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Photon statistics of multimode-laser light scattered by thermal particles

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Microscopic scattering processes are discussed for two structures of light incident on thermal particles. One is composed of two collinear light beams originating from different oscillating modes of a laser, and another is composed of two counter propagating beams originating from the same oscillating mode. A comparison of the cumulant expansions for the above two cases has allowed discerning which terms in the scattering structures led to the same statistical properties of the scattered photons. It is shown that, for the scattering of multimode-laser light, the statistical properties of scattered photons are not affected by the noncorrelating random phases of the incident laser fields but are the same as those in the case of single-mode laser light under practical scattering experiments. The factorial moments $F_k[=\langle n(n-1)\cdots(n-k+1)\rangle]$ up to sixth order of the scattered photons were measured for various scattering situations including the above two cases, and they were found to support the theoretical results.

I. INTRODUCTION

Quasimonochromatic lasers have been frequently used as stable light sources in many light scattering experiments. While the statistical nature of single-mode laser light scattered from random optical fluctuations has been examined in considerable detail,[1-12] the influence of the mode structure of incident laser light on the statistical properties of scattered photons has been left unsolved. In the multimode case, the instantaneous laser field of every mode scattered at one point in the scattering medium reflects the optical fluctuations at that point. Each fluctuating field is not directly correlated with the other because of random phase diffusion which is inherent in the oscillating modes of a laser.[13-14] The magnitude of the time correlation between the scattered photons, normalized by the average squared photon number might, therefore, be expected to become smaller than that in the case of single-mode laser light. In fact the second-order correlation of scattered light intensity for the case of a two-mode laser with equal intensity in each mode incident on the scattering medium, had been calculated. It was found to be two times smaller than that in a single-mode case in an earlier paper.[15] In a previous work[12] we examined the influence of the polarization properties of the incident laser light on the statistics of scattered photons. This time we report the influence of another fundamental property of the laser light, i.e., the laser-mode configurations.

We discuss the microscopic processes of light scattering for two mode structures of light incident on thermal particles and calculate the second-order factorial moment of scattered photons. First, the scattering of two collinear light beams of different laser modes will be treated. Next we consider the scattering process of two incident laser beams originating from the same laser mode but with opposite propagations. For both cases it is shown that under the practical experimental situations, the statistical properties of the scattered photons are substantially equal to those in the case of single-mode laser light. It will be also shown that although these cases result in similar statistical properties of scattered photons, the detailed scattering processes consist of different fine structures.

By using a standard photon-counting technique the factorial moments of the order from 2 to 6 of scattered photons were measured for various scattering configurations including the above cases, and we obtained the experimental results which support the theoretical conclusions.

II. THEORETICAL

Thermal particles whose kinetics are described by a Brownian motion, change their positions randomly while scattering the incident light field. Then the phase of the optical field at any observing point which is scattered by such a particle is thought to be random and diffusive. Assuming the scattering amplitude of an individual particle to be statistically deterministic, we present the optical field at the observation point by summing over the contributions from every particle in the scattering volume, and by examining the time correlation of the scattered photons.

We consider the situation where two light beams are incident on a system of random moving particles. These beams are denoted by 1 and 2. The analytic signal[16] $V(t)$ at the pin-point photodetector will be given by the superposition of the light fields scattered from each particle with a relative phase difference $\phi(t,r)$ scattered from each particle (Appendix),

$$V(t)=\sum_{\alpha}\eta_{\alpha}V_{\alpha}(t)\times\sum_{j}\exp\{i[\phi_{\alpha j}(t,r)-\omega_{\alpha}t+\varphi_{\alpha}(t)]\}, \quad (1)$$

where $V_\alpha(t)$ is incident light field of αth mode, and ω_α and r denote the frequency of the αth mode and the position of the particle, respectively. The coefficient η is the scattering efficiency and is assumed to be independent of an individual particle or its position. The time-dependent function $\varphi_\alpha(t)$ denotes the phase of αth incident laser field which is assumed to be random and obeying a Fokker-Planck equation. The number of scattered photons counted during a unit count interval can be directly calculated from Eq. (1) as

$$\begin{aligned} n(t)=V^*(t)V(t)=&\sum_{\alpha}\eta_{\alpha}^2|V_{\alpha}(t)|^2\sum_{jk}\exp\{i[\phi_{\alpha j}(t,r)-\phi_{\alpha k}(t,r)]\}\\ &+\eta_1\eta_2[V_1^*(t)V_2(t)\sum_{jk}\exp\{-i[\phi_{1j}(t,r)-\phi_{2j}(t,r)-(\omega_1-\omega_2)t+\varphi_1(t)-\varphi_2(t)]\}\\ &+\eta_1\eta_2[V_2^*(t)V_1(t)\sum_{jk}\exp\{i[\phi_{1j}(t,r)-\phi_{2j}(t,r)-(\omega_1-\omega_2)t+\varphi_1(t)-\varphi_2(t)]\}, \end{aligned}\quad (2)$$

here the count interval was assumed to be very short compared to the correlation time of the scattered field in order to avoid an influence of the finite count duration. The ensemble average of the number of photons over the incident light fields and the system of the random particles is, therefore, given as

$$\langle n(t)\rangle=\sum_{\alpha}\eta_{\alpha}^2\langle n^0(t)\rangle N=\langle n_1(t)\rangle+\langle n_2(t)\rangle, \quad (3)$$

here $n_\alpha[=\eta_\alpha^2 n_\alpha^0(t)N]$ is the scattered photon number of the αth mode, and $n_\alpha^0(t)[=|v_\alpha^*(t)V_\alpha(t)|]$ denotes the number of the incident photons of the αth mode. The time correlation of photons $g_s^{(2)}(t,t')(t\neq t')$ is also directly given from Eq. (1) as

$$\begin{aligned} \langle n(t)n(t')\rangle=&\Big\langle\Big(\sum_{\alpha}\eta_{\alpha}^2|V_{\alpha}(t)|^2\sum_{jk}\exp\{-i[\phi_{\alpha j}(t,r)-\phi_{\alpha k}(t,r)]\}\Big)\\ &\times\Big(\sum_{\beta}\eta_{\beta}^2|V_{\beta}(t')|^2\sum_{lm}\exp\{-i[\phi_{\beta l}(t',r)-\phi_{\beta m}(t',r)]\}\Big)\Big\rangle\\ &+\Big\langle\Big(\sum_{\alpha}\eta_{\alpha}^2|V_{\alpha}(t)|^2\sum_{jk}\exp\{-i[\phi_{\alpha j}(t,r)-\phi_{\alpha k}(t,r)]\}\Big)\\ &\times\eta_1\eta_2\Big(V_1^*(t')V_2(t')\exp\{-i[(\omega_1-\omega_2)t'-(\varphi_1(t')-\varphi_2(t')]\}\\ &\times\sum_{lm}\exp\{-i[\phi_{1l}(t',r)-\phi_{2m}(t',r)]\}+\text{c.c.}\Big)\Big\rangle\\ &+\Big\langle\Big(\sum_{\beta}\eta_{\beta}^2|V_{\beta}(t')|^2\sum_{lm}\exp\{-i[\phi_{\beta l}(t',r)-\phi_{\beta m}(t',r)]\}\Big)\\ &\times\eta_1\eta_2\Big(V_1^*(t)V_2(t)\exp\{-i[(\omega_1-\omega_2)t-(\varphi_1(t)-\varphi_2(t)]\}\sum_{jk}\exp\{-i[\phi_{1j}(t)-\phi_{2k}(t)]\}+\text{c.c.}\Big)\Big\rangle\\ &+\Big\langle\eta_1^2\eta_2^2\Big(V_1^*(t)V_2(t)\exp(-i\{(\omega_1-\omega_2)t-[\varphi_1(t)-\varphi_2(t)]\})\sum_{jk}\exp\{-i[\phi_{1j}(t,r)-\phi_{2k}(t,r)]\}+\text{c.c.}\Big)\\ &\times\Big(V_1^*(t')V_2(t')\exp(-i\{(\omega_1-\omega_2)t'-[\varphi_1(t')-\varphi_2(t')]\})\\ &\times\sum_{lm}\exp\{-i[\phi_{1l}(t',r)-\phi_{2m}(t',r)]\}+\text{c.c.}\Big)\Big\rangle\,(t\neq t'). \end{aligned}\quad (4)$$

We confine ourselves to the two scattering configurations mentioned earlier, before making any further algebraic manipulation of Eq. (4).

Case A: Two collinear laser beams originating from different laser modes

In this case, we can immediately obtain the reduced form of Eq. (4) by dropping oscillating terms

$$\langle n(t)n(t')\rangle = \sum_{\alpha\beta} \eta_\alpha^2\eta_\beta^2 \langle |V_\alpha(t)|^2 |V_\beta(t')|^2\rangle_f \sum_{jk}\sum_{lm} \langle \exp\{-i(\phi_{\alpha j}(t,r) - \phi_{\alpha k}(t,r) + \phi_{\beta l}(t',r) - \phi_{\beta m}(t',r))\}\rangle_p \quad (t \neq t') . \tag{5}$$

Here $\langle\ \rangle_f$ and $\langle\ \rangle_p$ denote the ensemble average over the incident light fields and the system of the random particles, respectively. The phase correlation function

$$\xi_{\alpha\beta} \equiv \langle \exp\{-i[\phi_{\alpha j}(t,r) - \phi_{\alpha k}(t,r) + \phi_{\beta l}(t',r) - \phi_{\beta m}(t',r)]\}\rangle_p \tag{6}$$

reflects the random movement of the thermal particles to the autocorrelation of the scattered photons. Among various combinations of particles in $\xi_{\alpha\beta}$, only the following combinations contribute: $j=k$ and $l=m$, or $j=m$ and $k=l$ $(j\neq k)$. The former contribution obviously gives rise to an accidental coincidence among scattered photons, and the latter combination results in the excess fluctuations in the scattered light fields. Equation (5) is then reduced as

$$\langle n(t)n(t')\rangle = \sum_{\alpha\beta} \eta_\alpha^2\eta_\beta^2 \langle n_\alpha^0(t) n_\beta^0(t')\rangle_f \left(N^2 + \sum_{j\neq k} \xi_{\alpha\beta}\right), \quad (t\neq t') . \tag{7}$$

We rewrite the form of $\xi_{\alpha\beta}$ as

$$\begin{aligned} \xi_{\alpha\beta} &= \langle \exp\{-i[\phi_{\alpha j}(t,r) - \phi_{\beta j}(t',r)]\}\rangle_p \langle \exp\{i[\phi_{\alpha k}(t,r) - \phi_{\beta k}(t',r)]\}\rangle_p \\ &= |\langle \exp\{-i[\phi_\alpha(t,r) - \phi_\beta(t',r)]\}\rangle_p|^2 . \end{aligned} \tag{8}$$

When $\alpha\neq\beta$, we call $\xi_{\alpha\beta}$ a crosscorrelation function between the scattered light beams or modes. It will be worth noting that $\xi_{\alpha\beta}(\alpha\neq\beta)$ determines the normalized magnitude of the fluctuations of the optical phase in two-mode laser beams scattered and does not involve any influence from the random phase diffusions of the incident laser fields. The scattering scheme of $\xi_{\alpha\beta}$ given by Eq. (7) is illustrated in Fig. 1(a).

Now we examine the amplitude of $\xi_{\alpha\beta}$ in the following. Expressing the relative phase $\phi(t,r)$ of the scattered light field with the scattering vector $\Delta\vec{K}$ and the position vector of the particle $\vec{r}(t)$ as

$$\phi(t,r) = \Delta\vec{K}\cdot\vec{r}(t) , \tag{9}$$

we can write down the apparent form of $\xi_{\alpha\beta}$ as

$$\xi_{\alpha\beta} = |\langle \exp\{i\Delta\vec{K}_\alpha\cdot[\vec{r}(t) - \vec{r}(t')]\}\rangle_p|^2 \, |\langle \exp[-i(\Delta\vec{K}_\alpha - \Delta\vec{k}_\beta)\cdot\vec{r}(t)]\rangle_p|^2 . \tag{10}$$

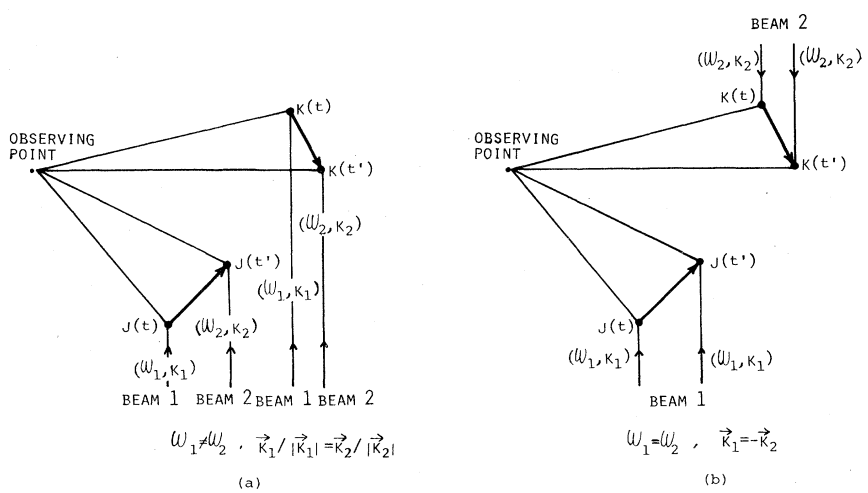

FIG. 1. Schematic diagrams of the light scattering of two light beams. (a) The case of two collinear laser beams originating from different laser modes (case A). (b) The case of two incident laser beams originating from the same oscillating mode but with opposite propagations (case B). These scattering structures give an origin to the additional $\frac{1}{2}$ in the normalized values of F_2.

For gas lasers which are usually used as a light source for scattering experiments, the frequency spread of the oscillating mode is at most 10^9 Hz. Then the second factor of Eq. (10) will be safely thought to be unity since under the usual experimental situations, the scattering volume is much larger than the wavelength $2\pi/|\vec{K}|$ but is much smaller than $2\pi/|\Delta\vec{K}_\alpha - \Delta\vec{K}_\beta|$, i.e.,

$$\gamma_{\alpha\beta} \equiv |\Delta\vec{K}_\alpha - \Delta\vec{K}_\beta| \times (\mathcal{S}) \cong 0 , \tag{11}$$

where $\mathcal{S}$ is the scattering dimension. Therefore

$$\xi'_{\alpha\beta} \equiv |\langle \exp[-i(\Delta\vec{K}_\alpha - \Delta\vec{K}_\beta)\cdot\vec{r}(t)]\rangle_p|^2 = 1 , \tag{12}$$

and then

$$\xi_{\alpha\beta} = |\langle \exp\{i[\phi_\alpha(t,r) - \phi_\alpha(t',r)]\}\rangle_p|^2 = \xi_{\alpha\alpha'} \tag{13a}$$

or

$$\xi_{11} = \xi_{12} = \xi_{21} = \xi_{22} . \tag{13b}$$

From Eq. (7) we finally obtain the equation

$$g_S^{(2)}(t,t') = g_L^{(2)}(t,t')\, g_M^{(2)}(t,t') . \tag{14a}$$

Here we assumed $N \gg 1$, and each correlation function is defined as

$$g_s^{(2)}(t,t') \equiv \langle n(t)n(t')\rangle , \tag{14b}$$

$$g_L^{(2)}(t,t') \equiv N^2 \sum_{\alpha\beta} \eta_\alpha^2 \eta_\beta^2 \langle n_\alpha^0(t) n_\beta^0(t')\rangle_f , \tag{14c}$$

and

$$g_M^{(2)}(t,t') \equiv 1 + |\langle \exp\{i[\phi(t) - \phi(t')]\}\rangle_p|^2 , \tag{14d}$$

respectively. Then the time correlation function of the scattered photons is factorized into those of incident photons and of the fluctuating motion of particles. Then the normalized correlation function is given by

$$\frac{\langle n(t)n(t')\rangle}{\langle n(t)\rangle\langle n(t')\rangle} = 1 + |\langle \exp\{i[\phi(t) - \phi(t')]\}\rangle_p|^2 (t \neq t') . \tag{15}$$

Here we assumed that the total number of the incident photons is constant.[17] When $t = t'$, we obtain the second-order factorial moment of the scattered photons from Eq. (15). The normalized second-order factorial moment F_2 is, therefore, given as

$$F_2 \equiv \frac{\langle n(n-1)\rangle}{\langle n\rangle^2} - 1$$

$$= |\langle \exp\{i[\phi(t) - \phi(t')]\}\rangle_p|^2_{t=t'} = 1 . \tag{16}$$

The above result is the same as for the case of a single-mode light scattering.[12] From the earlier work,[15] F_2 was given by $\frac{1}{2}$, i.e., one half of our resultant value. This disagreement can be attributed to Eq. (43) in that paper. In this equation the correlation between the light intensities $I(t)$'s of each scattered mode was factorized as

$$\langle I_1(t) I_2(t')\rangle = \langle I_1(t)\rangle\langle I_2(t')\rangle .$$

This will be equivalent to claiming in our language that $\xi_{\alpha\beta}$ *a priori* vanishes for $\alpha \neq \beta$. This will not generally hold.

When the light source is in multicolor operation instead of multimode operation as the case of simultaneous oscillation at 488.0 and 514.5 nm in an Ar ion laser, Eq. (11) may no longer stand but γ_{12} will vanish. This time $\xi_{\alpha\beta}$ becomes zero for $\alpha \neq \beta$ (α and β denote, this time, each oscillating line), then F_2 is given by

$$F_2 = (\langle n_1\rangle^2 + \langle n_2\rangle^2)/(\langle n_1\rangle + \langle n_2\rangle)^2 .$$

When $\langle n_1\rangle = \langle n_2\rangle$, F_2 becomes $\frac{1}{2}$.

If the propagation vectors of the αth and βth modes are opposite each other instead of being collinear, it will be seen that $\gamma_{\alpha\beta}$ becomes very large and $\xi_{\alpha\beta}$ can be approximated by zero for $\alpha \neq \beta$. This time F_2 will be again $\frac{1}{2}$ for the incident beams of equal intensities. However, when two incident light beams originate from the same oscillating mode, F_2 is shown to retain the magnitude 1 again as in the case of the two collinear laser beams of different modes (case A). But the original scattering configuration which gives an additive $\frac{1}{2}$ to the value of F_2 will be shown to be quite different.

Case B: Two incident laser beams originating from the same oscillating mode but with opposite propagations

In this case the first term in Eq. (4) does not change its form but its crosscorrelation function $\xi_{\alpha\beta}$ vanishes since their propagation vectors are opposite, $\vec{K}_1 = -\vec{K}_2$. Then the first term in Eq. (4) is given by the following.

First term:

$$\sum_{\alpha\beta} \langle n_\alpha^0(t)\rangle_f \langle n_\beta^0(t')\rangle_f N^2 \eta_\alpha^2 \eta_\beta^2 + \sum_\alpha \eta_\alpha^4 \langle n_\alpha^0(t)\rangle_f^2 N(N-1)\, |\langle \exp\{-i[\phi_\alpha(t) - \phi_\alpha(t')]\}\rangle|^2 . \tag{17}$$

The second term is rewritten as follows.

Second term:

$$\sum_{\alpha}\eta_\alpha^2\langle n_\alpha^0(t)\rangle_f\eta_1\eta_2\langle V_1^*(t')V_2(t')\exp\{i[\varphi_1(t')-\varphi_2(t')]\}\rangle_f\sum_{jk}\sum_{lm}\langle\exp\{-i[\phi_{\alpha j}(t,r)-\phi_{\alpha k}(t,r)+\phi_{1l}(t',r)-\phi_{2m}(t',r)]\}\rangle_p$$

$$+\sum_{\alpha}\eta_\alpha^2\langle n_\alpha^0(t)\rangle_f\eta_1\eta_2\langle V_1(t')V_2^*(t')\exp\{-i[\varphi_1(t')-\varphi_2(t')]\}\rangle_f$$

$$\times\sum_{jk}\sum_{lm}\langle\exp\{-i[\phi_{\alpha j}(t,r)-\phi_{\alpha k}(t,r)-\phi_{1l}(t',r)+\phi_{2m}(t',r)]\}\rangle_p\,.$$

Since the ensemble average of the phase diffusion

$$\langle\exp\{\pm i[\varphi_1(t)-\varphi_2(t')]\}\rangle_f=\langle\exp\pm i\varphi_1(t)\rangle_f\langle\exp\mp i\varphi_2(t')\rangle_f$$

vanishes, this term will not contribute. From the same argument, the third term also vanishes. The fourth term in Eq. (4) is reduced to

Fourth term:

$$\eta_1^2\eta_2^2\langle V_1^*(t)V_1(t')V_2(t)V_2^*(t')\exp\{i[\varphi_1(t)-\varphi_1(t')-\varphi_2(t)+\varphi_2(t')]\}\rangle_f$$

$$\times\sum_{jk}\sum_{lm}\langle\exp\{-i[\phi_{1j}(t,r)-\phi_{1l}(t',r)-\phi_{2k}(t,r)+\phi_{2m}(t',r)]\}\rangle_p+\text{c.c.}$$

$$+\eta_1^2\eta_2^2\langle V_1^*(t)V_1^*(t')V_2(t)V_2(t')\exp\{i[\varphi_1(t)+\varphi_1(t')-\varphi_2(t)-\varphi_2(t')]\}\rangle_f$$

$$\times\sum_{jk}\sum_{lm}\langle\exp\{-i[\phi_{1j}(t,r)+\phi_{1l}(t',r)-\phi_{2k}(t,r)-\phi_{2m}(t',r)]\}\rangle_p+\text{c.c.}$$

In this expression the second term vanishes since the ensemble averages over the system of the particles are zero. In the first term of the above equation, the ensemble average over the system of particles is different from zero for the following combination pair of particles: $j=1$ and $k=m$. The random phase factor in this term

$$\langle\exp\{i[\varphi_1(t)-\varphi_2(t)+\varphi_2(t')-\varphi_1(t')]\}\rangle_f$$

is thought to be almost unity since the light beams 1 and 2 originate from the same oscillating mode, so the equations $\varphi_1(t)=\varphi_2(t)$ and $\varphi_1(t')=\varphi_2(t')$ will stand always. Therefore, the fourth term is given by

Fourth term:

$$\eta_1^2\eta_2^2\langle n_1^0(t)\rangle_f\langle n_2^0(t)\rangle_f\sum_{jk}\langle\exp\{-i[\phi_{1j}(t,r)-\phi_{2k}(t,r)+\phi_{2k}(t',r)-\phi_{1j}(t',r)]\}\rangle_p+\text{c.c.}$$

$$=\langle n_1(t)\rangle\langle n_2(t)\rangle\langle\exp\{-i[\phi_1(t,r)-\phi_1(t',r)]\}\rangle_p\langle\exp\{i[\phi_2(t,r)-\phi_2(t',r)]\}\rangle_p+\text{c.c.}\qquad(18)$$

This time the crosscorrelation $\xi_{\alpha\beta}$ is given by

$$\xi_{\alpha\beta}=\langle\exp\{-i[\phi_{1j}(t,r)-\phi_{2k}(t,r)+\phi_{2k}(t',r)-\phi_{1j}(t',r)]\}\rangle_p\,.\qquad(19)$$

The scattering scheme of $\xi_{\alpha\beta}$ in Eq. (19) is illustrated in Fig. 1(b). From Eqs. (17) and (18), the correlation function of the scattered photons is given by

$$\langle n(t)n(t')\rangle=\sum_{\alpha\beta}\langle n_\alpha\rangle_f\langle n_\beta\rangle_f+\sum_{\alpha}\langle n_\alpha\rangle_f^2|\langle\exp\{i[\phi_\alpha(t,r)-\phi_\alpha(t',r)]\}\rangle|^2$$

$$+\langle n_1\rangle_f\langle n_2\rangle_f[\langle\exp\{-i[\phi_1(t,r)-\phi_1(t',r)]\}\rangle\langle\exp\{i[\phi_2(t,r)-\phi_2(t',r)]\}\rangle+\text{c.c.}]\quad(t\neq t')\,.\qquad(20)$$

Therefore, we obtain the second-order factorial moment F_2 of the scattered photons as

$$\langle n(n-1)\rangle=\sum_{\alpha\beta}\langle n_\alpha\rangle_f\langle n_\beta\rangle_f+\sum_{\alpha}\langle n_\alpha\rangle_f^2+2\langle n_1\rangle_f\langle n_2\rangle_f\,.$$

Then the normalized second-order factorial moment F_2 of the scattered photons is given by

$$F_2=\frac{\langle n(n-1)\rangle}{\langle n\rangle^2}-1=1\,.$$

This time we again obtain the same results as in the single-mode case. It will be clearly seen from the analysis so far and the illustrations in Fig. 1 that the nonzero crosscorrelating term ξ_{12} originates from the so-called Hanbury Brown-Twiss effect between the scattered modes in the case A but it comes from the direct product of the optical fields of the scattered light beams in the case B.

III. EXPERIMENTAL

The standard technique used for measuring the factorial moments of photoelectron pulses was employed to study experimentally the statistical properties of the scattered photons for the cases discussed above. The linearly polarized 514.5-nm Ar ion laser beam with a single or many longitudinal modes was used to irradiate polystyrene latex. The diameter of the polystyrene spheres was 481 nm with a 1.8-nm standard deviation. The scattered light was observed at right angle from the incident light beams through two irises whose diameters were 120 and 60 μm, respectively. They were 30 cm apart.

(i) First we measured the normalized factorial moments of the order from 2 to 6 for the scattered single-mode laser beam. The number of photons counted was 3×10^6.

(ii) Next the laser was operated longitudinally in multimode. This time about twenty longitudinal modes were oscillating simultaneously. All of these modes were collimated into the scattering cell. Data acquisition was 5×10^6.

(iii) The laser oscillated again longitudinally in a single mode. The transmitted light beam through the scattering cell was reflected back into the cell again on the same path. Both beams had equal intensity since the propagation path length through the cell was very short (less than 1 mm) and the transmitted beam suffered little scattering loss. Sampling number was 3×10^6.

(iv) Next the reflected beam was shifted from the position of the first-coming light beam by 2 mm in the lateral plane in the scattering cell so that the photodetector could see two separate scattering regions. The sample number was this time 5×10^6.

(v) Finally the laser oscillated in a multicolor. It emitted dominant laser lines at 488.0 and 514.5 nm and a few weaker lines. This time the reflecting mirror was again removed. The number of data was 2×10^6.

The photoelectron count interval was 42.14 μsec in all the above measurements. The correlation time of the scattered photons was calculated to be 445 μsec for all the above experiments, i.e., approximately ten times longer than the count interval.

IV. DISCUSSION

Table I shows the values of the normalized factorial moments F_k measured for the various scattering configurations mentioned in Sec. III. The first column gives the values obtained for the case of the single-mode light beam. The reduced value 0.92 instead of unity is attributed to the effects of a finite count interval and a spatial coherence. The factorial moments of higher orders are also smaller than the theoretical values neglecting these effects. In this paper the relative magnitude of the factorial moments for various experimental configurations will be sufficient to examine the theoretical results.

The second column gives the values for the multimode case. We can see no significant departure from the single-mode case except for the statistical error caused by the finite sample number. When the laser oscillated in multimode, the individual longitudinal modes had large fluctuations in their intensity.[17] This does not, however, affect the values of the factorial moments measured. This is due to the fact that the total light intensity was kept constant even though the longitudinal components changed their intensities,[17] and is consistent with the result given in Eq. (15).

The case of opposite propagation is shown in the third column and the values are almost equal to those in the single-mode case as expected from the theoretical results. As distinct from case (ii) in which the "unity" value of F_2 is attributed in part to the scattering scheme shown in Fig. 1(a),

TABLE I. Measured factorial moments $F_k[\equiv \langle n(n-1)\cdots(n-k+1)\rangle/\langle n\rangle^k - 1]$ for the five different mode configurations of the incident laser beams. In the parentheses in the fifth column the calculated values of F_k are shown.

	(i) single-mode	(ii) multimode	(iii) opposite (overlapped)	(iv) opposite (separated)	(v) multicolor
F_2	0.916 ± 0.008	0.909 ± 0.004	0.918 ± 0.009	0.910 ± 0.002	0.3282 ± 0.0002 (0.37)
F_3	4.50 ± 0.07	4.44 ± 0.04	4.57 ± 0.06	4.47 ± 0.04	1.267 ± 0.002 (1.39)
F_4	20.0 ± 0.8	19.6 ± 0.5	20.9 ± 0.4	20.0 ± 0.6	3.82 ± 0.02 (4.07)
F_5	98 ± 9	96 ± 5	109 ± 3	98 ± 7	11.5 ± 0.2 (9.6)
F_6	549 ± 96	551 ± 58	678 ± 46	560 ± 102	37 ± 1 (35.1)

the unity value of F_2 in this case can be in part be attributed to the direct product of the scattered light field of each mode. In this scattering scheme, as can be seen in Fig. 1(b), each scattering particle scatters photons of either incident light beam and does not interact with both the incident light beams. Two incident light beams will, therefore, need not be spatially overlapped. The fourth column shows the factorial moments obtained under this scattering configuration. As expected no significant departure can be seen from the case of the spatially overlapped light beams. For the scattering scheme shown in Fig. 1(a), each particle, on the contrary, interacts with both incident light beams which cannot occur when they are spatially separated.

Finally the last column shows the experimental values for the case of the multicolor laser light. In Sec. III it was seen that in the multimode case, the crosscorrelating functions ξ_{12} and ξ_{21} vanish since the difference of the scattering wave vectors are so large that the factor ξ_{12}' and ξ_{21}' vanish. In our experimental setup, the value of $|\Delta\vec{K}_\alpha - \Delta\vec{K}_\beta|$ is around 2×10^4 rad cm^{-1}. Then ξ_{12}' becomes very small. In the parentheses in this column we show the approximate values of F_k calculated by assuming that the kth cumulant of the scattered light intensity can be approximately reduced by a factor M^{1-k} with $M=2.2$; here M is an effective number of oscillating lines with equal intensity.

V. CONCLUSION

Although the detailed discussion of the scattering process was confined to the second-order time correlation of the scattered photons, the same treatment is applicable to the discussion of the higher-order statistical properties of the scattered photons. The multimode laser light scattered from thermal particles was shown to retain the same statistical properties as that in the case of a single-mode laser light. In the present discussions the influence of the heterodyning effect among the laser modes on the fluctuation properties of the scattered photons was neglected since it had been examined by Mandel.[15,18] Several experiments carried out supported all of the theoretical results.

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APPENDIX: ANALYTIC SIGNAL OF TWO LASER BEAMS SCATTERED BY RANDOM MOVING PARTICLES

We briefly introduce the analytic signal $V(t)$ of two laser fields scattered by random moving particles. The superposed field $V_L(t)$ of two laser fields incident on particles can be given by the sum of the analytic signals of each laser mode as

$$V_L(t)=\sum_\alpha V_\alpha(t)\exp\{-i[\omega_\alpha t-\varphi_\alpha(t)]\}\quad(\alpha=1,2)\,.$$

Here, α denotes αth mode and $V_\alpha(t)$ is the analytic signal of the αth laser mode, and ω_α and $\varphi_\alpha(t)$ represent the angular frequency and the phase of the αth optical field, respectively. When this superposed field is incident on the system of the random moving particles, each particle scatters independently the incident laser fields. Assuming the scattering efficiency η for each particle to be deterministic, the light field scattered by jth particle is given by

$$\sum_\alpha V_\alpha(t)\exp\{-i[\omega_\alpha t-\varphi_\alpha(t)]\}\{\eta_\alpha\exp[i\phi_{\alpha j}(t,r)]\}\,,$$

where $\phi_{\alpha j}(t,r)$ is the relative phase retardation introduced by the finite distance from the position r of jth particle to the observing point. This phase retardation fluctuates according to the random walk of the particle, and its fluctuation appears in the scattered light intensity when scattered light fields are summed over the scattering particles. Then the analytic signal $V(t)$ contributed from N particles at the detector is given by summing each scattered field over N particles as

$$T(t)=\sum_\alpha \eta_\alpha V_\alpha(t)\sum_i^N \exp\{i[\phi_{\alpha j}(t,r)-\omega_\alpha t+\varphi_\alpha(t)]\}\,.$$

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