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Photoelectron statistics and autocorrelation of Rayleigh scattering

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The statistics and autocorrelation function of the light scattered from polystyrene latex have been studied with a light-detecting system on-line with a small digital computer. The distributions of photoelectric counts have been measured during count intervals from 5×10^{-5} to 5×10^{-3} sec. The results were well accounted for by the approximate theory of Mandel. The factorial moments have been measured to sixth order and the results were in excellent agreement with theoretical calculations. For the first time, we have directly measured the autocorrelation function without using a clipping or scaling method. With this direct method, the comparison between the results of the composite measurements and the theoretical predictions can be quantitatively discussed.

I. INTRODUCTION

The statistics and the autocorrelation of photons have been theoretically studied by many authors.[1-3] Although the statistics of the laser light[4] and pseudothermal light[5] have been intensively investigated, those of a Rayleigh scattering have not been measured. While the autocorrelation function has been obtained for various systems by using the clipping or scaling method, the direct measurement of it has not been given.[6,7]

Here, we report the composite experiments of the statistics and the autocorrelation for a Rayleigh scattering by a new and precise method using an on-line computer system and we present the comparison of the results with the theoretical predictions.

When the intensity-stabilized and narrow-band light is incident on the medium of which refractive index is undergoing a thermal fluctuation, the scattered light reflects the statistics of the fluctuation. The distribution of the counted number of photoelectric pulses from a photocathode which is illuminated by the scattered light is given by a Bose-Einstein distribution assuming a count duration much shorter than the correlation time of the thermal fluctuation. On the contrary, when the duration becomes much longer than the correlation time, the correlation of the scattered light will be smoothed out and the observed photoelectric distribution is expected to have a Poisson distribution. For a count interval between the above extreme cases, the distribution becomes a function of the count interval and the correlation time of the fluctuation. We made a direct measurement of those photoelectric distributions.

One may more quantitatively grasp the fluctuation by the factorial moments than by the immediate distributions of the counted numbers. We measured the factorial moments up to sixth order for count intervals from 10^{-1} to 10^{2} times the correlation time of the scattered light. With modified software of the counting system, we measured the autocorrelation function of the photoelectric pulses.

It should be noted that although we actually discuss the statistics and the autocorrelation function of the photoelectric pulses, they are intrinsically equivalent to those of the scattered photons.[8] The theoretical treatment of the clipping or scaling method of the measurement of the autocorrelation[9,10] is rather complicated, while that of the direct measurement can be made more straightforwardly.

II. THEORY

The brief summary of the theory relevant to our experiments is presented as follows. Further detailed treatment shall be referred to the excellent review by Mehta.[11]

A. Probability distribution

The fluctuation of the light intensity is usually inferred from the photoelectric measurement. The probability $P(n)$ of detecting n photoelectric pulses in a time interval T is related to the probability density $P(W)$ of the integrated light intensity W as[12]

$$P(n)=\int_0^\infty \frac{(\alpha W)^n}{n!}\,e^{-\alpha W}P(W)\,dW\ , \tag{1}$$

$$W=\int_{t-T/2}^{t+T/2} I(t')dt'\ , \tag{2}$$

where α is the quantum efficiency of the detection and $I(t)$ is the light intensity. In the case of the count interval much shorter than the correlation time of the light fluctuation, T_c, the probability $P(n)$ of a Gaussian light becomes a geometrical distribution,[2]

$$P(n)=\frac{\langle n\rangle^n}{(1+\langle n\rangle)^{1+n}}\ ,\quad T\ll T_c \tag{3}$$

where $\langle n\rangle=\alpha\langle W\rangle$. Here $\langle W\rangle$ and $\langle n\rangle$ are the ensemble averages of the integrated light intensity and the number of photoelectric counts, respectively. On the other hand, when the count interval is much longer than the correlation time, $P(n)$ becomes a Poisson distribution,

$$P(n)=\frac{\langle n\rangle^n e^{-\langle n\rangle}}{n!},\quad T\gg T_c\ . \tag{4}$$

The probability $P(n)$ of the intensity stabilized light is also represented by the Poisson distribution.

When the count interval is comparable with the correlation time of the fluctuation, $P(n)$ is not given in an exact form since no explicit expression is available for $P(W)$. The approximate formula of $P(W)$, however, had been obtained by Mandel[13] for an arbitrary value of T as

$$P(n)=\frac{\Gamma(n+1/F(2))}{n!\,\Gamma(1/F(2))}\times\frac{1}{[1+\langle n\rangle/F(2)]^{1/F(2)}}\ \frac{1}{[1+1/\langle n\rangle F(2)]^n}\ , \tag{5}$$

where $\Gamma(x)$ is a gamma function and $F(2)$ is a normalized factorial moment of second order which will be introduced later.

B. Factorial moment

For the photoelectron distribution, we define the normalized factorial moment of kth order as

$$F(k)\equiv\frac{\langle n^{(k)}\rangle}{\langle n\rangle^k}-1\ . \tag{6}$$

Since the following relation holds between $\langle n^{(k)}\rangle$ and $\langle W^k\rangle$:

$$\langle n^{(k)}\rangle=\alpha^k\langle W^k\rangle\ , \tag{7}$$

the kth factorial moment $F(k)$ is represented by the integrated light intensity as

$$F(k)=\langle W^k\rangle/\langle W\rangle^k-1\ . \tag{8}$$

With the aid of the statistics, $\langle W^n\rangle/\langle W\rangle^n$ is given by the cumulants of W,[14]

$$\frac{\langle W^n\rangle}{\langle W\rangle^n}=\sum n!\prod_{i=1}^{k}\frac{\kappa_{n_i}^{l_i}}{n_i!\,l_i!}\ , \tag{9}$$

where κ_n represents nth normalized cumulant and the summation includes all the possible combinations of positive integers which satisfy the condition

$$\sum_{i=1}^{k} l_i n_i=n\ .$$

The nth normalized cumulant κ_n is calculated from the first-order correlation function $C^{(1)}(\tau)$ of the Gaussian light field by the next equation[13]:

$$\kappa_n=\frac{(n-1)!}{T^n C^{(1)}(0)^n}\int_0^T dt_1\cdots dt_{n-1}\times\int_0^T dt_n\, C^{(1)}(t_1-t_2)C^{(1)}(t_2-t_3)\cdots C^{(1)}(t_n-t_1)\ , \tag{10}$$

where

$$C^{(1)}(\tau)\equiv\langle E^{(-)}(t)E^{(+)}(t+\tau)\rangle\ , \tag{11a}$$

$$C^{(1)}(0)\equiv\langle E^{(-)}(t)E^{(+)}(t)\rangle=\langle I(t)\rangle\ , \tag{11b}$$

and $E^{(+)}$ and $E^{(-)}$ are the positive and the negative frequency parts of the electric field. From Eqs. (8) and (9), the first six $F(k)$'s are calculated as

$$F(1)=0\ , \tag{12a}$$

$$F(2)=\kappa_2\ , \tag{12b}$$

$$F(3)=\kappa_3+3\kappa_2\ , \tag{12c}$$

$$F(4)=\kappa_4+4\kappa_3+6\kappa_2+3\kappa_2^2\ , \tag{12d}$$

$$F(5)=\kappa_5+5\kappa_4+10\kappa_3+10\kappa_2+10\kappa_3\kappa_2+15\kappa_2^2\ , \tag{12e}$$

$$F(6)=\kappa_6+6\kappa_5+15\kappa_4+20\kappa_3+15\kappa_2+15\kappa_4\kappa_2+10\kappa_3^2+60\kappa_3\kappa_2+15\kappa_2^3+45\kappa_2^2\ . \tag{12f}$$

For the light field for which the first-order correlation function is known, those factorial moments can be readily evaluated from Eqs. (10) and (12).

C. Autocorrelation function

For the stationary light field, the second-order correlation function $G^{(2)}(\tau, T)$ of the photoelectric numbers registered during the time intervals of T at the times t_1 and t_2 depends only on the time difference $\tau(=|t_1-t_2|)$. It can be, furthermore, related to the correlation function of the light intensity as[12]

$$G^{(2)}(\tau,T)\equiv\langle n\,n(\tau)\rangle=\alpha^2\int_{-T/2}^{T/2}\int_{\tau-T/2}^{\tau+T/2}\langle I(t_1)I(t_2)\rangle\,dt_1dt_2,\quad \tau>T \tag{13}$$

where n and $n(\tau)$ are numbers counted at the proper-time origin and time τ, respectively. For Gaussian light, the correlation function $\langle I(t_1)I(t_2)\rangle$ is related to the first-order correlation function of the light field by the next expression[1]:

$$\langle I(t_1)I(t_2)\rangle = C^{(1)}(0)^2 + |C^{(1)}(t_1 - t_2)|^2 \ . \tag{14}$$

We studied experimentally the light scattered by polystyrene spheres suspended in water. They have no rotational diffusion and their translational diffusion constant D can be given by the Stokes-Einstein relation[15]

$$D = kT_0/6\pi\eta r \ , \tag{15}$$

where k is the Boltzmann constant and T_0, η and r are the temperature in K, the viscosity of the solvent, and the radius of the sphere, respectively. The spectral profile of the light scattered by the particles undergoing a Brownian motion is Lorentzian[16] when the incident light is monochromatic. The spectral half-width γ of the scattered light can be given by

$$\gamma = DK^2 \ , \tag{16}$$

where K is the scattering vector of the scattered light. The correlation time τ_c of the scattered light is defined to be the reciprocal of γ,

$$\tau_c = 1/\gamma \ . \tag{17}$$

III. EXPERIMENTAL APPARATUS

The system of the experiments is composed of a photoelectron counting system and an autocorrelation measuring system. Both systems are online with a minicomputer model YOHPAC 2100 by Yokogawa Hewlett Packard and the counted numbers are processed during and after the counting. The light source used was a 4880-Å argon laser model 52G-A by Coherent Radiation which was mode selected with a temperature-controlled etalon plate. The experimental arrangement is shown in Fig. 1. We observed the light scattered by polystyrene spheres through two irises at right angles with the direction of the incident laser light. The scattered light illuminated the photomultiplier with the second iris, 150 μm in diameter, which was 80 cm apart from the first iris in front of the sample cell. We used the first iris with a diameter of 150 μm for the measurement of the factorial moment and the autocorrelation, and with a diameter of 600 μm for the measurement of the probability distribution. The photomultiplier used was a Hamamatsu TV R464, from which dark pulses were a few per sec with our discriminating level. Photoelectric pulses were amplified by two wide-band amplifiers[17] and discriminated by a EG&G T105/N discriminator. The output of the discriminator was standardized to TTL level. The final shape of the pulses had a 2.5×10^{-8}-sec width. Those pulses were counted by a counting circuit on an interface card in the computer. The total counting speed was systematically limited to less than 30 MHz by the TTL counter used. The dead time of the counting system was, therefore, about 3×10^{-8} sec. An external clock prepared strobe pulses for transferring every counted number from latch memories and clear pulses for every beginning of count intervals. The details of the counting system and the processing procedure will be reported elsewhere. Polystyrene spheres of diameter 1090 Å with a standard deviation of 27 Å were suspended in water.

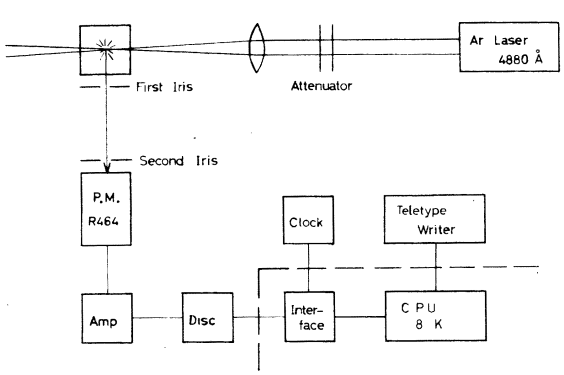

FIG. 1. Experimental arrangement.

The laser was carefully aligned in order to oscillate transversely in the TEM_{00} mode and longitudinally in a single mode. A part of the output of the laser was detected by a photosensor which was coupled to the feedback circuit and the discharge current of the laser tube was controlled in order to stabilize the laser intensity. Because the high-frequency part of the incident laser light causes the reduction of the amplitude of the correlation of the scattered light and a fluctuation of the frequency comparable to the correlation time may cause an error of the measured correlation time, it is desirable to eliminate the fluctuation of the incident laser light. The polarization ratio of the laser light was measured to be 100:1 for the electric field. It is, therefore, thought that the theoretical treatment with an assumption of a linear polarization well accounts for our experimental results.[18]

IV. EXPERIMENTAL RESULT

A. Probability distribution

The probability distributions which were obtained are shown in Fig. 2 for the count intervals 0.05 $\times10^{-3}$, 2×10^{-3}, and 5×10^{-3} sec [marked as (a), (b), and (c), respectively, in the figure]. The distribution for the incident laser light was also measured and is shown in the figure [marked as (d)]. The sample number is one million for the cases (a), (b), and (d) and 200 000 for that of (c). The values of $F(2)$ and $\langle n \rangle$ were evaluated from the obtained distributions and substituted into Eq. (5) to calculate the theoretical distributions. The solid curves in the figure show them. The approximate theory well interprets the measured distributions. The experimental value 25.44 of $\langle n \rangle$ for the laser light was substituted into Eq. (4) and the resultant curve of the Poisson distribution was also plotted in the figure.

B. Factorial moment

Before the calculation of the cumulants, let us consider the effect of the spatial coherence on the measured values of $F(k)$ and $G^{(2)}(\tau, T)$. For the amplitude of the second cumulant, the correction factor was derived by Jakeman *et al.*[19] It is a function of the radii of the employed irises and the distance between them. According to their results, our correction is calculated to be less than 0.002% for the measurements of the factorial moments and the autocorrelation function and 0.022% for the probability distributions. The effect of the spatial coherence is, therefore, safely neglected.

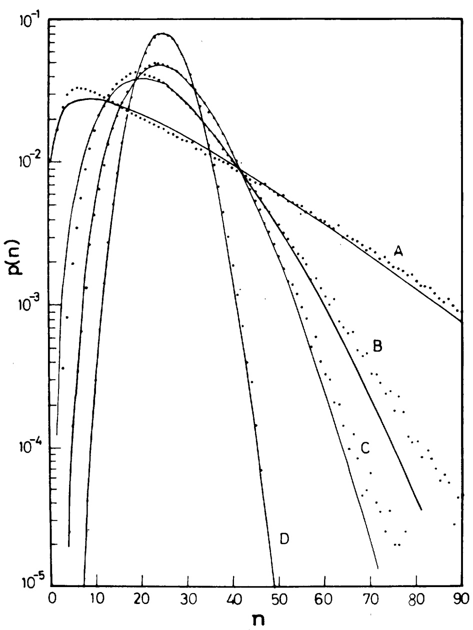

FIG. 2. Probability distribution. (a) Scattered light, $T = 0.05\times 10^{-3}$ sec and $\langle n \rangle = 25.39$. (b) Scattered light, $T = 2\times 10^{-3}$ sec and $\langle n \rangle = 24.91$. (c) Scattered light, $T = 5\times 10^{-3}$ sec and $\langle n \rangle = 26.52$. (d) Incident laser light, $T = 0.05\times 10^{-3}$ sec and $\langle n \rangle = 25.44$. The solid lines show the theoretical curves.

When the incident laser light fluctuates by a small but wide-band residual modulation, the spectral density $I(\omega)$ of the laser intensity is expressed as

$$I(\omega) = I_0\left((1-m)\delta(\omega-\omega_0) + \frac{m\gamma_0}{\pi[(\omega-\omega_0)^2+\gamma_0^2]}\right), \tag{18}$$

where ω_0 and I_0 are the center frequency and the total intensity of the laser light, respectively, and m is a fraction of the total fluctuating intensity. In the above equation, the noise spectrum of the laser light was assumed to be Lorentzian with a half-width of γ_0. The spectral density $I_s(\omega)$ of the scattered light is, therefore, given by the integration over the laser frequency,

$$I_s(\omega) = I_s\int_0^\infty \frac{\gamma}{\pi[(\omega-\omega')^2+\gamma^2]} \times\left((1-m)\delta(\omega'-\omega_0) + \frac{m\gamma_0}{\pi[(\omega'-\omega_0)^2+\gamma_0^2]}\right)d\omega' ,$$

where γ is the intrinsic spectral width of the scattered light, which is given by Eq. (16) for the light scattering by a thermal fluctuation.

The first-order correlation function $C^{(1)}(t_1-t_2)$ for the scattered light is obtained by the Fourier transform of $I_s(\omega)$,

$$C^{(1)}(t_1-t_2) = \langle I_s\rangle \iint_0^\infty e^{i\omega(t_1-t_2)} \times\left((1-m)\delta(\omega'-\omega_0) + \frac{m\gamma_0}{\pi[(\omega'-\omega_0)^2+\gamma_0^2]}\right) \times\frac{\gamma}{\pi[(\omega-\omega')^2+\gamma^2]}\,d\omega\, d\omega' .$$

Integrating the right-hand side of the above equation, one obtains the final form

$$C^{(1)}(t_1-t_2) = \langle I_s\rangle[(1-m) + m e^{-\gamma_0|t_1-t_2|}] \times e^{-\gamma|t_1-t_2| + i\omega_0(t_1-t_2)} . \tag{19}$$

When the noise spectrum is so wide that the condition

$$\gamma_0|t_1-t_2| \gg 1$$

is satisfied, the above relation is given by a simple expression

$$C^{(1)}(t_1-t_2) = (1-m)\langle I_s\rangle\, e^{-\gamma|t_1-t_2| + i\omega_0(t_1-t_2)}, \quad t_1 \neq t_2 . \tag{20}$$

The next equation is also, of course, obtained from Eq. (19):

$$C^{(1)}(0) = \langle I_s\rangle . \tag{21}$$

From Eqs. (10) and (20), it is easily seen that the

nth cumulant is reduced by a factor of $(1-m)^n$. The apparent form of the cumulants is calculated straightforwardly from Eqs. (10) and (20). The first six cumulants are obtained as follows:

$$\kappa_1 = 1 \ , \tag{22a}$$

$$\kappa_2 = \frac{2f^2}{a^2}\,[a + (e^{-a} - 1)] \ , \tag{22b}$$

$$\kappa_3 = \frac{12f^3}{a^2}\left(e^{-a} + 1 + \frac{2}{a}\,(e^{-a} - 1)\right) , \tag{22c}$$

$$\kappa_4 = \frac{12f^4}{a^2}\left(4e^{-a} + \frac{10}{a}(2e^{-a} + 1) + \frac{1}{a^2}(e^{-2a} + 28e^{-a} - 29)\right) , \tag{22d}$$

$$\kappa_5 = \frac{20f^5}{a^2}\left(8e^{-a} + \frac{72}{a}e^{-a} + \frac{12}{a^2}(e^{-2a} + 20e^{-a} + 7) + \frac{24}{a^3}(e^{-2a} + 12e^{-a} - 13)\right) , \tag{22e}$$

$$\kappa_6 = \frac{60}{a^2 f^6}\left(8e^{-a} + \frac{112}{a}e^{-a} + \frac{24}{a^2}(2e^{-2a} + 27e^{-a}) + \frac{72}{a^3}(3e^{-2a} + 25e^{-a} + 7) + \frac{4}{a^4}(e^{-3a} + 66e^{-2a} + 495e^{-a} - 562)\right) , \tag{22f}$$

where

$$a \equiv 2\gamma T = 2DTK^2 \tag{23}$$

and

$$f \equiv 1 - m \ . \tag{24}$$

The factorial moments $F(k)$ for $k=2$ to 6 were measured with the same experimental system as one for the measurement of the probability distributions but for the smaller first iris. The measurements were done for several count intervals from 2×10^{-5} to 5×10^{-2} sec. The experimental plots are shown in Fig. 3. Each experimental point has several hundreds of thousands data for the shorter count intervals and several tens of thousands data for the longer count intervals. In order to compare the theoretical expressions (12a)–(12f) to the experimental results, one must be given the values of τ_c and f. They were experimentally determined to be $(421.6\pm3.2)\times10^{-6}$ sec and 0.96, respectively, from the measurement of the autocorrelation function. By substituting these values into Eqs (22) and (23), the first six cumulants were evaluated. The theoretical curves of $F(k)$ are shown in Fig. 3. They are in good agreement with the experimental plots. This agreement implies that the factorial moments and the autocorrelation function were consistently measured.

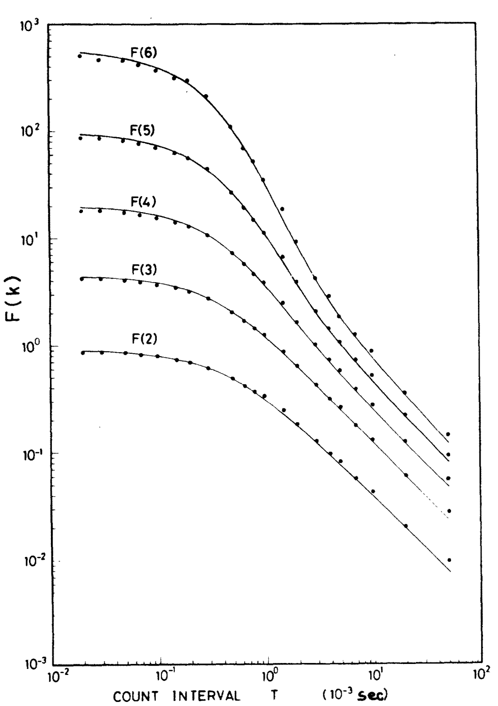

FIG. 3. Factorial moment. Solid lines show the theoretical curves evaluated with the values f and τ_c obtained by the measurement of $g^{(2)}(\tau, T)$.

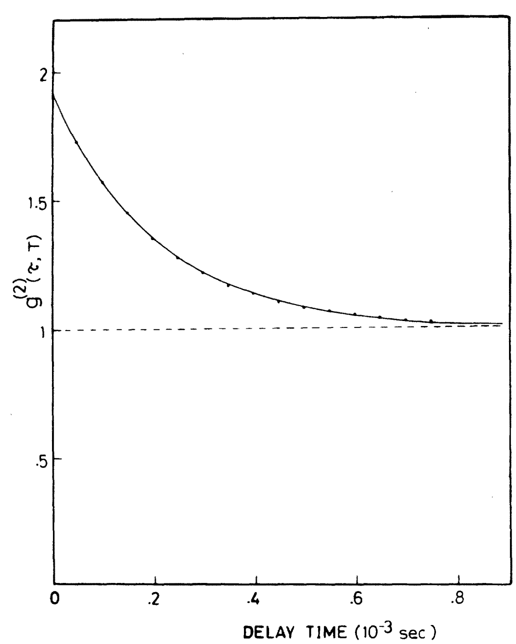

FIG. 4. Autocorrelation function. Each point consists of five million samples. Solid line shows the most probable curve of the experimental points from which the correlation time and the value of f were found to be $(421.6\pm3.2)\times10^{-6}$ sec and 0.96, respectively.

C. Autocorrelation function

The first-order correlation function for a Gaussian-Lorentzian light was given by Eqs. (20) and (21). Substituting Eqs. (20) and (21) into Eq. (14), one obtains the expression

$$\langle I_s(t_1)I_s(t_2)\rangle = \langle I_s\rangle^2 + (1-m)^2\langle I_s\rangle^2 e^{-2\gamma|t_1-t_2|}. \quad (25)$$

From the above relation and Eq. (13), the normalized second-order correlation function $g^{(2)}(\tau, T)$ is obtained

$$g^{(2)}(\tau, T) = \frac{\langle n n(\tau)\rangle}{\langle n\rangle^2} = 1 + f^2\left(\frac{\sinh(a/2)}{a/2}\right)^2 e^{-a\tau/T}, \quad \tau > T. \quad (26)$$

Figure 4 shows the experimental plot of $g^{(2)}(\tau,T)$. The count interval for each experimental point was 2×10^{-5} sec. Each point consists of five million samples. The average number of photoelectric pulses was typically 0.74. The correlation time was obtained as $(421.6\pm3.2)\times10^{-6}$ sec by a least-regression method for the decay slope of the experimental plot. The Stokes-Einstein relation predicted the correlation time to be $(421.6 \pm 3.6)\times10^{-6}$ sec, which was evaluated by the substitution of our physical quantities into Eqs. (15) and (16). The theoretical prediction of the correlation time, therefore, fits the experimental value well. By the extrapolation of the experimental plot to a zero time difference, the value of f was obtained to be 0.96. The fraction of the noise power of the laser light used was, therefore, estimated to be 4%.

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