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It is clear that an extension of the calculations to treat longer chains is required in order to resolve the differences noted above. A new method of generating freely rotating chains, based on the work of Scott *et al.* (1962) and Bernal (1964) on random packing of spheres, is being investigated, and it is hoped to present additional results in the near future.

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A measurement of optical linewidth by photon-counting statistics

Abstract. An optical linewidth of 20 Hz of laser light scattered by spherical particles undergoing Brownian motion has been measured experimentally by photon-counting statistics using the theory of the intensity-fluctuation distribution of Gaussian–Lorentzian light.

In a recent paper (Jakeman and Pike 1968) we presented the theory of the intensity fluctuations of Gaussian light having a Lorentzian spectrum of finite linewidth, and gave photon-counting distributions for such light as a function of counting rate and linewidth. In this letter we compare our theoretical predictions with experimental results obtained by scattering laser light from 0·6 μm diameter polystyrene spheres undergoing Brownian motion in water at room temperature. Such a system is expected theoretically (Glauber 1963, Pecora 1964) to produce Gaussian–Lorentzian scattered light, and has been used in previous investigations by Cummins *et al.* (1964) who used a heterodyne method to determine the linewidth, and by Dubin *et al.* (1967) and independently by Arecchi *et al.* (1967) who used a homodyne or self-beating method to determine the linewidth and also confirmed the Lorentzian form of the spectrum. The latter authors also verified the Gaussian nature of the scattered light by photon-counting statistics in the Bose limit, that is in the limit $\gamma\ (= \Gamma T)$ tends to zero, where Γ is the half-width at half-height of the spectral line and T is the sampling time.

The predicted theoretical value for the half-width is (Cummins *et al.* 1964)

$$\Gamma = |\mathbf{k}_s - \mathbf{k}_0|^2 \frac{kT_a}{12\pi^2\eta r} \tag{1}$$

where $\mathbf{k}_s$ and $\mathbf{k}_0$ are the wave vectors of the scattered and incident light, respectively, k is Boltzmann's constant, T_a the absolute temperature, η the viscosity and r the radius of the polystyrene spheres.

In the experiments reported here 300 μW of 6328 Å single-mode laser light from a Spectra Physics model 119 laser was scattered at a fixed angle of 33·5°. The rectangular scattering cell of cross section 5 mm × 5 mm was designed to avoid convection currents by maintaining a very small stabilizing vertical temperature gradient. The mean temperature was 23·5 °C. The density of polystyrene balls was such that the mean separation was of the order of fifteen diameters, giving a linear absorption coefficient of 0·2 cm^{-1}. The laser beam was focused to a waist diameter of 0·15 mm ($1/e^2$ points) at the centre of the cell, and the scattered light emerged normally to one face through a circular aperture of the same diameter. The photomultiplier detector was a cooled International Telegraph and Telephone type FW130 with dark-count rate at full sensitivity of 0·5 counts/s. This was placed behind a second aperture of 0·2 mm diameter at a distance of 7·4 cm from the first. The acceptance area at the detector was thus 0·073 of a coherence area; in the theoretical interpretation spatial-coherence effects were, therefore, neglected.

The probability $p(n, T)$ of detecting n photoelectrons in a sample time T is given by equations (42) and (49) of Jakeman and Pike (1968) and can also be found from the recurrence relation of Bédard (1966). It is a function of the parameter γ which was varied in these experiments over a range from 0·1 to 1·6 by varying T. The theoretical value of Γ from equation (1) for the numerical values of the parameters given above (η = 0·009 25 P) is 7·2 Hz. The times T, therefore, ranged from about 10 ms to 200 ms. At the longer times the dark-count rate was lowered to negligible values by reducing the efficiency.

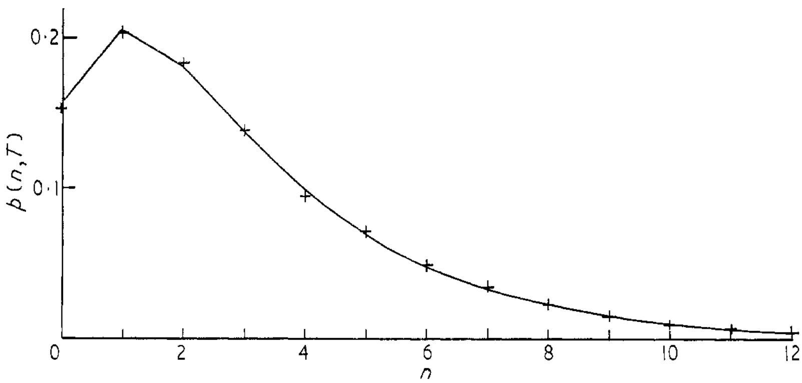

Figure 1. Photon-counting distribution for ΓT = 1·0, $\bar{n}$ = 3·0. The full curve is the theoretical value and the crosses are the experimental points.

A typical photon-counting distribution is shown by the crosses in figure 1. This was obtained with a sample time T of 100 ms and contains 10^4 samples. The mean $\bar{n}$ is approximately three events per sample time. The most accurate way to determine Γ from such a result is to use the formula for the normalized second factorial moment

$$n_2 = \sum_n \frac{n(n-1)p(n)}{\bar{n}^2} = 1 + \frac{1}{\gamma} - \frac{1}{2\gamma^2} + \frac{e^{-2\gamma}}{2\gamma^2}. \tag{2}$$

This formula follows from equation (50) of Jakeman and Pike (1968) and may also be obtained, along with exact expressions for higher factorial moments, from the recurrence relations given by Bédard (1966).

The value of Γ obtained from six experiments having different sample times was $9{\cdot}9 \pm 2{\cdot}2$ Hz and the theoretical photon-counting distribution for this linewidth is shown in figure 1 by the linked curve. The accuracy of the fit of the experimental results to the theory is conveniently assessed by comparing moments of the distributions; in figure 2 the normalized higher-order factorial moments obtained from the six experiments are compared with the behaviour predicted theoretically. The latter may be evaluated from

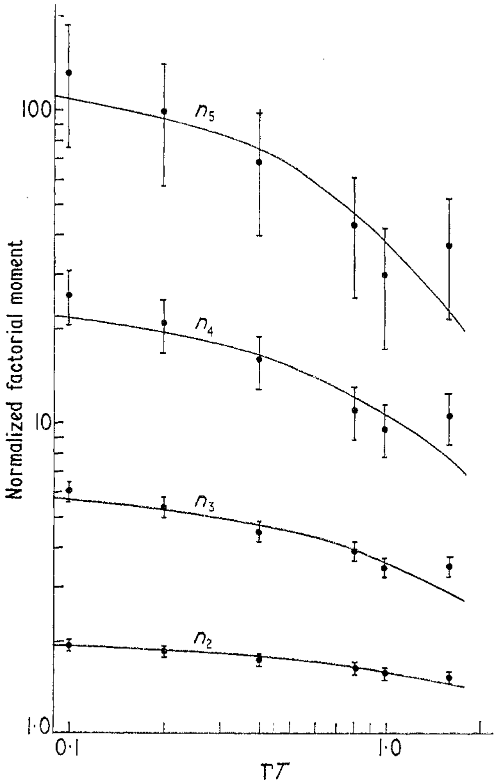

Figure 2. Normalized factorial moments of photon-counting distributions obtained with six different values of sample time T and a fixed linewidth of 20 Hz. The full curves are the theoretical values and the full circles are the experimental points.

the generating function $Q(s)$ given by equation (30) of Jakeman and Pike (1968) by employing the formula

$$n_r = \left(-\frac{1}{\bar{n}}\right)^r \left[\frac{d^r}{ds^r} Q(s)\right]_{s=0}. \qquad (3)$$

An erratum in Jakeman and Pike (1968) might be mentioned here; the unitary matrix which diagonalizes M in equation (19) is not S but

$$S' = \frac{S_{ik}^* \langle n_i \rangle^{1/2}}{\langle m_k \rangle^{1/2}}.$$

The results of the paper are unaffected.

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