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Single-interval statistics of light scattered by identical independent scatterers

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Abstract. We consider the statistical properties of coherent light Rayleigh-scattered by a solution containing an arbitrary number N of independent, identical particles. Expressions are obtained for the single-interval intensity probability distribution and the moments of this distribution, which are equivalent to the measurable photocount factorial moments. We discuss both the case of fixed N and the situation where N varies according to a Poisson distribution. For large N, the scattered electric field has, to a good approximation, gaussian statistics, but for small N, marked departures from gaussian statistics are predicted. It is argued that, in many scattering experiments, there is the potential to obtain from measurements in the non-gaussian regime (N small) useful system-dependent information which is not available in the gaussian regime.

1. Introduction

In the decade or so since the advent of the laser, the study of fluctuations in the intensity of scattered laser light has proved to be a powerful experimental tool, particularly in the investigation of fluid systems (see, for example, Benedek 1969, Pike 1969, Cummins and Swinney 1970, Jakeman and Pike 1974). It is the purpose of this paper to calculate the amplitude distribution of the electric field of light scattered by a model system of N independent scatterers, and, in doing so, to characterize the conditions under which useful information on a system under study can be extracted from the measured single-interval photocount distribution.

Hitherto emphasis has been on characterization of the time-dependence, or equivalently the power spectrum, of the fluctuating scattered intensity. A particular statistical form, gaussian statistics, has generally been *assumed* for the amplitude distribution, and the properties of that form have been used to relate measured quantities (photocount correlation functions, photocurrent spectra) to the dynamics of molecular motion. The gaussian field distribution is characterized by a single parameter, the average intensity. Thus no other system-dependent information can be extracted from the field statistics in the gaussian limit, and single-interval fluctuation statistics in scattering experiments have largely been ignored.

The gaussian assumption is generally quite good. In fact it is usually fairly difficult to observe non-gaussian statistics. Gaussian statistics are expected whenever the total scattered field can be viewed as the sum of *many*, *independent* contributions. This fact will be demonstrated below using a random walk approach to sum the independently-phased contributions (see, also, Cummins and Swinney 1970, Beckmann and Spizzichino 1963). In order to observe non-gaussian field statistics it is necessary to reduce the size of the scattering volume to the point where it no longer contains many independent scatterers. In the case of truly independent scatterers such as a dilute solution of macromolecules, marked deviations from gaussian statistics will be observed when the scattering volume contains less than about ten scatterers. Unless the scatterers are micron size or larger, the scattered field is usually too weak to be studied in this limit. Similarly, light scattered by correlated scatterers will deviate measurably from gaussian form when the volume over which the scatterer motions and positions are correlated becomes comparable to the scattering volume, even though the actual number of scatterers is large. However, correlation volumes, as, for instance, in pure fluids, are generally much smaller than diffraction-limited scattering volumes, making the non-gaussian regime difficult to achieve even near critical points.

Recent experiments have, nevertheless, demonstrated that the non-gaussian regime can be achieved both for independent and correlated scatterers. In the case of independent scatterers, non-gaussian statistics have been observed for aqueous suspensions of polystyrene spheres about 1 μm in diameter. (Schaefer and Berne 1972, Schaefer and Pusey 1972, 1973). In this case the unique system-dependent information obtained from the fluctuation statistics is the number density of scatterers. It should be remembered that in the gaussian limit neither the single-interval statistics nor the time-dependence of the intensity fluctuations yields this information. Non-gaussian statistics have also been observed recently for a nematic liquid crystal in its 'dynamic scattering' mode (Jakeman and Pusey 1973a, b). In this case the phase of the electric field transmitted by a thin film of liquid crystal is correlated over a range of several microns; this correlation range can be obtained from the statistics of the scattered light. Once again, this information cannot be determined from the statistics in the gaussian limit. Similar effects have been observed in scattering from coarse-grained ground glass (Bluemel *et al* 1972). It should be noted also that non-gaussian statistics have been encountered for several years in the field of laser anemometry (see, for example, Di Porto *et al* 1969, Bourke *et al* 1970). However, in these latter measurements, emphasis has been on the time-dependence, rather than the single-interval statistics, of the scattered light.

Thus it can be said that, if gaussian statistics are observed in an experiment, the experiment is, by virtue of a 'large' scattering volume, designed, intentionally or otherwise, to mask some potentially useful information about the system under study. If the scattering volume in such an experiment were reduced until it contained only a few correlation volumes, the statistics would cease to be gaussian and would be dependent on the system in question, thus providing further information.

Although correlated systems are potentially more interesting, this paper will concentrate on the statistics of light scattered by an arbitrary number of *independent* particles. The theory produced here is useful for estimating the deviation from gaussian statistics to be expected in any experiment on identical independent scatterers such as dilute solutions of polymers or biological macromolecules, cells etc. It can also be considered as the simplest model of a correlated system, where each correlation volume is regarded as an independent 'particle'. This approach has already been used with some success in the liquid crystal work mentioned above.

Several simplifying assumptions are made throughout this paper. These assumptions include a uniformly illuminated scattering volume, total spatial coherence at the detector, and sampling intervals short compared to the fluctuation time of the scattered intensity. These conditions, not always achieved in practice, allow exact expressions to be obtained for $P(I)$, the probability distribution of the integrated intensity, and for the moments of this distribution (or equivalently, the photocount factorial moments). If these assumptions are not made, exact expressions for the above quantities are difficult to obtain, although the first few intensity moments can be calculated in a rather tedious way (Schaefer and Pusey 1972, 1973).

The theoretical treatment in this paper leans heavily on studies of the two-dimensional random walk problem performed some seventy years ago (Kluyver 1905, Pearson 1906, Rayleigh 1919). Non-gaussian statistics in light scattering have also been discussed by Korenman (1970) using a rather general approach, and to some extent by Cantrell (1968).

2. Theory

Consider a solution of N identical non-interacting particles, dispersed in a liquid medium, illuminated by plane monochromatic light, whose scattering is observed in the far-field by a detector with active area much smaller than a coherence area. Let us assume the particles to be spherically symmetrical and/or small compared to the wavelength of the incident light, so that rotational effects are negligible. The complex analytic signal $E^+(t)$, the positive frequency part of the electric field (see, for example, Born and Wolf 1965, p 495), observed at the detector is then

$$E^+(t) = \beta\, \mathrm{e}^{\mathrm{i}\omega t} \sum_{j=1}^{N} \exp(\mathrm{i}\boldsymbol{K} \cdot \boldsymbol{r}_j) \tag{1}$$

where β^2 is the mean intensity scattered by one particle, ω is the angular frequency of the light, $\boldsymbol{K}$ is the scattering vector and $\boldsymbol{r}_j$ is the instantaneous position of the jth scatterer. (If the particles move in a random fashion as, for instance, in brownian motion, the electric field will fluctuate randomly with coherence time roughly equal to the time it takes one particle to move a distance $1/|\boldsymbol{K}|$.) We have assumed the incident illumination to be uniform throughout the scattering volume, and will also assume the dimensions of the scattering volume to be large compared to $1/|\boldsymbol{K}|$. It is then evident that if the scatterers move in such a way that $\boldsymbol{r}_j$ can have any value within the scattering volume, regardless of the positions of the other scatterers (uniform mean density of non-interacting scatterers), $\boldsymbol{K} \cdot \boldsymbol{r}_j$ is a random variable whose principal value takes on all values from 0 to 2π with equal probability. Thus equation (1) describes a two-dimensional random walk of N unit steps in the complex plane. For large N, it is well known that this leads to a gaussian probability distribution for E^+ (see, for example, Glauber 1965, equation (14.48))

$$P(E^+) = \frac{1}{\pi\langle|E^+|^2\rangle} \exp\left(\frac{-|E^+|^2}{\langle|E^+|^2\rangle}\right), \tag{2}$$

where

$$\langle|E^+|^2\rangle = N\beta^2. \tag{3}$$

Identifying

$$I \equiv |E^+|^2 \tag{4}$$

as the instantaneous intensity of the scattered light, the probability distribution of the intensity $P_N(I)$, in the large N limit, is given by

$$[P_N(I)]_{N\to\infty} = \frac{1}{\langle I \rangle} \exp\left(\frac{-I}{\langle I \rangle}\right). \tag{5}$$

Thus, though the electric field has gaussian statistics, $P_N(I)$ is exponential.

For small N equations (2) and (5) are not valid. However, an expression for $P_N(I)$ for arbitrary N can be obtained simply from Kluyver's (1905) work on the two-dimensional random walk, with the result

$$P_N(I) = \tfrac{1}{2} \int_0^\infty U J_0(U\sqrt{I})(J_0(U\beta))^N \, \mathrm{d}U \tag{6}$$

(see also Pearson 1906, Rayleigh 1919). Here J_0 is the zero-order Bessel function of the first kind. For one scatterer, $N = 1$, equation (6) reduces, as it must, to a δ function at $I = \langle I \rangle = \beta^2$. For $N = 2$,

$$P_2(I) = \frac{1}{\pi[I(2\langle I \rangle - I)]^{1/2}}, \tag{7}$$

where $\langle I \rangle = 2\beta^2$. This result can be obtained from equation (6); it can be derived more simply from first principles. An expression exists for $P_3(I)$ in terms of elliptic integrals (Pearson 1906, Rayleigh 1919). For $N > 3$, however, it appears that no closed-form solution of equation (6) has been obtained, although Pearson has performed a graphical solution for $N < 7$. With normalizations appropriate for our problem Pearson's results are plotted in figures 1 and 2 for $N = 2, 3, 4$ and 5; $\langle I \rangle P_N(I)$ is plotted

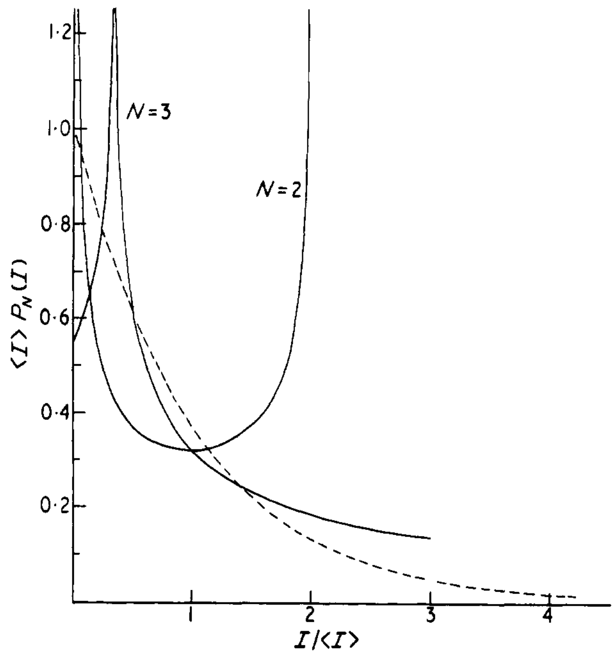

Figure 1. Single-interval intensity distribution $P_N(I)$ for fixed $N = 2$ and 3. The broken curve is the exponential limit (gaussian field distribution).

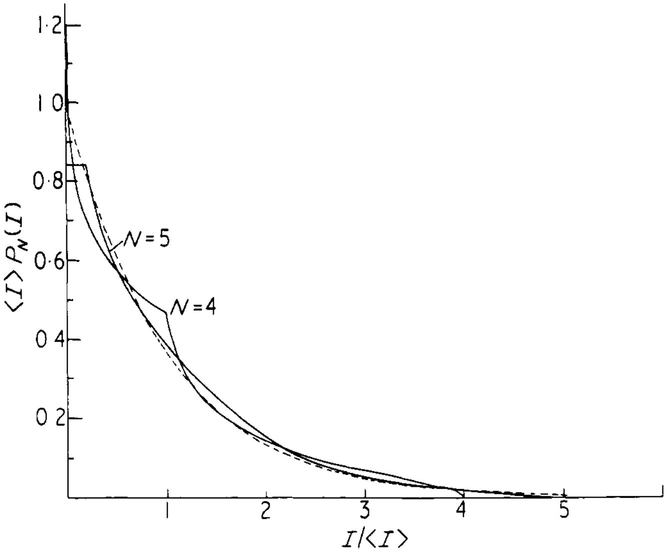

Figure 2. $P_N(I)$ for fixed $N = 4$ and 5.

against $I/\langle I\rangle$ so that direct comparison can be made with equation (5) (the broken curve). For small N, the light is clearly non-gaussian, $P_N(I)$ showing both infinities and discontinuities; however even for N as small as 5 the approach to exponential form is quite evident. For $N > 7$ there are no infinities or discontinuities and a good approximation to $P_N(I)$ can be obtained from the first few terms of a series expansion of equation (6), with the result:

$$P_N(I) = \frac{1}{\langle I\rangle}\mathrm{e}^{-I/\langle I\rangle}\left[1-\frac{1}{4N}\left(2-\frac{4I}{\langle I\rangle}+\frac{I^2}{\langle I\rangle^2}\right)\right.$$
$$\left.+\frac{1}{12N^2}\left(1-12\frac{I}{\langle I\rangle}+15\frac{I^2}{\langle I\rangle^2}-\frac{14}{3}\frac{I^3}{\langle I\rangle^3}+\frac{3}{8}\frac{I^4}{\langle I\rangle^4}\right)+\dots\right],\qquad N > 7. \qquad (8)$$

At first sight, the occurrence of infinities in the probability functions $P(I)$ might be considered rather surprising. It should be realized, however, that the *integrated* probability that I lies in an interval $\mathrm{d}I$ about some value will always be finite. A rather appealing comparison with a one-dimensional random walk can be invoked to explain, at least in part, the values of I at which the infinities or discontinuities occur. For a one-dimensional random walk of one step, the resultant length must, of course, be one step; the same holds true in two dimensions. For a one-dimensional random walk of two steps the resultant length will be either zero or two steps, each possibility having probability $\frac{1}{2}$. In two dimensions the probability distribution is smoothed somewhat, but there are still infinites in $P_2(I)$ corresponding to the δ functions in the one-dimensional case. For three steps in one dimension there are two possibilities: a resultant length of one with probability $\frac{3}{4}$, corresponding to the infinity at $I/\langle I\rangle = \frac{1}{3}$ in the two-dimensional case (figure 1), and a resultant length of 3 with probability $\frac{1}{4}$, corresponding to the cut-off at $I/\langle I\rangle = 3$ in figure 1.

The above results have been derived assuming the number of particles N in the scattering volume to be constant. Although such a situation can, in principle, be achieved it is simpler experimentally to allow N to vary. In a typical experiment one will define by optical means a scattering volume much smaller than the sample volume itself. Under the assumptions of particle independence and constant mean number density of particles within the sample, the distribution $P(N)$ of particles within the scattering volume will be Poisson (Chandrasekhar 1943). The coherence time associated with fluctuations in N will be much longer than that for the 'interference fluctuations' discussed earlier, the former being of the order of the time it takes a particle to move across the scattering volume (many wavelengths), while the latter is of order of the time it takes a particle to move one wavelength. Denoting the intensity probability distribution by $P_N(I)$ for fixed N and $P_{\langle N\rangle}(I)$ for N variable, we obtain an expression for $P_{\langle N\rangle}(I)$ by averaging equation (6) with a Poisson distribution for N, ie

$$P_{\langle N\rangle}(I) = \sum_{N=0}^{\infty} P(N)P_N(I), \tag{9}$$

where

$$P(N) = \frac{\langle N\rangle^N \exp(-\langle N\rangle)}{N!} \tag{10}$$

to give

$$P_{\langle N\rangle}(I) = \tfrac{1}{2}\int_0^\infty UJ_0(U\sqrt{I})\exp[\langle N\rangle(J_0(U\beta)-1)]\,\mathrm{d}U. \tag{11}$$

Figure 3 shows $P_{\langle N\rangle}(I)$ for $\langle N\rangle = 2$. Some 32% of the area under this curve consists of a δ function (not shown in figure 3) at $I/\langle I\rangle = 0{\cdot}5$, corresponding to the time when only one particle is in the scattering volume. The infinity shown in figure 3 at $I/\langle I\rangle = 0{\cdot}5$ arises from the $P_3(I)$ term in equation (9).

Although expressions have been derived above for the intensity distribution $P(I)$, one cannot measure $P(I)$ directly. Rather one measures $P(n)$, the probability of accumulating n detected photons, or photocounts, in a sample time, assumed here to be short

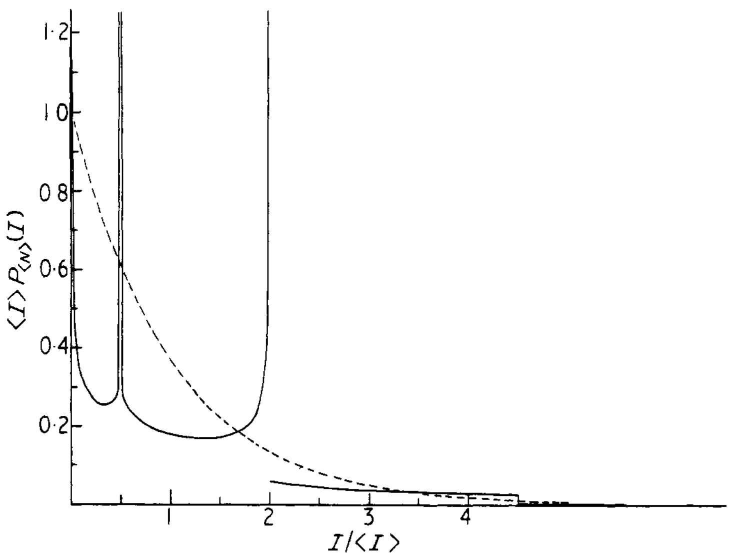

Figure 3. $P_{\langle N\rangle}(I)$ for variable N, $\langle N\rangle = 2$.

compared to the coherence time of the scattered light. Due to the stochastic nature of the detection process, $P(n)$ will not, in general, show the fine detail of $P(I)$. Only when the average number of photocounts detected per sampling interval is large, does $P(n)$ approach $P(I)$. One can obtain $P(n)$ from $P(I)$ using the Mandel relationship (Mandel 1963). This leads to a formal expression for $P(n)$ in terms of an apparently intractable sum to infinity of the moments of $P(I)$. A more productive approach is to calculate the moments of $P(I)$. The normalized intensity moments $\langle I^m \rangle / \langle I \rangle^m$ are equal to the normalized photocount factorial moments $\langle n(n-1)\ldots(n-m+1)\rangle/\langle n\rangle^m$, easily obtained from the measured $P(n)$ (this result is inherent in equation (10b) of Mandel 1959; see also Pike 1969, Cantrell 1970).

Consider the case for fixed N. We construct the generating function

$$\phi_N(\lambda) = \int_0^\infty e^{-\lambda I} P_N(I)\, dI \tag{12}$$

from which

$$\langle I^m \rangle = \left(\frac{-\partial}{\partial \lambda}\right)^m \phi_N(\lambda)\bigg|_{\lambda=0}. \tag{13}$$

From equations (6) and (12),

$$\begin{aligned}\phi_N(\lambda) &= \tfrac{1}{2}\int_0^\infty \left(\int_0^\infty e^{-\lambda I} J_0(U\sqrt{I})\, dI\right) U (J_0(U\beta))^N\, dU \\ &= \frac{1}{2\lambda}\int_0^\infty U \exp\left(-\frac{U^2}{4\lambda}\right)(J_0(U\beta))^N\, dU \\ &= \frac{1}{4\lambda}\int_0^\infty \exp\left(\frac{-x}{4\lambda}\right)(J_0(\sqrt{x}\beta))^N\, dx.\end{aligned}$$

Writing $(J_0(\sqrt{x}\beta))^N$ as a power series

$$(J_0(\sqrt{x}\beta))^N = \sum_{p=0}^{\infty} Q_p (x\beta^2)^p,$$

one finds

$$\phi_N(\lambda) = \sum_{p=0}^{\infty} Q_p \beta^{2p} p! (4\lambda)^p.$$

Thus, from equation (13)

$$\langle I^m \rangle = (m!)^2 \beta^{2m} (-4)^m Q_m. \tag{14}$$

The Q_m are evaluated in the appendix leading to the following expression:

$$\langle I^m \rangle = (m!)^2 \beta^{2m} \sum_{l=1}^{m} \frac{N!}{(N-l)!} \sum_{\{a\}} \left(\prod_{j=1}^{m} (j!)^{2a_j} (a_j)!\right)^{-1} \tag{15}$$

where the a_j are non-negative integers and the summation over $\{a\}$ is performed subject to the conditions

$$\sum_{j=1}^{m} j a_j = m \qquad \text{and} \qquad \sum_{j=1}^{m} a_j = l. \tag{16}$$

As before, equation (15) can be averaged with a Poisson distribution $P(N)$ (see equation (10)) for N to give $\langle I^m \rangle$ for variable N. Since

$$\sum_{N=0}^{\infty} \frac{N!}{(N-l)!} P(N) = \langle N \rangle^l, \tag{17}$$

we can write, using equations (3), (4), (15) and (17),

$$\frac{\langle I^m \rangle}{\langle I \rangle^m} = \frac{1}{\langle N \rangle^m} \sum_{l=1}^{m} C_{l,N} D_{m,l} \tag{18}$$

where

$$C_{l,N} = \begin{cases} \dfrac{N!}{(N-l)!} & \text{for } N \text{ fixed} \\ \langle N \rangle^l & \text{for } N \text{ variable} \end{cases} \tag{19}$$

and

$$D_{m,l} = (m!)^2 \sum_{\{a\}} \left(\prod_{j=1}^{m} (j!)^{2a_j} a_j! \right)^{-1}. \tag{20}$$

The $D_{m,l}$ are given in table 1 for the first six moments. The moments for variable N could equally well have been obtained by constructing the generating function for equation (11).

Table 1. Values of $D_{m,l}$

l \ m	1	2	3	4	5	6
1	1	1	1	1	1	1
2		2	9	34	125	461
3			6	72	650	5400
4				24	600	10500
5					120	5400
6						720

3. Discussion

For large N the term in equation (18) for $l = m$ will dominate. For this term, only $a_1 = m, a_2 \ldots a_m = 0$ fulfil the conditions of equation (16). Thus

$$\lim_{\langle N \rangle \to \infty} \frac{\langle I^m \rangle}{\langle I \rangle^m} = m!. \tag{21}$$

Comparison of equation (21) with the moments obtained from equation (5) shows that, in the limit $\langle N \rangle \to \infty$, the light is, as it must be, gaussian, both for fixed and variable N.

The first five intensity moments for variable N are, from equation (18) and table 1,

$$\langle I\rangle/\langle I\rangle = 1$$

$$\langle I^2\rangle/\langle I\rangle^2 = 2+1/\langle N\rangle$$

$$\langle I^3\rangle/\langle I\rangle^3 = 6+9/\langle N\rangle+1/\langle N\rangle^2$$

$$\langle I^4\rangle/\langle I\rangle^4 = 24+72/\langle N\rangle+34/\langle N\rangle^2+1/\langle N\rangle^3$$

$$\langle I^5\rangle/\langle I\rangle^5 = 120+600/\langle N\rangle+650/\langle N\rangle^2+125/\langle N\rangle^3+1/\langle N\rangle^4.$$

In figure 4 we plot $\langle I^m\rangle/m!\,\langle I\rangle^m$ against $\langle N\rangle$ for $m = 2, 3, 4$. For gaussian light these quantities would be unity. For fixed N they are found to be less than one indicating intensity fluctuations of smaller magnitude than for gaussian light, whereas for variable N the intensity fluctuations have magnitude greater than those of gaussian light. In both cases the gaussian value is approached as $N \to \infty$. It should be noted that, although for $\langle N\rangle = 7$ $P(I)$ looks exponential to within a few per cent, the fourth moment, for instance, differs from the gaussian value by nearly 50%.

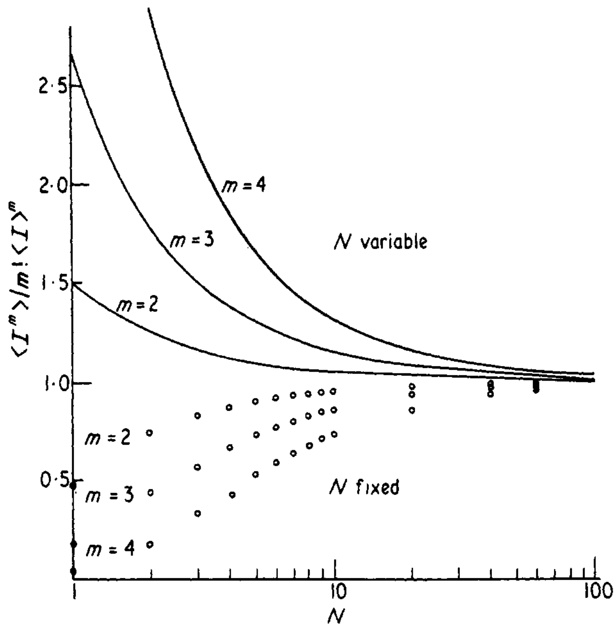

Figure 4. Second, third and fourth moments of $P(I)$ for fixed and variable N.

The expression for the moments of $P(I)$, equation (18), has also been obtained directly from equations (1) and (4), constructing $\langle I^m\rangle$ and using factorization properties valid for independent scatterers of the resulting average of products of sums (Chen *et al* 1973). This approach, however, does not yield expressions for $P(I)$.

As mentioned in the introduction experiments have been performed to test the predictions derived here (Schaefer and Pusey 1972, 1973). When account was taken of experimental effects such as non-uniform illumination of the scattering volume and incomplete spatial coherence, good agreement between experiment and theory for the first few moments was found.

Finally attention should be drawn to the fact that the mathematical treatment in this paper is similar to that used by Hodara (1965) when considering the statistics of multi-mode laser light under the somewhat unrealistic assumption of independently phased modes. The main difference between Hodara's work and ours is that he concentrated on the probability distribution of the electric field, the real part of equation (1), whereas we have been concerned with the experimentally accessible intensity probability distribution.

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Appendix. Calculation of Q_p

We require Q_p in the equation

$$(J_0(\sqrt{x}\beta))^N = \sum_{p=0}^{\infty} Q_p \beta^{2p} x^p. \tag{A.1}$$

We have, as an identity (Abramowitz and Stegun 1965, p 360),

$$J_0(\sqrt{x}\beta) = \sum_{q=0}^{\infty} \frac{(-\frac{1}{4}x\beta^2)^q}{(q!)^2} = 1 + \sum_{q=1}^{\infty} \frac{(-\frac{1}{4}x\beta^2)^q}{(q!)^2}.$$

Therefore

$$(J_0(\sqrt{x}\beta))^N = \sum_{l=0}^{N} \frac{N!}{(N-l)!\,l!} \left(\sum_{q=1}^{\infty} \frac{(-\frac{1}{4}\beta^2)^q}{q!} \frac{x^q}{q!} \right)^l.$$

Now

$$\left(\sum_{q=1}^{\infty} y_q \frac{x^q}{q!} \right)^l = l! \sum_{r=l}^{\infty} \frac{x^r}{r!} \sum_{\{a\}} \frac{r!\, y_1^{a_1} y_2^{a_2} \ldots y_r^{a_r}}{(1!)^{a_1}(2!)^{a_2} \ldots (r!)^{a_r} a_1!\, a_2! \ldots a_r!}$$

where the summation over $\{a\}$ is performed subject to the conditions

$$\sum_{j=1}^{r} a_j = l \qquad \text{and} \qquad \sum_{j=1}^{r} j a_j = r,$$

(Abramowitz and Stegun 1965, p 823). Thus

$$(J_0(\sqrt{x}\beta))^N = \sum_{l=0}^{N} \frac{N!}{(N-l)!} \sum_{r=l}^{\infty} x^r (-\tfrac{1}{4}\beta^2)^r \sum_{\{a\}} \left(\prod_{j=1}^{r} (j!)^{2a_j} a_j! \right)^{-1}.$$

Comparison with equation (A.1) gives

$$Q_p = (-\tfrac{1}{4})^p \sum_{l=0}^{N} \frac{N!}{(N-l)!} \sum_{\{a\}} \left(\prod_{j=1}^{p} (j!)^{2a_j} a_j! \right)^{-1}, \tag{A.2}$$

subject to

$$\sum_{j=1}^{p} a_j = 1, \qquad \sum_{j=1}^{p} j a_j = p. \tag{A.3}$$

Finally it can easily be shown that the limits on the summation over l in equation (A.2) can be changed to 1 and p.

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