Experimental photon statistics of multiscattered light

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The concentration dependence of the statistical properties of light scattered by a monodisperse sphere suspension has been studied. Theoretical predictions for photon-counting distributions, obtained by supposing the scattered field is a superposition of coherent signal S and noise excitation N, agree well with the experimental results, thus providing an alternative method to obtain quantitative information on the influence of multiple scattering on measurements. Moreover, the autocorrelation function is deformed from an exponential law to a stretched-exponential law, as expected, and the experimental results show that the parameter d_w, quantifying the exponential stretching, and the ratio S/N exhibit a similar dependence on the level of multiple scattering.

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It has been recently shown [1] that when the effective disorder increases, non-Gaussian statistics characterizes the intensity fluctuations of a propagating wave in a random medium. When the system under study contains a large number of independent scatterers, the amplitudes of the scattered field are Gaussian, and in the limit of short counting times, the intensity I is described by an exponential probability density. In this case, the probabilities $P(n,T)$ of counting n photoelectrons in a time T are given by the Bose-Einstein distribution.

Such a Gaussian model was widely used throughout the earlier calculations in photon correlation spectroscopy [2–4]. In fact, since light-scattering experiments are performed with a large number of statistically independent scattering centers, we aspect to detect quasithermal light, i.e., light with a Gaussian amplitude. However, several assumptions, not always achieved in practice, are necessary: uniformly illuminated scattering volume, total spatial coherence at the detector, and sampling intervals that are short compared to the fluctuation time of the scattered intensity. In particular, real experiments generally involve significant spatial integration, because a large detector aperture is highly desirable in order to improve statistical accuracy.

Moreover, $P(I)$ will no longer be described by negative exponential statistics as result of wave confinement, which leads to an enhancement of the probability of waves returning to a point in the medium, or as a result of increased strength of local scattering. Such a deviation of $P(I)$ from exponential statistics has been observed either in the case of strongly scattering samples, or in weakly scattering samples as the sample length increases (for constant cross section) [1,5]. This anomalous behavior of $P(I)$ has been considered not simply as the result of wave confinement, but also as associated to localization effects [6].

Another interesting effect of highly concentrated systems consists in the speed up of the decay of the intensity-intensity autocorrelation function $g^{(2)}(t)$ caused by elastic multiple scattering. Since doubly scattered light has a fixed phase relation with respect to single scattered light, the doubly scattered contribution may increase or decrease the optical amplitude at the detector depending on the relative phase [7]. Averagely, it leads to a small increase of amplitude and to enhanced autocorrelation functions at shorter delay times. Thus, dynamic light scattering on highly concentrated suspensions, where multiple scattering is inevitable, provides distorted correlation functions.

This effect, which does not permit the evaluation of parameters with standard methods, can be avoided by using a two wavelength setup and by measuring the cross correlation function [8]. However, it has been recently demonstrated [9–11] that the analysis of the distortion of $g^{(2)}(t)$ caused by multiple scattering gives important information about the degree of correlation between interacting scatterers. In particular, the transport of light through opaque suspensions is treated as a diffusive process in diffusing-wave spectroscopy (DWS) [9], and consequently the intensity autocorrelation function shows an exponential decay with the square root of time, thus permitting the evaluation of the dynamical properties of the suspended particles. This indicates that, by increasing the concentration of the suspension, multiple scattering distorts $g^{(2)}(t)$ from the simple exponential form of the single-scattering limit [12] to a stretched exponential form, which reaches the $\exp(-\sqrt{t})$ limit for high concentrations in the case of DWS.

In this paper, we analyze the influence of multiple scattering on the photon-counting distribution function $P(n,T)$ and of the intensity-intensity autocorrelation function $g^{(2)}(t)$ of light scattered from polystyrene latex sphere suspensions with different concentrations at room temperature. The scattered light was observed at $\theta=30°$ and detected by a photomultiplier tube cooled at $T=-20\,°\mathrm{C}$, with a dark current up to 10 counts/s. The suspension was placed in a cuvette having 1.5 mm of optical path and 130-μm walls. The light source (1-mW He-Ne laser) was focused on the samples by means of a 150-mm focal lens. A spot of about 100-μm diameter was obtained. The scattered light was collected just by using spatial filtering. Such an experimental geometry, basically different from that used in DWS [13], has been found to be profitable to follow the evolution of the statistical properties of light from the single- to the multiple-scattering case. The experimental evaluation of $P(n,T)$ and $g^{(2)}(t)$ is obtained by means of a PC based correlator [14] by using a single-beam scattering setup. A relatively high mean count rate, about 6×10^5 counts/s, is chosen in order to avoid the zero spike caused by photon shot noise, which could lead to misnormalization of $g^{(2)}(t)$. The same mean count rate was used in all measurements by controlling the light intensity by means of neutral density filters.

A light beam with a finite coherence time can be considered as a single mode excited to a noisy state [15]. As stated before, the amplitude distribution can be considered to be Gaussian. The probability distribution for an instantaneous measurement of the cycle-averaged intensity results in a negative exponential distribution, and the photon-counting probabilities result in a geometrical one, i.e.,

$$P(n,T)=\bar{n}^n/(1+\bar{n})^{n+1} \tag{1}$$

with a mean value $\bar{n}$ and a variance $\langle \Delta n^2\rangle=\bar{n}(1+\bar{n})$. This distribution is also called a Bose-Einstein distribution, since the equivalent-photon distribution shows the same statistical fluctuations of the quantum number around its average as that of a black-body single-mode field around its average photon number.

In the case of a pure coherent state, the density operator P, which gives the complete quantum-statistical specification of the state of the system, is a product of δ functions which constrains the amplitude to have a particular set of values. It has been shown that in this case the photon-counting distribution $P(n,T)$ is a Poissonian one,

$$P(n,T)=\frac{\bar{n}^n}{n!}\exp(-\bar{n}) \tag{2}$$

with $\langle \Delta n^2\rangle=\bar{n}$. In this the case the intensity is independent of time. The fluctuations which occur for a beam of constant intensity are called particle fluctuations. They are due to the discrete nature of the photoelectric process, in which energy can be removed from the light beam only through photons. These particle fluctuations are an intrinsic, irreducible feature of the photon counting experiment. Moreover, we note that the mean-square deviation contains information on the second-order coherence properties of the light.

In the case of superposition of coherent signal and noise excitation in a single mode, it has been shown [15] that the photon-counting distribution function is

$$P(n,T)=\frac{N^n}{(1+N)^{n+1}}L_m\left[-\frac{S}{N(1+N)}\right]\exp\left[-\frac{S}{1+N}\right], \tag{3}$$

where S and N are the number of quanta which are contained in the signal and noise fields measured independently. L_m are the Laguerre polynomials. When the signal strength vanishes this distribution assumes the geometrical form, and in the opposite extreme, when the noise excitation disappears, $P(m,T)$ reduces to the Poisson distribution characteristic of a coherent state. The mean values and the variance associated with this distribution are, respectively,

$$\bar{n}=S+N\ , \tag{4}$$

$$\langle \Delta n^2\rangle=S+N(1+N)+2SN\ . \tag{5}$$

Experimental results for $P(n,T)$ at different packing fractions Φ and theoretical predictions of Eq. (3) are compared in Fig. 1. N and S are computed by means of Eqs. (4) and (5), by using the experimental evaluated mean values and variances. It is worth noting that no fitting parameter is used. The packing fraction Φ is defined as the ratio between ρV_D, where V_D is the volume of the particle and ρ the number density, and Φ_C, the maximum value of Φ, the hexagonal-solid, close-packed value ($\Phi_C\simeq0.74048$). Figure 1 shows clearly the excellent agreement between the photon-counting distribution for mixed light (3) and the experimental results. The effect of multiple scattering on the photon-counting distribution is evident: $P(n,T)$ results are more and more similar to the Poisson distribution of coherent states as the Φ is increased, thus indicating that the number of quanta in the coherent field is enhanced and that the scattered intensity became progressively more independent of time.

Although the general validity of Eq. (3) is well known and any further confirmation is certainly not of special interest, the experimental results exhibit a quite interesting feature. In fact, the quantitative evaluation of the parameters characterizing $P(n,T)$, i.e., S and N, can be obtained just by measuring the mean value and the standard deviation of the scattered light, thus providing a simple experimental method to quantify the influence of the multiple scattering on the measure.

The experimental results obtained for $g^{(2)}(t)$ at different packing fractions Φ are shown in Fig. 2(A). It is evident that the $g^{(2)}(t)$ decay is enhanced by increasing Φ. This effect confirms the fact that the photon flow of

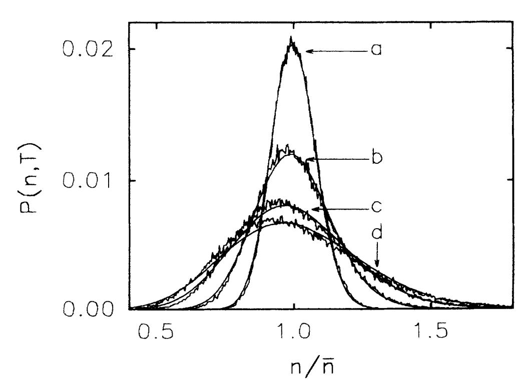

FIG. 1. Concentration dependence of the experimental photon-counting distribution functions $P(n,T)$ and theoretical predictions for mixed light of coherent and noise states. No fitting parameter is used. The packing fractions are (a) $\Phi=1.07\times10^{-2}$, (b) $\Phi=0.53\times10^{-2}$, (c) $\Phi=0.26\times10^{-2}$, (d) $\Phi=0.13\times10^{-2}$. The particle diameter is 303 nm and the sampling time is $T=500\ \mu$s.

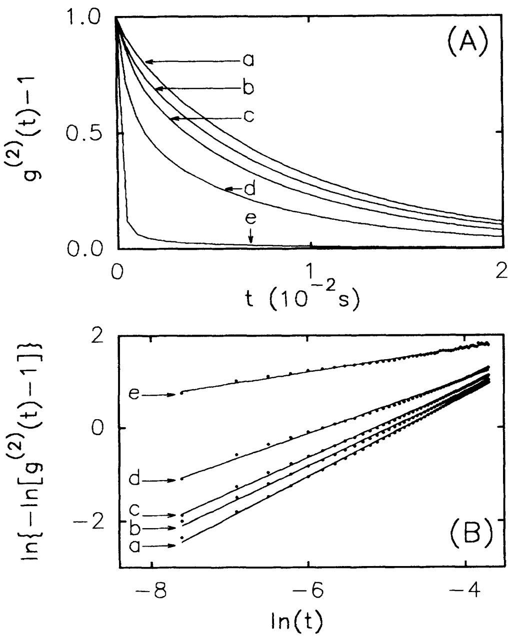

FIG. 2. (A) Particle concentration dependence of the experimental intensity-intensity autocorrelation function $g^{(2)}(t)$. (B) Log-log plot of the data (dots) shown in (A) fitted to Eq. (6). The packing fractions are (a) $\Phi=0.06\times10^{-2}$, (b) $\Phi=0.13\times10^{-2}$, (c) $\Phi=0.26\times10^{-2}$, (d) $\Phi=0.53\times10^{-2}$, (e) $\Phi=1.06\times10^{-2}$. The particle diameter is 215 nm and the sampling time is $T=400$ μs.

the scattered light becomes progressively time independent, as it is argued by the above analysis on photon-counting distribution functions. Moreover, in such concentrated systems, $\ln[g^{(2)}(t)-1]$ is no longer a linear function of t, as in the case of dilute suspensions. Figure 2(B) shows the log-log plot of $|\ln[g^{(2)}(t)-1]|$ versus time. The fact that all the data for different concentrations result to be straight lines in this log-log plot, clearly indicates the existence of a power-law dependence of $|\ln[g^{(2)}(t)-1]|$ on time t, i.e., multiple-scattering distorts the correlation functions $g^{(2)}(t)$ from single exponential form to stretched-exponential law, as expected.

In order to take into account the effect of the multiple scattering on $g^{(2)}(t)$, an anomalous diffusion exponent d_w is introduced to quantify the level of stretching of the exponential law

$$\ln[g^{(2)}(t)-1]\propto t^{2/d_w} . \tag{6}$$

Obviously, in the case of dilute suspension, the exponent d_w is about 2, as predicted by the single-scattering theory [12], while for highly concentrated systems $g^{(2)}(t)$ will be a stretched exponential with $d_w>2$.

The anomalous diffusion exponent d_w, evaluated by fitting $g^{(2)}(t)$ with Eq. (6), is compared in Fig. 3 with the ratio S/N, obtained by means of Eqs. (4) and (5). Both the parameters are increased by multiple scattering, and

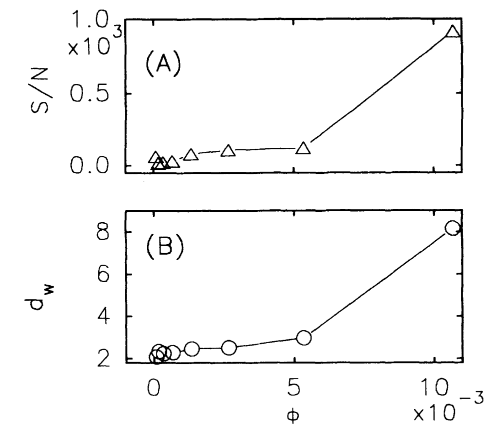

FIG. 3. Particle concentration dependence of (A) the ratio S/N between the number of quanta in coherent and noise state, (B) and of the anomalous diffusion exponent d_w. The particle diameter is 215 nm and the sampling time is $T=400$ μs.

tend to be constant values in the single-scattering limit, as expected. Moreover, S/N and d_w show a similar dependence on Φ, thus suggesting that the degree of multiple scattering involved in the experimental measure influences in a quite analogous way the level of stretching of the correlation function and photon-counting distributions. However, we remark, that, in our knowledge, no analytical relationship between the level of exponential stretching d_w and the ratio S/N has been developed, up to now.

In conclusion, we have analyzed the effect of the multiple scattering on the photon-counting probability distribution function $P(n,T)$ and on the intensity-intensity autocorrelation function $g^{(2)}(t)$. We have shown that $P(n,T)$ can be accurately described by assuming that the scattered light is a superposition of coherent field, containing S quanta, and a noise excitation field of N quanta. The ratio S/N has been found directly related to the influence of the multiple scattering on the measure. S and N can be easily evaluated by measuring the mean and standard deviation of the scattered light, thus providing a quantitative measure of the degree of multiple scattering involved in the measure. By analyzing the level of stretching of the intensity autocorrelation function caused by multiple scattering, it has been observed that the parameter d_w that quantify the exponential stretching and the ratio S/N are strictly related.

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