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Theory of optical spectroscopy by digital autocorrelation of photon-counting fluctuations

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Abstract. Digital autocorrelation of optical signals is discussed with particular reference to the technique of clipping, reported briefly in an earlier publication. A detailed analysis of the relationship between the autocorrelation functions of clipped and unclipped photon-counting fluctuations is given. By evaluating the generating function for the joint distribution of intensity fluctuations, corrections to the earlier results are calculated which arise if the time over which the signal is sampled is not negligible compared with any coherence time of the light. The effect of clipping in heterodyne experiments is also analysed.

1. Introduction

Statistical techniques are finding application in an ever increasing number of problems in optical spectroscopy, ranging from turbulence measurements in fluids to the determination of diffusion constants of macromolecules. These problems necessitate the analysis of optical spectra with linewidths in the range 1–10^8 Hz. Conventional optical interference techniques become impractical for such measurements owing to the enormous paths (perhaps as much as 10^7 m) required to resolve small frequency differences in the optical region.

The fluctuating quantity observed in statistical experiments is the intensity or, more precisely, the square of the envelope of the field defined by

$$I(t) = \mathscr{E}^+(t)\mathscr{E}^-(t). \tag{1}$$

Here $\mathscr{E}^+(t)$ is the positive frequency part of the electric field corresponding quantum-mechanically to the annihilation of photons by the detection process. The space dependence of the field has been dropped since detection at a single space point will be assumed throughout the present work. The quantity $I(t)$ is modulated on a time scale determined by the spectral properties of the light field and these may be recovered by suitable measurement of its fluctuations. To this end, the techniques of intensity-fluctuation spectroscopy familiar to workers in the radar field (Atlas 1964) have in recent years been applied successfully at optical frequencies (for reviews see Benedek 1968, Cummins and Swinney 1969, Pike 1969). Such experiments have usually been carried out using a scanning electrical filter or spectrum analyser. Several other statistical methods can be used to extract spectral information from the fluctuations in $I(t)$, including the measurement of integrated photon-counting statistics (Jakeman *et al.* 1968). The relative merits of the various techniques are discussed by Foord *et al.* (1969), who conclude that direct autocorrelation of $I(t)$ in real time offers most of the advantages and few of the disadvantages of the other methods available.

When a high-intensity electric field is detected by a photomultiplier the output train of photoelectrons is essentially a continuous current. Although it is possible to digitalize artificially this current (as is sometimes done at microwave frequencies) an analogue autocorrelation technique seems more appropriate. In the case of light fields which give rise to rather low rates of photoelectron emission, however, analogue autocorrelation introduces an unnecessary dependence on fluctuations in the experimental apparatus and a digital technique, whereby the emissions of individual photoelectrons are correlated, is to be preferred.

The process of digital autocorrelation of optical intensities in real time is at first sight complex and the cost of instrumentation prohibitive owing to the rapidity with which multiplication and storage must be carried out. A rather similar problem confronted workers in the radar field many years ago, however, and was overcome by the technique of 'clipping'. This entails replacing the signal, before autocorrelation, by a series of ones and zeros according as it lies above or below a specified, though not necessarily constant, 'clipping' level. Autocorrelation of the clipped signal is a relatively simple matter and can in certain circumstances give information about the true autocorrelation function. For example, Van Vleck (1943, see also Van Vleck and Middleton 1966) has shown that if a Gaussian field with zero mean is replaced by one or zero according as it is positive or negative (an extreme form of clipping known as 'hard limiting') then the autocorrelation function of the clipped signal is $2/\pi$ times the arc sine of the original one. The technique takes advantage of the fact that any signal contains more information than is necessary for the construction of its first-order autocorrelation function. In practice, simplification of the autocorrelation process by clipping is obtained at the expense of a small increase in the number of samples (and hence the experimental time) required to attain the same statistical accuracy as a comparable direct autocorrelation.

Since it was originally formulated in connection with the measurement of field fluctuations in the microwave region of the spectrum, the method of clipping is not immediately applicable to intensity measurements at optical frequencies. By analogy, one possibility is to replace the intensities to be correlated (multiplied) by zero or one according as they lie below or above some predetermined level. However, the extraction of the true correlation function from such measurements is normally a difficult mathematical problem and a somewhat different technique—that of clipping in only one channel—proves to be more useful in practice. Carrying over such techniques into the digital analysis of trains of photon detections is a relatively straightforward step and some preliminary results, valid for Gaussian light of arbitrary spectral profile, were presented in an earlier publication (Jakeman and Pike 1969 b).

In the present paper the relationship between autocorrelation functions of clipped and unclipped photon-counting distributions is examined in more detail. Contrary to the assumption implicit in the previous work, photon-counting distributions can in practice be sampled over times which are not negligible compared with any coherence time of the light under investigation. The consequent modification of the earlier results may be calculated with the help of the generating function for the joint probability distribution of the integrated intensities and the present work is devoted in part to the evaluation of this quantity. The complexity of the problem necessitates the assumption of a simple model field, and in order to gain some insight into the size of corrections to the previous results they are calculated for the case of Gaussian–Lorentzian light.

A further problem tackled in the present paper is that of the clipped Doppler experiment. In many situations, particularly when the statistics of the signal are unknown, Doppler or heterodyne spectroscopy has proved to be a more useful technique for the measurement of field spectra than intensity-fluctuation spectroscopy. The digital autocorrelation of trains of photon detections in such an experiment might also be simplified by clipping the signal prior to multiplication, and new calculations will be presented which demonstrate the effect of this process. In order to obtain the clipped Doppler autocorrelation function it is necessary to evaluate the generating function of the joint probability distribution of intensities containing both incoherent and coherent components. Finite sample time effects will be neglected for the purposes of this latter calculation.

The next section is devoted to a general discussion of the intensity autocorrelation function, its connection with the field spectrum and with digital experiments of the integrated photon-counting statistics type. In § 3 the technique of clipping is defined mathematically and expressed in terms of photon-counting distributions and the related generating functions. These are calculated for certain special cases in § 4 and the autocorrelation functions of clipped signals are evaluated and discussed in the last section of the paper.

2. Analysis of intensity fluctuations by autocorrelation

This section is intended to serve mainly as a notational introduction to quantities of interest in the field of optical spectroscopy, although some results are presented which to the author's knowledge have not previously appeared in the literature.

For a wide class of optical fields a measurement of the intensity autocorrelation function is sufficient to determine completely the field autocorrelation function. For example, when the probability distributions of the Fourier amplitudes of the field are Gaussian the normalized second-order autocorrelation function $g^{(2)}(\tau)$ factorizes (Siegert 1943, Glauber 1963)

$$g^{(2)}(\tau) = \frac{\langle I(\tau)I(0)\rangle}{\langle I\rangle^2} = 1+|g^{(1)}(\tau)|^2 \tag{2}$$

where

$$g^{(1)}(\tau) = \frac{\langle \mathscr{E}^+(\tau)\mathscr{E}^-(0)\rangle}{\langle I\rangle} \tag{3}$$

is the normalized optical autocorrelation function. If the field spectrum is symmetric about some mean frequency, $g^{(1)}(\tau)$ is determined by a measurement of $g^{(2)}(\tau)$ through equation (2). Relations analogous to (2) may be derived for statistics other than Gaussian but if the statistics are unknown Doppler experiments are more useful. In such experiments the field of interest $\mathscr{E}_s^{\,+}(t)$ is mixed with the coherent beam from a strong local oscillator source of frequency ω:

$$\mathscr{E}^+(t) = \mathscr{E}_s^{\,+}(t)+I_c^{1/2}\exp(-\mathrm{i}\omega t). \tag{4}$$

Here I_c is the constant intensity of the coherent contribution to the field. If I_c is much larger than the average intensity $\langle I_s\rangle$ then the normalized intensity autocorrelation function is given by

$$g^{(2)}(\tau) = 1+\frac{\langle I_s\rangle}{\langle I_c\rangle}\{g^{(1)}(\tau)\exp(\mathrm{i}\omega\tau)+g^{(1)*}(\tau)\exp(-\mathrm{i}\omega\tau)\}+\mathrm{O}\left(\frac{\langle I_s\rangle}{\langle I_c\rangle}\right)^2 \tag{5}$$

and $g^{(1)}(\tau)$ can again be recovered from a measurement of $g^{(2)}(\tau)$.

A rather different technique by which intensity fluctuations may be analysed is that of photon-counting statistics (for a list of references see Jakeman and Pike 1969 a). In the simplest experiment a single probability distribution $p(n; T)$ of the number of counts n arriving in a sample of duration T of the scattered intensity is determined. This can be related to parameters of simple model fields (Jakeman and Pike 1968, 1969 a), though in practice its normalized factorial moments defined in terms of the mean number of counts $\bar{n}$ per sample by

$$n^{(r)}(T) = \sum_{n=0}^{\infty}\frac{n(n-1)\ldots(n-r+1)p(n;T)}{\bar{n}^r} \tag{6}$$

prove more useful for fitting purposes. This technique is only useful when the statistics of the light and the shape of the spectrum are of a known simple form. Even if the statistics were known many photon-counting distributions for different values of T would be needed if a completely unknown spectral profile was to be determined. A knowledge of this dependence on sample time is equivalent to a knowledge of the intensity autocorrelation function, however, as may be seen from the following considerations. It is not difficult to show, using Mandel's (1959) formula

$$p(n, T) = \frac{1}{n!}\int_0^\infty (\alpha E)^n \exp(-\alpha E)P(E)\, \mathrm{d}E \tag{7}$$

where α is the quantum efficiency of the detector and the integrated intensity is given by

$$E(\tau; T) = \int_{\tau-T/2}^{\tau+T/2} I(t)\, \mathrm{d}t \tag{8}$$

that the normalized factorial moments of $p(n; T)$ are identical with the normalized moments of the integrated intensity-fluctuation distribution $P(E)$. Thus

$$n^{(2)}(T) = \frac{1}{\langle E\rangle^2}\int_{-T/2}^{+T/2} \mathrm{d}t \int_{-T/2}^{+T/2} \mathrm{d}t\, \langle I(t)I(t')\rangle\, \mathrm{d}t\, \mathrm{d}t'. \tag{9}$$

Multiplying this equation by T^2 (recalling that $\langle E\rangle = \langle I\rangle T$) and differentiating twice with respect to the sample time leads to the formula

$$\frac{\mathrm{d}^2}{\mathrm{d}T^2}\{T^2 n^{(2)}(T)\} = g^{(2)}(T) + g^{(2)}(-T). \tag{10}$$

Turning now to the direct intensity autocorrelation experiment by which $g^{(2)}(\tau)$ is constructed in real time, it is convenient to introduce a modified function to take account of the fact that sampling is never instantaneous, but characterized by a finite time interval T. This will be defined as follows:

$$g^{(2)}(\tau; T) = \frac{\langle E(\tau; T)E(0; T)\rangle}{\langle E\rangle^2} \tag{11}$$

where $E(\tau; T)$ is defined by equation (8) and

$$g^{(2)}(\tau; 0) \equiv g^{(2)}(\tau). \tag{12}$$

The autocorrelation function of the integrated intensity may be related to that of the instantaneous intensity by differentiating equation (11) with respect to the sample time. For non-overlapping samples,

$$\frac{\mathrm{d}^2}{\mathrm{d}T^2}\{T^2 g^{(2)}(\tau; T)\} = g^{(2)}(\tau+T; 0) + g^{(2)}(\tau-T; 0), \qquad \tau \geqslant T. \tag{13}$$

As an example, consider the case of Gaussian–Lorentzian light

$$g^{(2)}(\tau; 0) = 1 + \exp(-2\Gamma|\tau|). \tag{14}$$

The correction to the intensity autocorrelation function due to finite sampling is obtained by substituting (14) into (13) and integrating. Applying the boundary condition (14) at $T = 0$ then gives (Pike 1969)

$$g^{(2)}(\tau; T) = 1 + \exp(-2\Gamma\tau)\left(\frac{\sinh \Gamma T}{\Gamma T}\right)^2, \qquad \tau \geqslant T. \tag{15}$$

Although the discussion and results presented in the preceding part of this section are confined explicitly to the properties of the intensity autocorrelation function, they apply equally well to the autocorrelation function of photon-counting distributions since these two quantities are identical except for zero time difference, when there is a statistical correction. These properties may be demonstrated using the following generalization of equation (7):

$$p(n, n'; T) = \int_0^\infty \mathrm{d}E \int_0^\infty \mathrm{d}E' \exp\{-\alpha(E+E')\} \frac{(\alpha E)^n}{n!} \frac{(\alpha E')^{n'}}{n'!} P(E, E') \qquad (16)$$

where $p(n, n'; T)$ is the probability of counting $n(t; T)$ photoelectrons in the sample time T at time t and $n'(t'; T)$ photoelectrons in the same sample time at time t'. $P(E, E')$ is the corresponding distribution of integrated intensities. Using (16) it is not difficult to establish the relation

$$g^{(2)}(\tau; T) = \frac{\langle n(\tau; T) n(0; T)\rangle}{\bar{n}^2}, \qquad \tau \geqslant T \qquad (17)$$

where $\bar{n} = \alpha\langle E\rangle$. At zero time difference the quantity on the left-hand side of (17) satisfies the previously mentioned relationship between the factorial moments of $p(n; T)$ and the moments of $P(E)$, namely

$$g^{(2)}(0; T) = \frac{\langle n(0; T) n(0; T)\rangle - \langle n(0; T)\rangle}{\bar{n}^2} = n^{(2)}(T). \qquad (18)$$

3. Autocorrelation of clipped intensities

Earlier theoretical work has been concerned with the analysis of continuous signals at microwave frequencies and it is convenient to extend this work into the optical region by first considering the possibilities of intensity clipping. By straightforward analogy with the earlier work (for example Van Vleck and Middleton 1966) a normalized autocorrelation function of clipped integrated intensities may be defined as follows:

$$G_{bb}^{(2)}(\tau; T) = \int_b^\infty \mathrm{d}E \int_b^\infty \mathrm{d}E' \, P(E, E'). \qquad (19)$$

This definition describes the autocorrelation of intensities which are set equal to zero if they are less than b and equal to 1 if they are greater than that value. The double subscript on the left-hand side is used since, in principle, clipping could be carried out at different levels in each channel. In practice the most commonly occurring and useful form for $P(E, E')$ is that given by Siegert (1943) for Gaussian light in the limit of small sample times. The integral on the right-hand side of (19) cannot be performed analytically for this function, however, and $G_{bb}^{(2)}(\tau; T)$ can only be expressed as a complicated sum of terms containing the true autocorrelation function. The latter cannot be extracted from a measurement of the former quantity, therefore, without a good deal of computation. Fortunately, the mathematical situation is greatly improved if only one channel is clipped and a single clipped autocorrelation function may be defined which proves to be of much more practical use:

$$G_b^{(2)}(\tau; T) = \int_0^\infty E' \, \mathrm{d}E' \int_b^\infty \mathrm{d}E \, P(E, E'). \qquad (20)$$

It is a relatively small step, now, to define corresponding autocorrelation functions of clipped photon-counting distributions with which the remainder of this paper will be concerned. Appropriately normalized and with integer subscripts kk' they may be written in the following form:

$$g_{kk'}^{(2)}(\tau; T) = \frac{\sum_{n>k}^{\infty} \sum_{n'>k'}^{\infty} p(n, n'; T)}{\sum_{n>k}^{\infty} p(n; T) \sum_{n>k'}^{\infty} p(n; T)} \tag{21}$$

$$g_k^{(2)}(\tau; T) = \frac{\sum_{n>k}^{\infty} \sum_{n'=1}^{\infty} n' p(n, n'; T)}{\bar{n} \sum_{n>k}^{\infty} p(n; T)}. \tag{22}$$

The right-hand sides of equations (21) and (22) are conveniently expressed in terms of the generating function of the joint integrated intensity-fluctuation distribution

$$Q(s, s'; T) = \langle \exp\{-(Es + E's')\} \rangle \tag{23}$$

through the relation

$$p(n, n'; T) = \frac{(-\alpha)^n}{n!} \frac{(-\alpha)^{n'}}{n'!} \frac{d^n}{ds^n} \frac{d^{n'}}{ds^{n'}} Q(s, s'; T)|_{s=s'=\alpha} \tag{24}$$

where, as before, α is the quantum efficiency of the detector and n, n' denote $n(t; T)$, $n'(t'; T)$, the number of counts arriving in the interval T at times t and t' respectively. In the earlier paper (Jakeman and Pike 1969 b) $g_{00}^{(2)}(\tau; 0)$ and $g_k^{(2)}(\tau; 0)$ were evaluated in this way. A finite sample time calculation is in general much more difficult, however, and will be restricted in the present paper to the case of Gaussian–Lorentzian light.

4. Generating functions

The first part of this section is devoted to an evaluation of $Q(s, s'; T)$ defined by (23) for Gaussian–Lorentzian light. This will allow the autocorrelation functions defined by (21) and (22) to be determined using equation (24). Since the effect of clipping in Doppler experiments is also of interest, the generating function $Q_D(s, s'; 0)$ for a mixture of Gaussian light of arbitrary spectral profile and a single coherent mode will be derived in the second part of the section.

4.1. *Intensity-fluctuation experiment*, $T \neq 0$

The mathematical techniques employed to find $Q(s, s'; T)$ are analogous to those used by Meyer and Middleton (1954) and more recently by Dialetis (1969) and represent a natural generalization of the approach used to obtain $Q(s; T)$ in earlier papers (see, for example, Jakeman and Pike 1968). It is shown in appendix 1 that, for fields whose Fourier amplitudes have Gaussian probability distributions, (23) is given by

$$Q(s, s'; T) = \prod_k \frac{1}{1 + (\langle E \rangle / T)\lambda_k(s, s')} \tag{25}$$

where the dependence of λ_k on s and s' is determined through the integral equation

$$\left(s \int_{\tau - T/2}^{\tau + T/2} + s' \int_{-T/2}^{+T/2}\right) g^{(1)}(t - t')\phi_k(t')\, dt' = \lambda_k \phi_k(t) \tag{26}$$

and $g^{(1)}(t)$ is defined by equation (3). For a Lorentzian spectrum centred at ω_0,

$$g_{\text{Lor}}^{(1)}(t-t') = \exp\{-\Gamma(t-t')+\mathrm{i}\omega_0(t-t')\} \tag{27}$$

the integral equation (26) can be solved exactly and following the method given in appendix 1 it may be shown that the eigenvalues are given through the relation

$$\lambda_k = -\frac{T}{\langle E\rangle \xi_k} \tag{28}$$

in terms of the roots ξ_k of the transcendental equation

$$F(\xi) = 0 \tag{29}$$

where

$$F(\xi) = \exp(-2\gamma)\left\{\cosh y + \frac{1}{2}\left(\frac{\gamma}{y}+\frac{y}{\gamma}\right)\sinh y\right\}\left\{\cosh y' + \frac{1}{2}\left(\frac{\gamma}{y'}+\frac{y'}{\gamma}\right)\sinh y'\right\}$$
$$-\tfrac{1}{4}|g_{\text{Lor}}^{(1)}(\tau)|^2\left(\frac{\gamma}{y}-\frac{y}{\gamma}\right)\left(\frac{\gamma}{y'}-\frac{y'}{\gamma}\right)\sinh y \sinh y' \tag{30}$$

$$\gamma = \Gamma T \tag{31}$$

$$y^2 = \gamma^2 + 2\gamma s\langle E\rangle\xi \tag{32}$$

and

$$y'^2 = \gamma^2 + 2\gamma s'\langle E\rangle\xi. \tag{33}$$

Now $F(\xi)$ is an entire function of ξ of order $\frac{1}{2}$ and genus zero, so that applying Hadamard's factorization theorem (Ahlfors 1966, p. 207) it may be expressed as an infinite product over its zeros. Since $F(0)$ is unity this takes the form

$$F(\xi) = \prod_k \left(1-\frac{\xi}{\xi_k}\right) = \prod_k \left(1+\frac{\xi\langle E\rangle}{T}\lambda_k\right). \tag{34}$$

Comparison of (25) and (34) gives the desired closed form for the generating function

$$Q_{\text{Lor}}(s, s'; T) = \frac{1}{F(1)}. \tag{35}$$

The generating function for the single integrated intensity-fluctuation distribution of Gaussian–Lorentzian light (Bédard 1966, Jakeman and Pike 1968) may be obtained by setting s' equal to zero in this equation.

4.2. *Doppler experiment*, $T = 0$

Consider now a field consisting of a single coherent mode of amplitude β displaced to a frequency ω from the centre of the spectrum (assumed symmetric) of a Gaussian component (Jakeman and Pike 1969 a). The generating function

$$Q_{\text{D}}(s, s'; 0) = \langle\exp\{-(sI+s'I')\}\rangle \tag{36}$$

of the joint intensity-fluctuation distribution of such a field in the small sample time limit is conveniently obtained from the moment generating function of the fourfold probability distribution $P\{\mathscr{E}^+(t), \mathscr{E}^-(t), \mathscr{E}^+(t'), \mathscr{E}^-(t)\}$. This generating function takes the rather simple form

$$M_{\text{D}}(s_1, s_2, s_1', s_2'; 0) = \exp[-\{s_1\beta\exp(\mathrm{i}\omega t)+s_2\beta^*\exp(-\mathrm{i}\omega t)$$
$$+s_1'\beta\exp(\mathrm{i}\omega t')+s_2'\beta^*\exp(-\mathrm{i}\omega t')\}]$$
$$\times \exp[\langle E\rangle\{(s_1s_2+s_1's_2')g^{(1)}(0)+(s_1s_2'+s_2s_1')|g^{(1)}(\tau)|\}]. \tag{37}$$

By expressing the generating function $Q(s, s'; T)$ in terms of angular integrations of the fourfold field probability distribution it may be shown quite generally that

$$Q(s, s'; T) = -\frac{1}{4\pi^2 ss'} \int\int_{-\infty}^{+\infty} \int\int ds_1\, ds_2\, ds_1'\, ds_2' \exp\left(\frac{s_1 s_2}{s} + \frac{s_1' s_2'}{s'}\right) \times M(s_1, s_2, s_1', s_2'; T). \tag{38}$$

Substituting from (37) into (38) leads to the result

$$Q_D(s, s'; 0) = \exp(-\langle W\rangle Q(s, s'; 0)[s+s'+2ss'\langle E\rangle\{g^{(1)}(0) - |g^{(1)}(\tau)| \cos \omega\tau\}]) \times Q(s, s'; 0) \tag{39}$$

where

$$Q(s, s'; 0) = \{(1+\langle E\rangle s)(1+\langle E\rangle s') - \langle E\rangle^2 ss' |g^{(1)}(\tau)|^2\}^{-1} \tag{40}$$

is the generating function of the joint intensity-fluctuation distribution $P(I, I')$ given in the earlier paper (Jakeman and Pike 1969 b), and $\langle W\rangle$ $(= |\beta|^2)$ is the intensity of the coherent mode.

5. Results and discussion

In order to assess the order of magnitude of the corrections to previous results, which arise from sample times which are finite but small compared with the coherence time of the signal being analysed, it is interesting as a first approximation to consider an expansion of (35) to first order in γ:

$$Q_{\text{Lor}}(s, s'; T) \simeq Q_{\text{Lor}}(s, s'; 0)\left(1 - \frac{\gamma}{3}\langle E\rangle^2 Q_{\text{Lor}}(s, s'; 0) \times [s^2 + s'^2 + \langle E\rangle ss'(s+s')\{1 - |g_{\text{Lor}}^{(1)}(\tau)|^2\}]\right) \tag{41}$$

where $Q_{\text{Lor}}(s, s'; 0)$ is obtained by setting $T = 0$ in (35) and is identical with (40) when $g^{(1)}(\tau)$ is replaced by $g_{\text{Lor}}^{(1)}(\tau)$. The required correction to the autocorrelation function of the clipped signal can be calculated to first order in the sample time T using the definitions (21), (22) and (24) and the approximate expression for the generating function (41).

The exact autocorrelation function generated by clipping intensities in both channels at zero is of the form

$$g_{00}^{(2)}(\tau; T) = \frac{1 - 2Q(\alpha; T) + Q(\alpha, \alpha; T)}{\{1 - Q(\alpha; T)\}^2} \tag{42}$$

where $Q(s; T) \equiv Q(s, 0; T)$ is the generating function corresponding to $P(E)$. Clipping at higher levels in both channels gives rise to increasingly complicated formulae. For a Lorentzian spectrum and small γ (42) may be evaluated with the help of (44) to give the approximate result

$$g_{00}^{(2)}(\tau; T) = \frac{1 + \{(1-\bar{n})/(1+\bar{n})\}|g_{\text{Lor}}^{(1)}(\tau)|^2}{1 - \{\bar{n}/(1+\bar{n})\}^2 |g_{\text{Lor}}^{(1)}(\tau)|^2} - \frac{2\gamma}{3}\frac{\bar{n}^2 |g_{\text{Lor}}^{(1)}(\tau)|^2}{(1+\bar{n})^3} \times \frac{1 - \{\bar{n}/(1+\bar{n})\}|g_{\text{Lor}}^{(1)}(\tau)|^2}{[1 - \{\bar{n}/(1+\bar{n})\}^2 |g_{\text{Lor}}^{(1)}(\tau)|^2]^2}. \tag{43}$$

Calculation of the correction in the single clipped case is not quite so straightforward though the final formula is rather simple. In appendix 2 it is shown that this is

$$g_k^{(2)}(\tau;\, T) = 1 + |g_{\text{Lor}}^{(1)}(\tau)|^2 \left\{\frac{1+k}{1+\bar{n}} + \frac{\gamma}{3}\frac{(2k\bar{n}+k-\bar{n}^2)}{(1+\bar{n})^2}\right\}. \tag{44}$$

If γ is set equal to zero in (43) and (44), a special case (i.e. Lorentzian spectral profile) of the results presented in the earlier paper (Jakeman and Pike 1969 b) is recovered. For small values of $\bar{n}$ the right-hand side of (43) then reduces to (2) and only statistical accuracy is lost by clipping. On the other hand, for large $\bar{n}$ all spectral information is lost by clipping at zero as might be expected. Similarly when $T = 0$ (44) reduces to (2) when $k \sim \bar{n}$ and little spectral information is lost by making this choice of clipping level whatever the value of $\bar{n}$.

Relations (43) and (44) reduce to the expected results when $\bar{n}$, $k \ll 1$. The correction term then vanishes since clipping has little effect on the signal and $g^{(2)}(\tau;\, T)$ is an even function of γ from (15). For clipping at the mean (44) reduces (for integer $\bar{n}$) to

$$g_{\bar{n}}^{(2)}(\tau;\, T) = 1 + |g_{\text{Lor}}^{(1)}(\tau)|^2 + \frac{\gamma}{3}\frac{\bar{n}}{1+\bar{n}}|g_{\text{Lor}}^{(1)}(\tau)|^2. \tag{45}$$

It is interesting to compare (45) with an expansion of (15) in powers of γ^2:

$$g^{(2)}(\tau;\, T) = 1 + |g_{\text{Lor}}^{(1)}(\tau)|^2 + \frac{\gamma^2}{3}|g_{\text{Lor}}^{(1)}(\tau)|^2 + O(\gamma^4). \tag{46}$$

Whereas for $\gamma = 0{\cdot}1$ an error of $0{\cdot}3\%$ would be made in the coefficient of $|g_{\text{Lor}}^{(1)}(\tau)|^2$

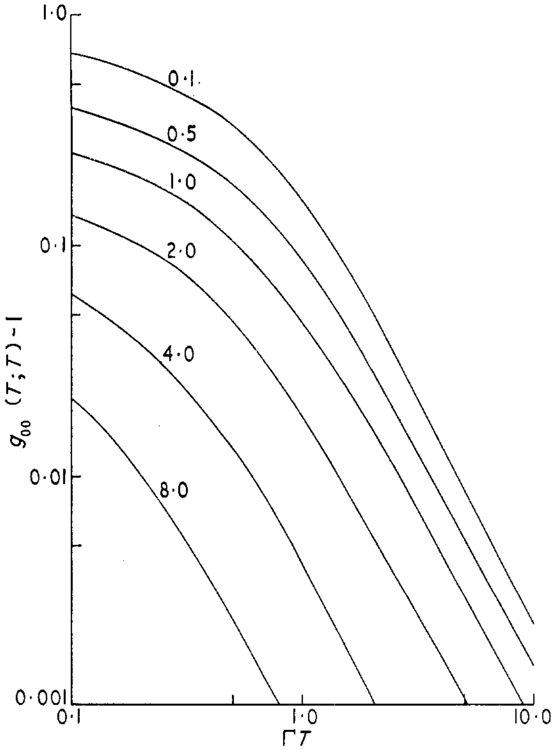

Figure 1. Maximum decay of the intensity autocorrelation function of Gaussian–Lorentzian light when clipping at zero is carried out in both channels. The values of $\bar{n}$ are shown against the curves.

by neglecting sample-time effects in a full autocorrelation, an error of 1·7% would be made in the autocorrelation of the clipped signal for $\bar{n} = 1$, rising to 3·3% for $\bar{n} \gg 1$. Comparison of (43) and (46) is not so instructive since for zero sample time the two autocorrelation functions are not identical unless $\bar{n}$ is vanishingly small. If the latter condition is satisfied the correlation vanishes to first order in γ since, as mentioned above, clipping at zero then has little effect on the signal. If $\bar{n}$ is not small the correction factor is a function of the correlation time τ.

The effect of arbitrary sample times (i.e. γ not small) on the autocorrelation function of a clipped Gaussian–Lorentzian signal can be ascertained using the exact form of $Q_{\mathrm{Lor}}(s, s'; T)$ given by equations (30) and (35). Figures 1 and 2 show the

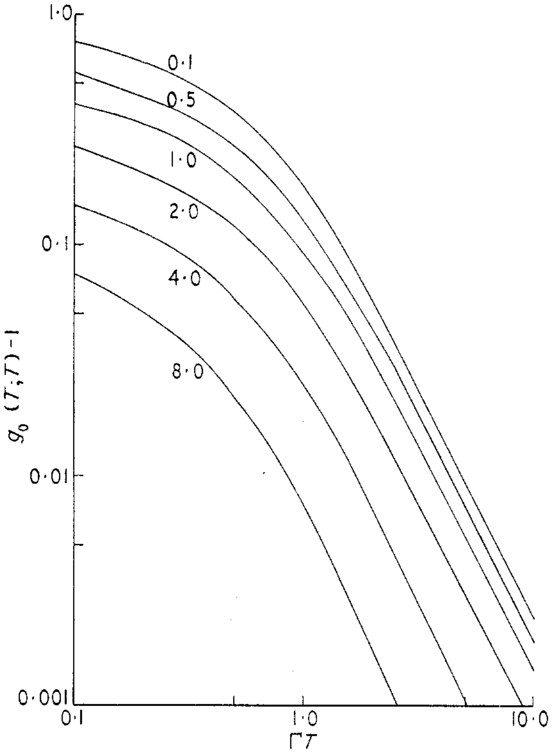

Figure 2. Maximum decay of the intensity autocorrelation function of Gaussian–Lorentzian light when clipping at zero is carried out in one channel. The values of $\bar{n}$ are shown against the curves.

results of such calculations for various $\bar{n}$ when the clipping levels are set at zero. Since the largest effect occurs for small correlation times, the graphs are plotted for $\tau = T$, i.e. the smallest value of τ for which sample times do not overlap. The quantity plotted is $g^{(2)}(T; T)-1$ and in fact represents the overall maximum decay of the autocorrelation function which can be measured experimentally; it is thus an indication of the sensitivity decrease with sample time. For values of $\gamma \gg 1$ it decreases like $1/\gamma^2$. For $\gamma \ll 1$ the correction terms in (43) and (44) have a small effect by comparison with that of the condition $\tau \geq T$, since in the case of a Lorentzian spectrum this restriction reduces the sensitivity exponentially for small γ. For example, setting $\Gamma\tau = 0{\cdot}1$ in (45) with $T = 0$ yields a value of 0·8 for the $|g_{\mathrm{Lor}}{}^{(1)}(\tau)|^2$ factor as compared with unity when $\tau = 0$—a reduction of 20%. When clipping is carried out in one channel at the mean the additional correction which appears in (45) tends to diminish this fall in sensitivity slightly, but when clipping is carried out in both channels at zero the effect is enhanced by the additional correction present in equation (43). The size of the term proportional to γ in equation (44) can in fact be reduced by suitably adjusting the values of k and $\bar{n}$ but this tends to reduce the sensitivity of the technique.

The effect of clipping on Doppler-type experiments is more complicated, as can be judged from the form of the generating function (39). However, expressions for the clipped autocorrelation functions can in certain cases be obtained using the definitions (21), (22) and (24) as before. For example, clipping at zero in both channels gives rise to the autocorrelation function

$$g_{00}^{(2)}(\tau;0) = 1+\frac{\exp[\{2\bar{n}_i\bar{n}_c|g^{(1)}(\tau)|\cos\omega\tau\}/(1+\bar{n}_i)^2]+\{\bar{n}_i/(1+\bar{n}_i)\}^2|g^{(1)}(\tau)|^2-1}{[1-\{\bar{n}_i/(1+\bar{n}_i)\}^2|g^{(1)}(\tau)|^2][(1+\bar{n}_i)\exp\{\bar{n}_c/(1+\bar{n}_i)\}-1]^2} \tag{47}$$

where $\bar{n}_i = \alpha\langle E\rangle$ and $\bar{n}_c = \alpha\langle W\rangle$. Clipping in one channel at an arbitrary level k leads to a more complicated result, but when $\bar{n}_c \gg \bar{n}_i$ the approximate formula

$$\begin{aligned}(-\alpha)^m \frac{d^m}{ds'^m}\frac{dQ}{ds}\bigg|_{\substack{s=0\\ s'=\alpha}} \simeq{}& \frac{\bar{n}_c^{\,m+1}}{(1+\bar{n}_i)^{2m+3}}\\ &\times\exp\left(-\frac{\bar{n}_c}{1+\bar{n}_i}\right)\{2n_i|g^{(1)}(\tau)|\cos\omega\tau(1+\bar{n})-\bar{n}_i^2|g^{(1)}(\tau)|^2\\ &-(1+\bar{n}_i)^2\}\end{aligned} \tag{48}$$

may be used to obtain the following result

$$g_k^{(2)}(\tau;0) = 1-\frac{[\{n_i/(1+\bar{n}_i)\}|g^{(1)}(\tau)|^2-2|g^{(1)}(\tau)|\cos\omega\tau]\{n_i/(1+\bar{n}_i)\}}{[k!(1+\bar{n}_i)^{2k+1}/\bar{n}_c^{\,k}\exp\{\bar{n}_c/(1+\bar{n}_i)\}-1]}. \tag{49}$$

If clipping is carried out at zero in one channel it is possible to obtain a fairly simple exact formula. This is

$$g_0^{(2)}(\tau;0) = 1+\frac{\bar{n}_i\bar{n}_c|g^{(1)}(\tau)|}{(1+\bar{n}_i)(\bar{n}_i+\bar{n}_c)}\,\frac{2\cos\omega\tau+\bar{n}_i|g^{(1)}(\tau)|\{1/\bar{n}_c-1/(1+\bar{n}_i)\}}{(1+\bar{n}_i)\exp\{\bar{n}_c/(1+\bar{n}_i)\}-1}. \tag{50}$$

In the limit $\bar{n}_c \gg \bar{n}_i$ this reduces to (49) with $k = 0$.

These results indicate that the use of clipping techniques in a Doppler type of experiment is feasible in that the true autocorrelation function could be extracted from that of the clipped signal. However, one of the main advantages of the usual Doppler experiment, namely its independence of the statistics of the scattered light, is lost by clipping at a fixed mean because the autocorrelation function of the clipped signal is in this case a function of the higher-order natural correlation functions and therefore depends on their factorization properties. The use of a statistical distribution of clipping levels to overcome this difficulty is a possibility worth considering.

Several more general conclusions may be drawn from the calculations of this paper. It is evident that, at least in the case of Gaussian light, clipping in one channel rather than two may prove more useful in practice owing to the ease with which the true autocorrelation function can be recovered from that of the single clipped signal. A suitable experimental arrangement is described by Foord *et al.* (1969) (figure 3) who have measured the diffusion constant of protein molecules using this technique. It is also advantageous both from the point of view of experimental sensitivity and theoretical simplicity to minimize finite sampling time effects. Only when these are small is it possible, in general, to recover an arbitrary spectral profile from the clipped autocorrelation function. The obvious way to achieve this is to ensure that the sampling time is small compared with any coherence time of the light under investigation, but

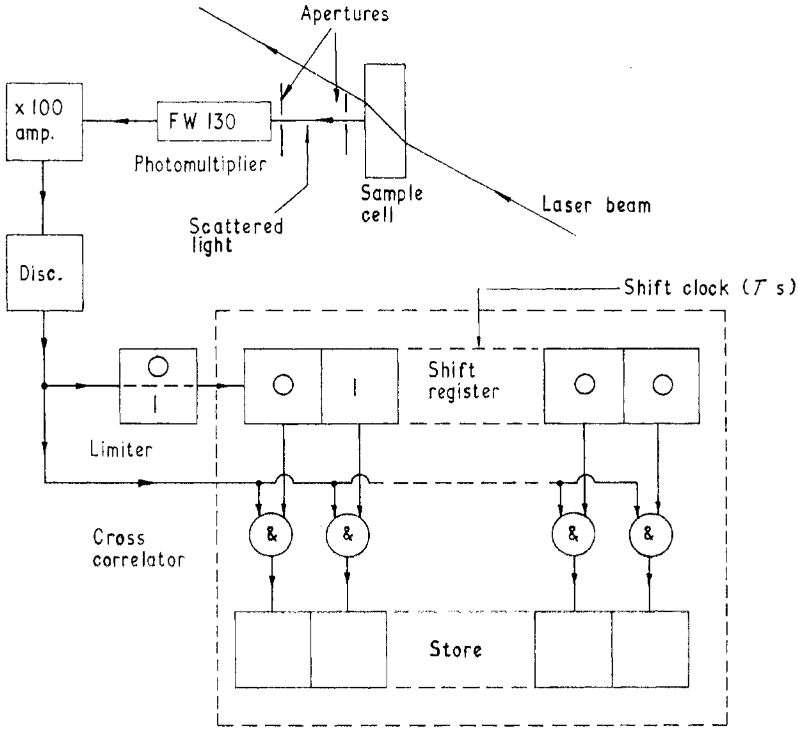

Figure 3. Block diagram of the use of a single-clipping autocorrelator after Foord *et al.* (1969). The clipped signal passing through the shift register in real time controls '&' gates which allow storage of the true signal when the clipped value is unity. The quantity $\langle n_k(0)n(\tau)\rangle$ accumulates in the store.

working with low values of $\bar{n}$ and low clipping levels also seems to be beneficial in this respect. The latter conditions are also advantageous in Doppler experiments because the effect of clipping signals prior to autocorrelation is thereby minimized and the particular merits of such experiments, mentioned above, are not lost by clipping. Earlier in this section the maximum measurable decay of the autocorrelation function was taken to be an indication of the sensitivity of the technique. It is not the only factor affecting the sensitivity, however, as inspection of equation (44) demonstrates. The maximum decay of the single clipped autocorrelation function given by this formula is seen to be $(1+k)/(1+\bar{n})$, neglecting finite sample time effects, a quantity which can be increased indefinitely by raising the clipping level k. The sensitivity of the technique is not necessarily increased by this process because it is also a function of the statistical accuracy of the measurement. As the clipping level is raised, the frequency of non-zero correlation for a given $\bar{n}$ is decreased so that, unless the number of samples of the signal (and hence the total experiment time) is increased, the statistical accuracy of the measurement is lowered. In this context it is, of course, of great interest to know exactly how much statistical accuracy is lost by clipping prior to autocorrelation. This and other problems related to the experimental errors incurred during digital autocorrelation of optical signals will be discussed in a future publication.

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Appendix 1. Evaluation of $Q(s, s', T)$

Using the notation of Jakeman and Pike (1968) the joint generating function

$$Q(s, s'; T) = \langle \exp(-sE - s'E') \rangle \tag{A1}$$

where

$$sE + s'E' = s \int_{\tau - T/2}^{\tau + T/2} I(t)\, dt + s' \int_{-T/2}^{T/2} I(t)\, dt \tag{A2}$$

may be expressed in terms of the normal modes α_k of the electric field defined by

$$\mathscr{E}^+(r, t) = \sum_k \alpha_k e_k(r, t) \tag{A3}$$

where

$$e_k(r, t) = i(\tfrac{1}{2}\hbar\omega_k)^{1/2} u_k(r) \exp(-i\omega_k t). \tag{A4}$$

For Gaussian light the probability distribution of the α_k is

$$p(\alpha_k) = \frac{1}{\pi \langle n_k \rangle} \exp\left(-\frac{|\alpha_k|^2}{\langle n_k \rangle}\right) \tag{A5}$$

with

$$\langle \alpha_i^* \alpha_j \rangle = \langle n_i \rangle \delta_{ij}. \tag{A6}$$

The transformation

$$e_k(t) = \sum_{k'} S_{kk'} \phi_{k'}(t) \tag{A7}$$

with the normalization

$$\left(s \int_{\tau - T/2}^{\tau + T/2} + s' \int_{-T/2}^{+T/2}\right) \phi_k(t) \phi_{k'}^*(t)\, dt = \delta_{kk'} \tag{A8}$$

leads immediately to the result

$$sE + sE' = \sum_k |a_k|^2 \tag{A9}$$

where

$$a_k = \sum_{k'} \alpha_k S_{kk'}. \tag{A10}$$

Now

$$\langle \mathscr{E}^+(t) \mathscr{E}^-(t') \rangle = \sum_{k'k''} \langle a_{k'} a_{k''}^* \rangle \phi_{k'}(t) \phi_{k''}^*(t') \tag{A11}$$

so that, if the a_k's are now required to be statistically independent,

$$\langle a_i a_j^* \rangle = \langle m_i \rangle \delta_{ij} \tag{A12}$$

with

$$p(a_k) = \frac{\exp(-|a_k|^2 / \langle m_k \rangle)}{\pi \langle m_k \rangle} \tag{A13}$$

multiplication of (A11) by $\phi_k(t')$ and integrating over t' using (A8) leads to the integral equation

$$\left(s \int_{\tau - T/2}^{\tau + T/2} + s' \int_{-T/2}^{+T/2}\right) \langle \mathscr{E}^+(t) \mathscr{E}^-(t') \rangle \phi_k(t')\, dt' = \langle m_k \rangle \phi_k(t). \tag{A14}$$

Equations (A1), (A9), (A12) and (A13) give the generating function in the form of an infinite product over the eigenvalues of (A14):

$$Q(s, s', T) = \prod_k \frac{1}{1+\langle m_k \rangle}. \tag{A15}$$

(A15) and (A14) reduce to (25) and (26) when (A14) is normalized using the relation

$$\langle \mathscr{E}^+(t)\mathscr{E}^-(t) \rangle = \frac{\langle E \rangle}{T} \tag{A16}$$

and the quantity

$$\lambda_k = \frac{T}{\langle E \rangle} \langle m_k \rangle. \tag{A17}$$

In solving (A14) for a Lorentzian spectrum (equation (27)) only the form of $\phi_k(t)$ inside the two sample times centred on 0 and τ need be considered. There are two cases assuming $\tau < 0$.

(i) $\tau - T/2 < t < \tau + T/2$

$$s\int_{\tau-T/2}^{t} \exp\{-\Gamma(t-t')\}\phi_k(t')\,\mathrm{d}t' + s\int_{t}^{\tau+T/2} \exp\{-\Gamma(t'-t)\}\phi_k(t')\,\mathrm{d}t' + s'\int_{-T/2}^{+T/2} \exp\{-\Gamma(t'-t)\}\phi_k(t')\,\mathrm{d}t' = \lambda_k\phi_k(t). \tag{A18}$$

(ii) $-T/2 < t < +T/2$

$$s\int_{\tau-T/2}^{\tau+T/2} \exp\{-\Gamma(t-t')\}\phi_k(t')\,\mathrm{d}t' + s'\int_{-T/2}^{t} \exp\{-\Gamma(t-t')\}\phi_k(t')\,\mathrm{d}t' + s'\int_{t}^{+T/2} \exp\{-\Gamma(t'-t)\}\phi_k(t')\,\mathrm{d}t' = \lambda_k\phi_k(t). \tag{A19}$$

These integral equations may be solved by differentiating twice with respect to t. This leads to differential equations with solutions of the form

$$\phi_k(t) = A\cos\omega t + B\sin\omega t. \tag{A20}$$

In case (i) above, $\omega^2 = (2\Gamma s - \Gamma^2\lambda)/\lambda$ and, in case (ii), $\omega^2 = (2\Gamma s' - \Gamma^2\lambda)/\lambda$. Substituting back into the integral equations leads to a criterion for non-trivial solutions which takes the form given in the text by equations (28)–(33).

Appendix 2. Evaluation of $g_k^{(2)}(\tau; T)$

The autocorrelation function of the single clipped signal may be written

$$g_k^{(2)}(\tau; T) = \frac{\bar{n} + \sum_{m=0}^{k} \{(-\alpha)^m/m!\}(\mathrm{d}^m/\mathrm{d}s^m)(\mathrm{d}/\mathrm{d}s')Q(s, s'; T)\Big|_{\substack{s'=0\\ s=\alpha}}}{\bar{n}\left[1 - \sum_{m=0}^{k} \{(-\alpha)^m/m!\}(\mathrm{d}/\mathrm{d}s^m)Q(s; T)\right]} \tag{A21}$$

where, to first order in γ, $Q(s, s'; T)$ is given for Gaussian–Lorentzian light by (41) and $Q(s; T)$ is obtained by setting s' to zero in the same equation. (A21) can be evaluated with the help of the formula (derived from equation (41))

$$\frac{(-\alpha)^m}{m!}\frac{d^m}{ds^m}\frac{dQ}{ds'}(s,s';T)\bigg|_{\substack{s=\alpha\\s'=0}} = \frac{\gamma\bar{n}}{3}(1-g^2)\delta_{m0}+\left(\frac{\bar{n}}{1+\bar{n}}\right)^{m+1}\left[g^2-1-g^2\frac{1+m}{1+\bar{n}}\right.$$
$$\left.+\frac{\gamma}{3}\left\{4g^2-2+(1-5g^2)\frac{1+m}{1+\bar{n}}+g^2\frac{(1+m)(2+m)}{(1+\bar{n})^2}\right\}\right] \qquad \text{(A22)}$$

where $g\ (\equiv |g_{\mathrm{Lor}}{}^{(1)}(\tau)|)$ is given by equation (27).

The numerator of (A21) takes the form

$$\bar{n}\left(\frac{\bar{n}}{1+\bar{n}}\right)^{k+1}\left[1+g^2\frac{1+k}{1+\bar{n}}+\frac{\gamma}{3}\left\{\frac{\bar{n}-k}{1+\bar{n}}+g^2\frac{(\bar{n}+3k\bar{n}-k^2-\bar{n}^2)}{(1+\bar{n})^2}\right\}\right] \qquad \text{(A23)}$$

whilst the denominator is given by

$$\bar{n}\left(\frac{\bar{n}}{1+\bar{n}}\right)^{k+1}\left\{1+\frac{\gamma}{3}\,\frac{\bar{n}-k}{1+\bar{n}}\right\}. \qquad \text{(A24)}$$

Expansion of the ratio of (A23) to (A24) to first order in γ then leads to equation (44) of the text.

References

AHLFORS, L. V., 1966, *Complex Analysis* (New York: McGraw-Hill).

ATLAS, D., 1964, *Adv. Geophys.*, **10**, 317–488.

BÉDARD, G., 1966, *Phys. Rev.*, **151**, 1038–9.

BENEDEK, G. B., 1968, *Polarisation Matière et Rayonnement, Livre de Jubilé en l'Honneur du Professeur A. Kastler* (Paris: Presses Universitaires de France).

CUMMINS, H. Z., and SWINNEY, H. L., 1969, *Progress in Optics*, Vol. VIII, Ed. E. Wolf (Amsterdam: North-Holland).

DIALETIS, D., 1969, *J. Phys.* A: *Gen. Phys.*, **2**, 229–35.

FOORD, R., *et al.*, 1969, *Proc. Southampton Conf. on Lasers and Opto-Electronics* (*Proc. I.E.R.E.*, to be published).

GLAUBER, R. J., 1963, *Phys. Rev.*, **131**, 2766–88.

JAKEMAN, E., OLIVER, C. J., and PIKE, E. R., 1968, *J. Phys.* A: *Gen. Phys.*, **1**, 406–8.

JAKEMAN, E., and PIKE, E. R., 1968, *J. Phys.* A: *Gen. Phys.*, **1**, 128–38.

—— 1969 a, *J. Phys.* A: *Gen. Phys.*, **2**, 115–25.

—— 1969 b, *J. Phys.* A: *Gen. Phys.*, **2**, 411–2.

MANDEL, L., 1959, *Proc. Phys. Soc.*, **74**, 233–43.

MEYER, M. A., and MIDDLETON, D., 1954, *J. Appl. Phys.*, **25**, 1037–52.

PIKE, E. R., 1969, *Proc. Inaugural Conf. European Phys. Soc.* (*Nuovo Cim.*, in the press).

SIEGERT, A. J. F., 1943, *M.I.T. Rad. Lab. Rep.*, No. 465.

VAN VLECK, J. H., 1943, *Harvard University Radio Res. Lab. Rep.*, No. 51.

VAN VLECK, J. H., and MIDDLETON, D., 1966, *Proc. Inst. Elect. Electron. Engrs*, **54**, 2–18.