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PREFACE

This second volume in the series "Advances in Quantum Electronics" comprises three major articles by groups of leading experts in their respective fields of research.

The first monograph by Pike and Johnson on Photon Statistics and Photon-correlation Spectroscopy is a review of a topic which has both theoretical and practical interest. A review of the statistical properties of optical fields and spectral processing techniques is followed by a summary of the authors' own work on the use of photon correlation techniques to measure scattering effects in a number of different media.

The second monograph by Brown, Cox, Shand and Williams on the spectroscopy of the rare earth doped chalcogenides follows the paper by Brown and Shand in Volume 1 on the Quantum Counter. The use of optical, E.P.R. and excitation spectroscopic techniques is described in detail as techniques for establishing the location of impurity ions in the chalcogenides.

The final monograph by Greenhow and Schmidt surveys the field of mode locked lasers, the theory of mode locking, techniques and applications. These three comprehensive review articles will be of value to graduate students, researchers and others wishing to survey a specific research field without the involvment of a full literature survey.

December, 1973

D. W. GOODWIN

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PHOTON STATISTICS AND PHOTON-CORRELATION SPECTROSCOPY

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I. INTRODUCTION

This review is concerned with the description and analysis of electrical signals generated by detectors of optical radiation in their role as sensors of a wide range of optical phenomenon. We shall refer to these generically as "optical signals". The quantum nature of the optical field, together with field fluctuations arising from the superposition, in a typical optical experiment, of many randomly phased contributions from the source, necessitates such description and analysis in terms of randomly fluctuating quantities possessing definite statistical correlations. These correlations are experimentally measureable and amenable to theoretical interpretation. Although optics is a subject of long standing, such statistical considerations are comparatively recent, essentially following the advent of the laser. This is because first, the "beating" together of the randomly phased components in "classical" experiments gives rise to very rapid fluctuations—even the most monochromatic "classical" light source fluctuates on a timescale of nano-seconds, too fast to be easily analysed by electronic processing techniques—and secondly, individual photodetections can only be recorded with comparatively modern detectors. The development of the laser and modern detectors has made it possible to generate and study processes occurring on much slower time scales, using methods of electronic-statistical analysis. Thus although the first successful statistical experiments were performed in the fifties (Forrester *et al.*, 1955) before the laser was invented, systematic studies of the statistical properties of a variety of optical fields were not carried out until the mid sixties (see for example Pike, 1969). From these original photon-counting experiments a whole new field of optical spectroscopy in the frequency range 1–10^8 Hz has evolved (Pike, 1970).

As indicated above, fluctuations in an optical signal arise in two ways; firstly, from the basic processes producing the signal whether one considers, for instance, the field produced by some arbitrary source, say a lamp or laser, or produced by scattering of a (usually) coherent beam from a target; secondly, from the quantum nature of the light field and the detection process itself. In the weak-signal limit the second source of fluctuations dominates, whereas in conditions of higher signal strength it becomes less significant. It is interesting to note that the first situation does not arise in the microwave region where the photon energies are much lower than room-temperature thermal energies. Moreover, differences in the detection process due to the high energy of the optical photon mean that even at high signal strengths analyses of fluctuations of optical and microwave fields differ considerably. Fluctuations in radar signals have of course been studied for many years (see for example Atlas, 1964; Skolnik, 1962). Indeed, a first requirement nowadays in the design of new radar equipment is to postulate (usually from suitable measurements) some model for the statistical fluctuations to be expected in wanted and unwanted returns. There is thus a large body of literature available to the optical spectroscopist with problems in the realm of signal processing and by adapting some of the more sophisticated techniques used in the microwave region for use at optical frequencies it has been possible both to take advantage of the digital nature of the signal and to optimize signal usage. This will be shown to make possible accurate spectroscopic measurements on weak light signals fluctuating on time scales ranging from 1–10^{-8} s.

Our review is planned broadly as follows. The next section is intended to familiarize the reader with some of the more relevant statistical qualities characterizing light fields. Section III discusses the statistical properties of the detected signal and generally follows the early historical development of the subject whilst section IV is concerned with the signal processing techniques currently being exploited in the field of photon-correlation spectroscopy. Section V reviews recent calculations relating to optimum system design whilst the final section is devoted to applications. The following somewhat more detailed description of the contents of each section will serve to clarify, for introductory purposes, the material to be covered.

In section II we discuss the statistical properties of the optical field before detection. Any statistically fluctuating quantity is described mathematically by a set of probability distributions which in the case of an optical field will be characteristic of the source. The same information is contained in the moments of the distributions and all the distributions or all the moments provide equivalent knowledge of the complete field statistics. The optical spectrum is in general only part of the information carried by the field and can therefore be determined without measuring the totality of distributions or moments. Its relationship with these quantities is all-important and will be discussed in some detail. In real experiments finite sample times and source areas must be used and their effect on the statistical properties of the signals will also be described.

In section III we concentrate on the post-detection signal. The incident field

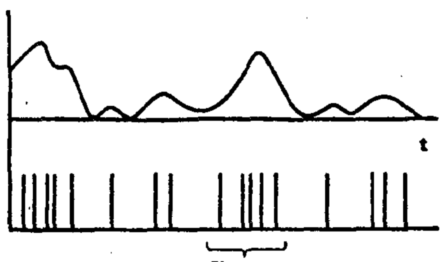

FIG. 1. Detection of scattered light.

is photodetected as a Poisson process and the post-detection signal consists of a series of amplified photo-electron pulses which are more bunched or thinned out as the intensity falling on the detector increases or decreases (Fig. 1). Such individual pulses recording each photodetection event are easily achieved with modern photomultiplier tubes. The distribution of photo-electron pulses at the output of the detector is therefore related to the properties of the source in a somewhat more complicated way than the incident field (Mandel, 1959). Nevertheless the optical spectrum can, in principle, be extracted from measurements of the photon-counting distributions or their moments and the relationship between these quantities will be discussed.

Section IV is devoted to the signal processing techniques which can be used in practice to obtain spectral information from an optical signal fluctuating in the 1–10^8 Hz frequency range. These techniques can be classed as reference-beam or direct methods. Reference beam methods include the familiar light beating techniques of heterodyne and homodyne (heterodyne at carrier frequency) "Doppler" spectroscopy (Pike 1969, 1970; Atlas, 1964; Benedek

1968; Cummins and Swinney, 1969) whilst the direct methods of intensity fluctuation measurement range from electrical filtering (see last mentioned references) (sometimes erroneously referred to as homodyning), through simple photon counting and coincidence counting to the more sophisticated digital autocorrelation techniques taken over from the microwave region (Jakeman, 1970).

Section V is given over to a discussion of the accuracy of spectral measurements using the methods described in section IV. Statistical fluctuations in the results of a set of experiments occur due to the finite duration over which time averages can be carried out in any real measurement. There is a good deal of current interest in devising processing schemes which minimize statistical errors arising from this source and recent work will be reviewed.

Finally in section VI we review recent applications of the techniques developed in this area of optical spectroscopy. These are many and range from fundamental studies such as Rayleigh scattering in liquids to measurements of more obvious practical importance such as those of fluid flow and turbulence.

II. Statistical Properties of Optical Fields

The statistical properties of light are contained in the correlations of the randomly fluctuating values of the real electric field operator $\mathscr{E}(\mathbf{r}, t)$ at different points in space and time. $\mathscr{E}(\mathbf{r}, t)$ is a real solution of Maxwell's equations and for its physical effects could be compared with a real solution of Schrodinger's equation. Wave or particle properties can be deduced from the field operator in the usual way, and it must be symmetrically quantized in the field volume according to the unit spin of the photon. An ideal light detector at a point in space responds not to the electric field itself but, by the theory of photoemission, to the modulus of the square of its positive-frequency part $\mathscr{E}^{+}(\mathbf{r}, t)$ which by second quantization gives the quantum mechanical photon annihilation operator in the coordinate representation.

$\hat{\mathscr{E}}^{+}(\mathbf{r}, t)$ is defined by

$$\hat{\mathscr{E}}(\mathbf{r}, t) = \sum_k [a_k e_k(\mathbf{r}, t) + a_k^{+} e_k^{+}(r, t)]$$
$$= \hat{\mathscr{E}}^{+}(\mathbf{r}, t) + \hat{\mathscr{E}}^{-}(\mathbf{r}, t) \tag{1}$$

with

$$e_k(\mathbf{r}, t) = i[\tfrac{1}{2}h\omega]^{1/2} u_k(\mathbf{r}) \, e^{-i\omega_k t} \tag{2}$$

where $u_k(\mathbf{r})$ is a normal mode of the field volume and the Bose operators a_k and a_k^{+} annihilate and create field quanta respectively. In a real detector, temporal and spatial integration will occur so that the statistical behaviour of the following quantity contains the information of interest:

$$E(\mathbf{r}, t; A, T) = \frac{1}{AT} \int_A \int_{t-T/2}^{t+T/2} d^2\mathbf{r}' \, dt' \, I(\mathbf{r}', t') \tag{3}$$

where T is a sample or integration time, A is an integration area centred on position r and

$$I(\mathbf{r}, t) = \langle \hat{\mathscr{E}}^{-}(\mathbf{r}, t)\hat{\mathscr{E}}^{+}(\mathbf{r}, t)\rangle_{Q} \tag{4}$$

where the Q-angle brackets indicate a quantum mechanical average. I and E will be referred to rather loosely as the intensity and integrated intensity respectively, and the properties of these quantities for various light sources is the subject of this section.

More specifically $I = \langle|\hat{\mathscr{E}}^{+}|^2\rangle_Q$, is the direct analogue of Born's familiar probability of electronic charge $\langle|\psi(\mathbf{r}, t)|^2\rangle_Q$, in terms of the second quantized wave function, and is interpreted similarly as the probability density of photons in the Maxwell field. We should point out that the use of the classical $|\mathscr{E}|^2$ as a measure of detected intensity, which is sometimes found in the literature, gives rise to spurious sum-frequency terms which are unphysical in the optical region and leads to incorrect photon-counting distributions but fortunately does not always give seriously misleading results.

It may also be worthwhile to point out that quantum mechanical averages of normally ordered operators such as occur in equation (4) can be calculated with some facility when the field state is expressed in Glauber's "P-representation" (Glauber, 1966).

A. THE PROBABILITY DISTRIBUTION OF INTENSITY FLUCTUATIONS

One of the simplest statistical properties of an optical field which can be investigated is the probability distribution of intensities, $P(I)$, illustrated in Fig. 2.

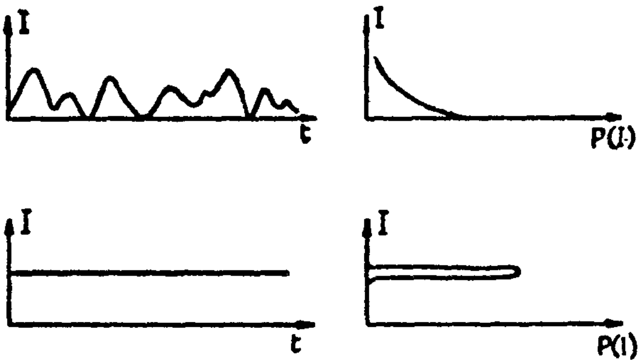

FIG. 2. Intensity fluctuation distribution of (a) incoherent and (b) coherent light.

Optical sources can be classified as incoherent or coherent according as the emitted intensity fluctuates or remains constant. An ideal laser operating well above threshold emits constant intensity radiation and its characteristic intensity fluctuation distribution (Fig. 2(b)) is essentially a δ-function. On the other hand thermal sources and incoherently scattered light fluctuate, leading to probability distributions of the form shown in Fig. 2(a). Many, though not all, incoherent sources may be more specifically classified technically as giving rise to "Gaussian" light. In this case the temporal field amplitude fluctuations are due to the fact that each frequency component of the light field from an incoherent source is usually made up from a sum of randomly phased contributions. According to the solution of the two-dimensional random-walk problem, the probability distribution of such a sum tends to become Gaussian as the number of contributions increases. This situation arises both in the case of thermal sources, each element of the source contributing in arbitrary phase, and also in the case of most scattering experiments, in which the scattering particles or dielectric fluctuations are randomly distributed over the scattering volume, thus again producing randomly phased contributions at the detector.

From these elementary statistical considerations, simple theoretical models for light fields have emerged. For example, the field from an amplitude stabilized laser operating well above threshold may be represented in the form

$$\mathscr{E}_c^+(\mathbf{r}, t) = \mathscr{E}_c\, e^{i[\mathbf{k}_c\cdot\mathbf{r} - \omega_c t + \phi(t)]} \tag{5}$$

where $\phi(t)$ is a random slowly fluctuating phase variable, ω_c and $\mathbf{k}_c$ the characteristic frequency and wave vector, respectively and $\mathscr{E}_c$ is constant.

A simple consequence of (5) is that the intensity defined by equation (4) is constant and equal to $|\mathscr{E}_c|^2$. This property has enabled light scattering techniques to be used to investigate processes occurring on time scales as slow as seconds. On the other hand, a Gaussian fluctuating field is completely defined by the following statements (referring to equations (1) and (2))

$$\mathscr{E}^+(\mathbf{r}, t) = \sum_k \alpha_k e_k(\mathbf{r}, t) \tag{6}$$

$$p(\{\alpha_k\}) = \prod_k p(\alpha_k) \tag{7}$$

which implies that $$\langle|\alpha_k|^n|\alpha_{k'}|^m = S_{kk'}\langle|\alpha_{k'}|^n\rangle\langle|\alpha_k|^m\rangle \tag{8}$$

$$\langle\alpha_k\rangle = 0 \tag{9}$$

$$\langle|\alpha_k|^2\rangle = S(\omega_k) \tag{10}$$

where $S(\omega_k)$ is the optical or first order spectrum.

By a simple calculation the intensity fluctuation distribution is found to be a negative exponential

$$P(I) = \exp(-I/\langle I\rangle)/\langle I\rangle \tag{11}$$

where

$$\langle I \rangle = \int_0^\infty IP(I)\,\mathrm{d}I \tag{12}$$

the angle brackets denoting an average over an ensemble of samples or, alternatively for an ergodic field, a time average. The fact that only $S(\omega_k)$ is needed to characterize a Gaussian signal is a unique property and an arbitrary incoherent field in general requires the specification of higher order spectral properties.

Equations (10)–(12) follow from (6)–(10) but it is clear that measurement of $P(I)$, if it were possible, provides statistical information about $\mathscr{E}^+(\mathbf{r}, t)$ and not spectral information. The reason for this becomes clearer when it is recalled that the optical spectrum of a stationary field is just the Fourier transform of the auto-correlation function

$$g^{(1)}(t, t') = \langle \mathscr{E}^+(\mathbf{r}, t)\mathscr{E}^-(\mathbf{r}, t')\rangle / \langle I \rangle \tag{13}$$

which we have defined in normalized form. For stationary processes $g^{(1)}(t, t')$ is a function only of the time difference $\tau = t - t'$ and decays to zero from a value of unity at $\tau = 0$ with a characteristic time τ_c equal to the inverse spectral bandwidth. Evidently a measurement of the instantaneous intensity or its distribution $P(I)$ contains no information about two-time correlations in the signal and therefore no information about the spectrum. A simple way of obtaining spectral information is to take advantage of temporal integration which will be present in any case in a real detection system. Neglecting the effect of spatial integration, for small integration times the quantity E defined by equation (3) will reduce to I and in the Gaussian-field case would be distributed according to relations (11) and (12). However, for integration times which are long compared to τ_c, the correlation time of the light, the signal fluctuations are averaged out and E appears constant from sample to sample. The distribution $P(E)$ therefore changes from that shown in Fig. 2(a) to that in Fig. 2(b) as T is increased from values smaller than to values greater than τ_c. Measurements of $P(E)$ for a range of values of T will thus provide spectral information as well as statistical information about the signal.

It is frequently more convenient both experimentally and theoretically to study the moments of the intensity fluctuation distributions

$$n^{(r)} = \frac{\langle E^r \rangle}{\langle E \rangle^r} = \int_0^\infty E^r P(E)\,\mathrm{d}E / \langle E \rangle^r \tag{14}$$

In the small-sample-time small-detector-area limit measurements of these quantities can provide a simple check on the statistics of the source. For example

$$\lim_{A,T\to 0} n^{(r)} = 1 \qquad \text{coherent light (Glauber, 1965)} \tag{15}$$

$$= r! \qquad \text{thermal (Gaussian) light (Mandel, 1958)} \tag{16}$$

$$= (r!)^2 \qquad \text{Gaussian light scattered by a Gaussian medium (Bertolotti } et\ al.\text{, 1970)} \tag{17}$$

In order to obtain spectral information the effect of finite integration times must again be used. There has been a good deal of theoretical effort devoted to the evaluation of sample-time effects on $P(E)$ and its moments. A generating function

$$Q(s) = \langle e^{-sE} \rangle \tag{18}$$

is commonly employed. Only integrated laser light and the case of Gaussian statistics and Lorentzian spectral profile (Gaussian–Lorentzian light) characterized by a linewidth Γ, has been fully treated in the literature (Lax and Zwanziger, 1970, 1973; Jakeman *et al.*, 1970a; Bédard, 1966; Jakeman and Pike, 1968a) $P(E)$ is the inverse Laplace transform of (18) and in principle all the moments of the distribution can be derived from the generating function by differentiating at $s = 0$. In the case of Gaussian light the second moment of $P(E)$ can be calculated rather simply from the identity (Jakeman and Pike, 1969a) (neglecting spatial integration effects)

$$\frac{d^2(T^2 n^{(2)})}{dT^2} = 2(1 + |g^{(1)}(T)|^2) \tag{19}$$

$n^{(2)}$ is plotted against T for a Lorentzian line shape in Fig. 3 which is given by the relation

$$n^{(2)} = 1 + \frac{1}{\Gamma T} - \frac{1}{2(\Gamma T)^2} + \frac{1}{2(\Gamma T)^2} e^{-2\Gamma T}$$

We have so far neglected the effect of spatial integration due to finite aperture sizes in the optical system. In general, light from a perfectly incoherent quasi-monochromatic source of radius S attains a degree of coherence during propagation to the detector so that the electric field at two points on the detector surface separated by a distance R are in phase and interfere constructively if, roughly speaking (Mandel and Wolf, 1965)

$$R < \frac{Z}{k_0 S}$$

where k_0 is the wave vector of the light and Z the source-detector separation. This defines a coherence area at the detector, which if exceeded by the sensing area leads to averaging out of the signal fluctuations and consequently loss of information. This is compensated by the increased collecting power of large area detectors which may improve the accuracy of measurements in certain

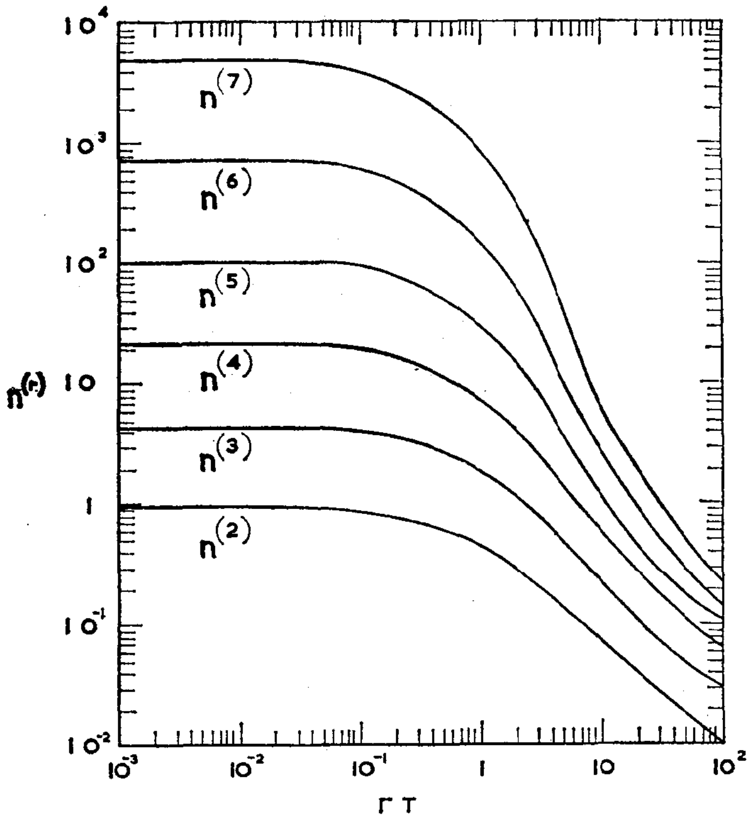

FIG. 3. Excess normalized factorial moments for Gaussian–Lorentzian light, after Bédard (1966).

circumstances. The spatial integration effect can, of course, be used to determine the source size (see for example Hanbury-Brown, 1968) but the increased complexity of the interpretation of results led early workers in the field of photon-counting spectroscopy to minimize this effect by choice of suitably small apertures in the optical system.

B. DOUBLE-TIME PROBABILITY DISTRIBUTIONS

The right-hand side of equation (19) may be expressed more generally in