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Experimental analysis of the noise in photon-correlation and photon-structure functions

G. F. Lorusso, V. Capozzi, and A. Minafra
Dipartimento di Fisica, Università di Bari, Via Amendola 173, I-70126 Bari, Italy
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We present an experimental analysis of the variance of photon-correlation and photon-structure functions obtained from dynamic light-scattering measurements. The results are compared with theoretical predictions. The noise evaluation of the statistical estimators has been obtained by using a fluctuation analyzer, specifically built for this purpose, which uses linearly spaced sampling time. The noise dependences on the experimental bounds (mean count rate and sampling time) are investigated. The predicted behavior of the estimator variances has been confirmed. However, the measured accuracies of correlation and structure functions show a discrepancy between their analytical expressions and the experimental results in the high-intensity limit of the scattered light. The results confirm that the structure function has better noise performance for relatively high mean count rates. This strongly suggests a wider use of this statistical estimator in measurements requiring a good accuracy, such as in the analysis of polydispersed microparticle size distribution, in the case of uniform sampling.

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I. INTRODUCTION

The analysis and the interpretation of the fluctuations of light field has been a widespread field of research in recent years. The most used experimental technique is based on the computation of the intensity autocorrelation function $g(t,T)$, i.e., the autocorrelation function of the photon counts $n(t,T)$ detected at time t during the sampling time T [1]. This is accomplished by using a fast real-time digital autocorrelation apparatus. An alternative method has been proposed by Oliver and Pike [2]: It is based on the evaluation of photon-structure function $d(t,T)$, widely used in fields like turbulence and atmospheric optics. However, most commercial apparatus are not capable of evaluating the structure function.

The influence of the noise of the statistical estimator used for the interpretation of dynamic light-scattering (DLS) measurements on the information available from experiments has been discussed by several authors [3,4]. In fact, to obtain the size distribution of suspended microparticles in polydispersity analysis, it is necessary to find the solution of a Fredholm integral equation of the first kind [5]. In general the solution of this equation is an "ill-posed" problem, and it leads to difficulties because of the existence of an infinite number of oscillating solutions satisfying the investigated integral within the experimental errors. Hence, the knowledge of the behavior of noise of the used statistical estimator permits an improvement of the accuracy of DLS measurements.

In this paper the variance of the statistical estimators $g(t,T)$ and $d(t,T)$ are experimentally evaluated in DLS measurements with a single time constant (i.e., monodisperse particle suspension) without a reference beam (i.e., homodyne detection) by using uniform sampling technique. The dependence of the variances on mean count rate and sampling time are investigated; the results are compared with the theoretical predictions.

II. ANALYTICAL METHODS

The intensity correlation function is theoretically defined by the following relation:

$$g(k,T)=\langle n(0,T)n(kT,T)\rangle \tag{1}$$

where $n(t,T)$ is the photon counts detected at time t during the sampling time $T(t=kT;\ k=1,2,3,\ldots)$. This definition is valid for stochastic processes $n(t,T)$ with a stable mean value. Hence the correlation function has no meaning in processes showing time drifts.

The photon-structure function is defined for stochastic processes with stationary increments [6]. This assumption is weaker than the stability of the mean value. The photon-structure function is expressed as follows:

$$d(k)=\langle [n(0,T)-n(kT,T)]^2\rangle\,. \tag{2}$$

Since the structure function is a relative measurement, it is less sensitive to low-frequency noise. This allows an improvement of the signal-to-noise ratio for short time lags at relatively high mean count rates. On the contrary, the correlation function is less sensitive to high-frequency noise, and therefore it should be used when experimental conditions allow only weak signals.

To compare the variances of the statistical estimators $g(k,T)$ and $d(k,T)$ it is necessary to introduce the modified estimators, which are defined as

$$G(k,T)=g(k,T)-\bar{n}^2\,, \tag{3}$$

$$D(k,T)=d(k,T)-2\bar{n}\,, \tag{4}$$

where $\bar{n}$ is the mean count rates of the stochastic process. The normalized modified estimators $\hat{G}(k,T)$ and $\hat{D}(k,T)$ can be obtained by dividing Eq. (3) by $\bar{n}^2$ and Eq. (4) by $2\bar{n}^2$.

For the direct detection of Gaussian-Lorentian light the

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theoretical estimator variance for $G(k,T)$ is given by [6]

$$\sigma^2(G)=\frac{\bar{n}^4}{N}[\coth 2\gamma+2e^{-2\gamma k}\coth\gamma+e^{-4\gamma k}(6k+3\coth 2\gamma)]+\frac{\bar{n}^3}{N}(2+4e^{-2\gamma k}+6e^{-4\gamma k})+\frac{\bar{n}^2}{N}(1+e^{-2\gamma k})\,. \tag{5}$$

Similarly, the analytical form for the estimator variance of $D(k,T)$ is [6]

$$\sigma^2(D)=\frac{4\bar{n}^4}{N}[4\coth\gamma+5\coth 2\gamma-e^{-2\gamma k}(16k+4\coth\gamma+8\coth 2\gamma)+e^{-4\gamma k}(6k+3\coth 2\gamma)]$$
$$+\frac{4\bar{n}^3}{N}[14+4\coth\gamma-e^{-2\gamma k}(20+4k+4\coth\gamma)+6e^{-4\gamma k}]+\frac{4\bar{n}^2}{N}(5+e^{-2\gamma k})\,, \tag{6}$$

where γ is the ratio between the sampling time T and the coherence time T_c, and N is the number of recorded counts.

The behavior of these two analytical expressions is shown in Fig. 1. For very low mean count rates ($10^{-3}\leq\bar{n}\leq 10^{-1}$) the noise of the correlation function (solid line) is always lower than the noise of the structure function (dashed line), while for relatively high mean count rates ($\bar{n}\geq 1$) the structure function has better noise performance than correlation function at short time lags. Moreover, Fig. 1 indicates that the analytical expressions [Eqs. (5) and (6)] do not show significant variations for $\bar{n}$ greater than 10 counts per channel.

III. RESULTS AND DISCUSSION

The experimental evaluation of the variance of the correlation and structure functions has been obtained by means of a purposely built apparatus [7]. This apparatus is a PC-based fluctuation analyzer, composed of an acquisition board and a dedicated elaboration software. The acquisition board counts the scattered photons detected in the chosen sampling time and stores the results in the PC memory by using direct memory access (DMA). The acquisition board uses linearly spaced sampling times. The software permits the calculation of the structure function as well as the correlation function from the collected data. Moreover, it permits the evaluation of the noise for each channel of the experimentally measured G and D.

The noise in $\hat{G}$ and $\hat{D}$ has been evaluated by means of DLS measurements of light scattered from polystyrene standard latex spheres (215 nm nominal size, 2.6 nm standard deviation) suspended in distilled water, filtered by a 200-nm sieve. A 4-mW He-Ne laser source has been used. The scattered light has been detected at 30° from the incident direction by a photomultiplier tube cooled at $-20\,°$C, which has a dark mean count rate of 3.5 counts/s. This very low dark level is a good specification for the noise evaluation. The experimental variances have been obtained by comparing 800 photon-correlation or photon-structure functions, each one computed from 1.2×10^4 recorded counts.

The experimental results and theoretical predictions on variance of $\hat{G}$ and $\hat{D}$ obtained for different values of γ are compared in Fig. 2. The theoretical behavior of $\sigma^2(\hat{G})$ and of $\sigma^2(\hat{D})$ has been confirmed by our measurements, although the experimental variances show a dependence of the crossover region on the chosen sampling time. In fact, by increasing $\bar{n}$, the crossover shifts toward longer values of time lags.

The intensity dependence of the noise in photon-correlation and photon-structure functions have been investigated by evaluating experimental variances for different mean count rates, as shown in Fig. 3. The analytical expressions of the variances of $\hat{G}$ and $\hat{D}$ result independent of the mean count rate in the investigated intensity range, in contrast with the experimental results. Moreover, the experimental crossover region always occurs at longer time lags than theoretical predictions.

In conclusion, our experimental results confirm that, in the investigated intensity range, the variance of the photon-structure function is much smaller than the variance of the photon-correlation function for short time lags (about $t\leq 15$ ms). Our results are restricted to the case of linearly spaced delays and sampling times. We remark that the main amount of information about the particle

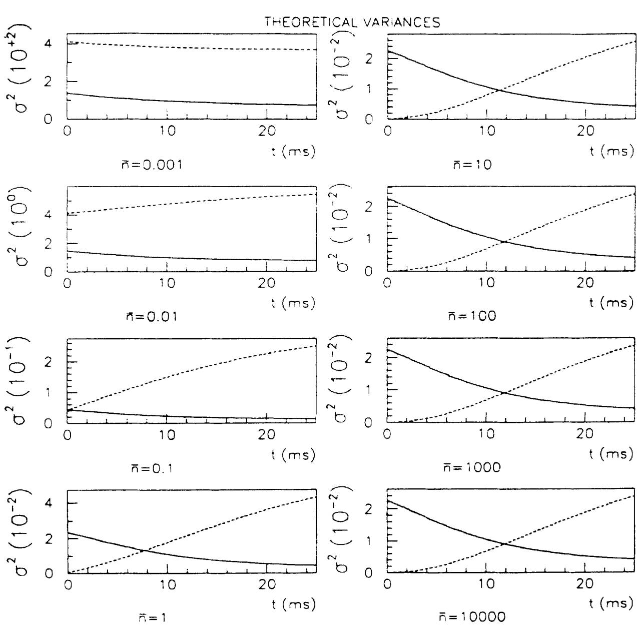

FIG. 1. Theoretical predictions of the variances of photon-structure function (dashed line) and photon-correlation function (solid line) for different mean count rates $\bar{n}$. The ratio between the sampling time and the coherence time is $\gamma=1.22\times 10^{-2}$. The analytical expressions do not show significant variations when $\bar{n}$ is greater than 10 counts per channel.

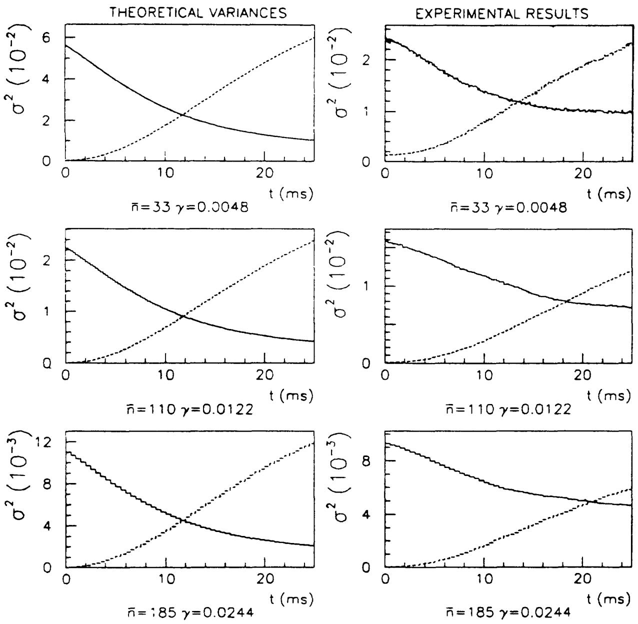

FIG. 2. Comparison between theoretical predictions and experimental results of the variances of normalized photon-structure function (dashed line) and photon-correlation functions (solid line) for different sampling times T. γ and $\bar{n}$ have the same meanings as in Fig. 1.

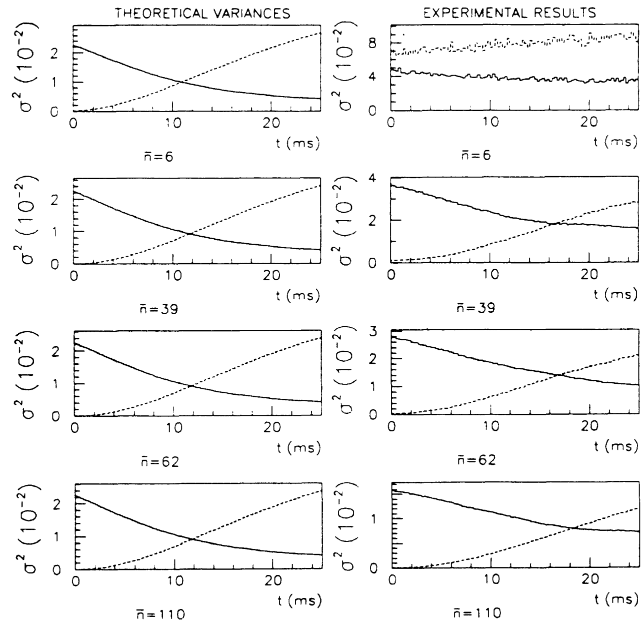

FIG. 3. Comparison between theoretical predictions and experimental results of variances of normalized photon-structure function (dashed line) and photon-correlation function (solid line) for different $\bar{n}$. γ and $\bar{n}$ have the same meanings as in Fig. 1. The ratio between the sampling time and the coherence time is $\gamma=1.22\times10^{-2}$.

size distribution photon is just in the short-time-lag region of the structure and the correlation functions. So, the measurement of $\hat{D}$, rather than $\hat{G}$, strongly improves the accuracy of the methods of data analysis needed to solve the integral equation of Fredholm of the first kind.

The predicted general behavior of the variances [Eqs. (5) and (6)] has been experimentally confirmed. The observed discrepancies between theoretical and experimental values suggest the need for an improvement of the analytical model for what regards the dependence of noise on the mean count rates. Eventually, the theory should be refined in its variance crossover predictions.

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