u>Removing "Triangular Averaging Distortions" from</u> <u>Correlation Functions</u>

As was shown in the above sections, "Triangular Averaging Distortions" seldom have a significant influence on Multiple Tau Correlation correlation functions, even for MT-16/8 implementation, but are normally covered by the much larger estimator noise of such a correlation function even for longer to very long measurement times.

While this already should rule out the use of MT-32/16 or MT-64/32 implementations of Multiple Tau Correlation by itself, simply because both show decrease in overall statistical accuracy of the resulting correlation functions compared to the classical MT-16/8 scheme, the idea of using "more channels per sampling time block" should be fully dropped once knowing that "Triangular Averaging Distortions" can always be removed from the correlation function by several methods.

Method 1: Numerical Correction via the Second Derivative

Let's presume as correlation function any function with an existing Taylor-series expansion in the lag time t around t = 0 (for convenience, any other point would do as good), thus :

$$F(t) = F(0) + F'(0)t + F''(0)\frac{t^2}{2!} + F'''(0)\frac{t^3}{3!}\dots$$
(5.1)

Computing the time averaged correlation function of this function at any point t then leads to

$$g_{\mu}(k) = \frac{1}{\tau^{2}} \int_{-\tau}^{\tau} (F(0) + F'(0) t + F''(0) \frac{t^{2}}{2!} + F'''(0) \frac{t^{3}}{3!} \dots) (\tau - |t|) dt$$

$$= \frac{1}{\tau^{2}} \int_{-\tau}^{\tau} (F(0) (\tau - |t|) dt + \int_{-\tau}^{\tau} F'(0) t (\tau - |t|) dt + \int_{-\tau}^{\tau} F''(0) \frac{t^{2}}{2!} (\tau - |t|) dt \dots$$

$$= \frac{1}{\tau^{2}} (\tau^{2} F(0) + \tau^{3} F'(0) + \frac{7}{12} \tau^{4} F''(0) + R^{6})$$

$$= F(0) + \tau F'(0) + \frac{1}{2} \tau^{2} F''(0) + \frac{1}{12} \tau^{2} F''(0) \dots + O(\tau^{4} F^{(4)}(0)$$

$$\sim F(0) [1 + \frac{1}{12} \tau^{2} F''(0)]$$
(5.2)

where all terms of higher order in τ than second order were dropped. This is an interesting result, because it shows that the effects of temporal averaging can be (practically) fully described by the second derivative of the original correlation function for all decaying correlation functions (see comments on undamped cosine functions in 2.0). In addition, it gives an elegant insight, why for a single exponential correlation function the pre-factor describing the "Triangular Averaging Error" remains fully constant, whereas it does not for a hyperbolic correlation function, for example: while in the case of a single exponential, all even derivatives (uneven derivatives as well, just for the sake of correctness) are exactly this exponential function scaled only by the inner derivatives, this does not hold true for a hyperbolic function obviously. Notably, an undamped cosine function should show constant pre-factor behaviour as well, because all even derivatives are again cosine functions (however, in the $(\tau \omega) \leq 1$ limit only ! For larger $\tau \omega$ the higher than second order terms in 5.2 must no longer be dropped).

As a matter of fact, computing the second derivate of a given correlation function analytically is usually less time-consuming than performing the (straightforward, but lengthy) integration over the triangular weight and in addition it can easily be performed on the measured correlation function as well, because a simple (though rather precise) numerical approximation to the second derivate exists:

$$g_2''(k\tau) \sim \frac{g_2((k-1)\tau) - 2 \cdot g_2(k\tau) + g_2((k+1)\tau)}{\tau^2}$$
(5.3)

which can directly be applied using the correlation data estimators themselves, at least as long as they are already precise enough; in this case, the "Triangular Averaging Error/Distortion" can be computed as

$$\Delta_{Triang}(k\tau) = g_2(k\tau) - g_\mu(k) \sim \frac{g_\mu(k-1) - 2 \cdot g_\mu(k) + g_\mu(k+1)}{12} + \varepsilon$$
(5.4)

where the error term mainly depends on the so far reached accuracy of the approximation data $g_{\mu}(k)$ (note : the error term of using the numerical representation of the second derivative was dropped – it can be shown to be much too small to contribute significantly for the purposes outlined here. Still, the error term shown above does indeed depend additionally on the fact that $g_{\mu}(k)$ do have the "Triangular Averaging Errors" added – however, as was shown above, the noise level usually is significantly larger. If the data is extremely precise, additional recursions should be used to recomputed the "Triangular Averaging Errors" from the values $g_{\mu}(k)$ - $\Delta_{Triang}(k\tau)$ etc.) and can be shown to scale with

$$\varepsilon \sim \frac{1}{12} \cdot \sqrt{\left(\delta g_{\mu}(k-1)\right)^{2} + \left(2\,\delta g\mu_{2}(k)\right)^{2} + \left(\delta g_{\mu}(k+1)\right)^{2}} \sim \frac{1}{30}\,\delta_{Av_{-}}(g_{2}(k))$$
(5.5)

presuming gaussian distributed noise on the correlation estimators, which holds true for the case of photon noise being the dominant noise source. For signal noise being the dominant factor, the average error usually is even smaller due to the strong correlation of the signal noise contribution.

Method 2: Correction via "Absorption into the Fit Model"

While the direct computation of the second derivative, or it's numerical cousin, are very useful for accurate data and/or model function computations, for everyday use an even simpler and yet exactly as precise method exists. For certain the correlation function as such is not the goal of any of the photon correlation experiments, but instead some further data analysis is usually applied to extract the decay rates in one or the other way. All these "fits" are numerical procedures trying to fit a certain number of model functions to the correlation function while trying to minimize certain statistical criteria (least-squares-fitting, which minimizes the sum of the quadratic distances from model function to the measured data points, for example).

The most obvious way to incorporate "Triangular Averaging Distortion" correction into the fitting procedure thus is to, instead of using the pure model functions for the fit, use model functions that already incorporate the additional "Triangular Averaging Distortion".

With the methods shown in this text this is particularly easy to do – the to be expected contribution of the "Triangular Averaging Distortion" for a certain model function at lag time $k\tau_k$ and sampling time τ_k will correctly be computed (with either or approach) and added to the model function accordingly, for example, for a single exponential as model function for the intensity correlation function, this yields

$$g_2^*(k) = A e^{-2\Gamma T_k} \left(1 + \frac{(2\Gamma \tau_k)^2}{12}\right)$$
 (5.6)

were the result of the integration over the triangular weight of the model function for the specific sampling time range $-\tau_k \dots + \tau_k$ at lag time T_k was used. Unfortunately, this can not be linearly adopted to multi-exponential models in DLS, because the fit models in DLS are linear in field correlation function and quadratic in the intensity correlation function (where the triangular averaging takes place as described herein), thus

$$g_{2}(t) = \beta g_{1}(t)^{2} = \beta \left(\sum_{j} c_{j} e^{-\Gamma_{j}t}\right)^{2}$$
 (5.7)

and cross-terms arise in the intensity correlation function (and it's second derivative, of course) requiring the simultaneous knowledge of all $\Gamma_1 \dots \Gamma_N$ and their relative amplitudes. At least the later (if not both) are the scope of the fit though ... thus, this approach always requires non-linear fitting strategies (which is not a problem for multi-exponential force fits – they require non-linear fitting strategies anyway, but is a problem for the well known "grid-based" fitting strategies, such as NNLS, CONTIN and ALV-NonLin as well as most implementations of MEM).

In this case (though not restricted to this case, but a generally valid approach) it is more easy to proceed in a "Three Step Algorithm":

- 1. Perform a (usually least-squares) fit using the desired fit-model and the data $g_u(k)$, with the fit results compute $g_u^*(k)$
- 2. Using $g_{\mu}^{*}(k)$ compute $\Delta_{Triang}(k\tau)$ via (5.4)
- 3. Redo the fit using as data $g_{\mu}(k) \Delta_{Triang}(k\tau)$

Because $g_{\mu}^{*}(k)$, the recomputed correlation data from the fit model is used, there is no need to be careful about the potential problems of the numerical computation of the second order derivative due to the noise level in $g_{\mu}(k)$.

Although the above method is generally valid and convergent for a broad class of correlation functions (in essence, the requirement is that the second derivative exists for all *k* and the function is decaying to zero for $k \rightarrow \infty$. An additional, practical requirement is that the function can be fitted via a least-squares fit the fit model to at least reasonable precision for all *k*), extreme care should be taken for correlation functions composed of undamped cosine functions.

For these, the Multiple Tau Correlation scheme quickly involves sampling times (much) larger than the cosine inverse period and the triangular averaging more and more corresponds to classical aliasing effects and usually any fit not taking these into account will give rather poor results with rather noticeable (and systematic) deviation between $g_{\mu}^{*}(k)$ and $g_{\mu}(k)$. In addition, due to the non-decaying nature of undamped cosine functions, the approximation shown in (5.4) for the "Triangular Averaging Distortion" will only be valid for $\omega t_{s} < 1$ for $g_{\mu}(k) = A \cos(\omega k t_{s})$

In these cases, either the fit must be restricted to data points satisfying the above condition, or the closed form result of the integration of the cosine functions with triangular weighting must be used (2.10). In both cases the convergence of the method should be carefully inspected.

It should, nevertheless, be stressed that for pure cosine correlation functions the use of a constant sampling time correlation scheme with a reasonably large number of correlation channels usually is the better choice, though the MT-16/8 implementation was succesfully applied to such problem classes correctly fitting the data even for $\omega t_s >> 10$ using the closed form equation along with non-linear fitting methods.

To prove the schemes extreme efficiency, data taken over 12 hours (to be able to perfectly visualize the "Triangular Averaging Distortions") from a special random test generator with pure single exponential correlation function expectation (proprietary development of ALV, implemented as test generator in the most modern ALV-7004 and ALV-7004/FAST correlators) was analyzed with and without the correction via "Absorbing into the Fit Model". The test data generated has an expectation for the

decay rate of 4,817/ms +/- 0.023/ms, an expectation for β of 1.0 +/ 0.001 and an expectation for the baseline of <10⁻⁸. The average output count rate was 312,5 kcps.



After <u>~12 hours</u> of measurement time using an ALV-7004 Multiple Tau Digital Correlator, the average noise level can be shown to be about $\delta \leq 10^{-4}$ for the complete lag time range shown. Clearly, for the uncorrected fit version, thus fitting the pure model functions only, the triangular averaging distortions become easily visible within the residuals $(g_{\mu}(k) - g_{\mu}^{*}(k))$.

Fitting the corrected model function incorporating the to be expected distortions removes this problem completely and yields smooth residuals (they are smooth because they are correlated, as we outlined above) in this lag time regime, all well within the average noise level – the "Triangular Averaging Distortions" are thus fully removed.

In both cases a non-linear least-squares fit approach was used force fitting

$$g^{\text{uncorrected}}(k) = A e^{-2\Gamma T_k} + B$$
(5.8)

resp.

$$g^{corrected}(k) = A e^{-2\Gamma T_k} \left(1 + \frac{(2\Gamma \tau_k)^2}{12}\right) + B$$
(5.9)

with A, Γ and B being optimized in the fit. The numerical results of the fit are given in the below table, again with the values to be expected from the test generator data

	Uncorrected Fit Model	Corrected Fit Model	Test Generator's Expectations
A	1.0	1.0	1.0 +/- 0.001
G[1/ms]	4.8125	4.8169	4.817 +/- 0.023
В	4.437 10 ⁻⁵	0	0 +/- 10 ⁻⁸
LSQ [arb.]	1.0704 10-4	5.1564 10 ⁻⁵	

Test Generator Data, Fit Results

Although the uncorrected version of the fit shows significantly worse residuals then the corrected version, the results are much less different than could be expected. The primary parameter of interest, namely the decay rate differs not more than just 0.1% from the expected result. Obviously, this minor change in the decay rate already minimizes the effect of the "Triangular Averaging Distortions" in the least-squares sense. In fact, we can equate:

$$\varepsilon = \sum_{k} \left(e^{-k\tau 2\Gamma^*} - e^{-k\tau 2\Gamma} \left(1 + \frac{(2\Gamma\tau)^2}{12}\right) \right)^2 \to \text{minimum}$$
(5.10)

The below plot shows this graphically for different Γ/Γ^* relations. Obviously, the minimum for noiseless data can be found at $\Gamma/\Gamma^* \sim 1.0007$, which is in very good agreement with the results obtained from the actual fit of the data of ~1.00094. Interestingly, the additional B parameter, if taken into account as well, does not change the minimum position of Γ/Γ^* significantly, although it does indeed change the absolute value of the LSQ-sum.



The same data was analysed using the "Three Step Algorithm" with a single iteration. As was expected, it gave identical results compared to the analytical approach, see below graph of the residuals :



In summary, the effects of the "Triangular Averaging Distortions" on the results of a single exponential fit to the correlation data are surprisingly small and unless the data is of extreme accuracy, the results inherent inaccuracy due to the correlation estimator noise level is by magnitudes larger. The explanation of this effect lies in the strict systematic and non-zero contribution on just a few points of the correlation function due to the "Triangular Averaging Distortion".

Method 3: Modified LSQ-Sum Correction

In the case of non-linear fitting, it is not necessarily required to restrict the fit to a "pure" least-squares approach, thus to compute the least-squares sum using the corrected fit model (recall 5.10)

$$\varepsilon = \sum_{k} \left(g_{\mu}(k) - g_{\mu}^{* \operatorname{corr}}(k) \right)^{2} \to \min.$$
(5.11)

Instead, the "Triangular Averaging Distortion" can as well be incorporated in the process of computing a "corrected" LSQ-sum, thus

$$\varepsilon = \sum_{k} \left(g_{\mu}(k) - g_{\mu}^{*}(k) - \frac{1}{12} [g_{\mu}^{*}(k-1) - 2g_{\mu}^{*}(k) + g_{\mu}^{*}(k+1)] \right)^{2} \to \text{min.}$$
 (5.12)

using again the numerical approach of computing the second derivative of the model function. Indeed, this method is the preferred method for all non-linear fits implemented in the ALV-Correlator Software for WINDOWS® package, showing exactly the same accuracy, as Method 1 and Method 2 described above.

While such an approach could principally be implemented for the case of grid based "standard" LSQ-methods, such as CONTIN, ALV-NonLin or MEM as well, the number of grid points required would be three-fold compared to the uncorrected fit model (because the fit matrix would have to be extended per row for the resulting model data of $g_{\mu}^{*}(k-1)$ and $g_{\mu}^{*}(k+1)$, requiring additional 2N columns in the fit matrix per row) and such approach would suffer from significant performance penalties compared to the "Three Step Algorithm" described above, without yielding to any better results.

Methods 1...3 outlined above show, that removing "Triangular Averaging Distortions" from a correlation function is simple and highly efficient, though, keeping in mind the effective influence of these on the fit results, not desperately required in most cases.

Nevertheless, the ALV-Correlator Software for WINDOWS® package uses either the "Modified LSQ-sum Correction" for all non-linear fits implemented or the "Three Step Algorithm" for the grid based fitting strategies implemented per default to safely correct for potential "Triangular Averaging Distortions". No such correction is applied for the "Cumulant Fit", were this would not pay off anyway.

Conclusion

One particular effect of sampling data from a time continuous process for correlation function computation is that sampling time dependent scale transformation and/or distortions due to "Triangular Averaging Effects" must be expected.

While not problematic for a small enough and constant sampling time correlation function, the use of several, different sampling times in parallel and along the lag time axis, as is used in particular in Multiple Tau Correlation schemes, always makes these "Triangular Averaging Effects" a systematic distortion of the correlation function – the so called "Triangular Averaging Distortions".

The absolute magnitude of these distortions stays rather small, no matter of the specific implementation of Multiple Tau used (MT-16/8, MT-32/16 or MT-64/32), for most experimental conditions even too small to become visible – they will be deeply buried in the photon and signal noise.

Still, in some cases it is worth considering a full correction of these distortions. This can be performed at very high accuracy, in some cases even analytically, by the methods outlined herein.

In none of these cases it seems useful to instead increase the number of correlation channels per sampling time from the 16/8 scheme used in the classical Multiple Tau Correlation scheme to decrease the total magnitude of the "Triangular Averaging Distortions", because doing so has significant impact on the overall statistical accuracy achievable for the correlation functions computed. Correlation functions computed using MT-32/16 or even MT-64/32 sampling time implementations of Multiple Tau show less statistical accuracy within the same measurement time and thus reduced, and by no means higher, "resolution" compared to the 16/8 channel scheme.

References

[ref 1]	K. Schätzel: "Noise on Photon Correlation Data: I.", Quantum Optics 2, 287-305 (1990)
[ref 2]	W. Brown, editor. "Dynamic Light Scattering : The Method and some Application", Clarendon Press, Oxford, (1993), therein the contributions of K. Schätzel and R. Peters
[ref 3]	ALV-Application Notes: "Seeing each other" – distortions at small lag times on correlation functions obtained using pseudo-cross correlation detection with APD-based single photon detectors, R. Peters for ALV-GmbH

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