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1973 J. Phys. A: Math. Nucl. Gen. 6 1327

(http://iopscience.iop.org/0301-0015/6/9/008)

J. Phys. A: Math., Nucl. Gen., Vol. 6, September 1973. Printed in Great Britain.

Photon-correlation spectroscopy: dependence of linewidth error on normalization, clip level, detector area, sample time and count rate

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Received 6 March 1973

Abstract. This paper contains a detailed investigation of the factors affecting the accuracy that can be achieved in a given intensity-fluctuation measurement by clipped photon-correlation spectroscopy. The effects of different normalization methods, selection of clip level, spatial and temporal integration over detector area and sample time, and varying count rate are investigated. Comparison is made with theoretical predictions and also with computer simulation.

1. Introduction

The technique of clipped photon correlation (Jakeman and Pike 1969) is becoming increasingly widely applied in a variety of fields. A full theory has been given by Jakeman (1970) who derived expressions for the effects of clipping, count rate and temporal integration on the observed spectral parameters. This theory has been extended by Jakeman *et al* (1971a) and Koppel (1971) to include the effect of detector dead time and spatial integration over the detector area. In addition to these analyses of the correlation coefficients a number of workers have performed a theoretical study of the statistical errors involved. While this work has led to a reasonable qualitative understanding of the effects of the various parameters, none of the treatments have been wholly satisfactory since, because of the complexity of the problem, approximations or simplifications of some kind have had to be made.

In this paper we describe experimental measurements and further computer simulations for a gaussian–lorentzian source, namely scattering from the brownian motion of a spherical monodisperse macromolecule, using the technique of single-clipped correlation†. The dependence of the measured linewidth accuracy on various parameters is determined and compared with the available theory or with the computer simulation. Direct comparison with the detailed results of Jakeman *et al* (1970b, 1971b, to be referred to as JPS70, 71), shows their theory to be adequately verified. From the various sets of results optimum operating conditions for single-clipped correlation will be deduced.

† The experiments were performed on a Malvern system 4300 photon-correlation spectrometer manufactured by Precision Devices and Systems Ltd, Spring Lane, Malvern, Worcestershire.

2. Experimental parameters

Analysis of the effects of various parameters on the errors in linewidth measurements using intensity-fluctuation spectroscopy have been made by many authors (Benedek 1968, Haus 1969, Cummins and Swinney 1970, JPS70, 71, Degiorgio and Lastovka 1971 (to be referred to as DL71), Kelly 1971 (to be referred to as K71)); the last three publications are the most comprehensive. However these three are not directly comparable since DL71, JPS70, 71 and K71 analyse the data with three-, two- and one-parameter fits respectively. In the single-clipped correlation measurements under discussion the correlation coefficient for the rth delay has the form

$$G_k^{(2)}(rT) = X + Y\exp(-2\Gamma rT) \tag{1}$$

where T is the sample time, Γ is the optical linewidth and X and Y are constants. However, as was shown by Jakeman and Pike (1969) this expression can be factorized so that

$$G_k^{(2)}(rT) = \langle n\rangle \langle n_k\rangle \{1 + C\exp(-2\Gamma rT)\} \tag{2}$$

where $\langle n\rangle$ and $\langle n_k\rangle$ are the mean unclipped and clipped counts per sample time respectively, and C is a constant determined by the operating conditions. This implies that analysis of the data using three variables as in equation (1) such as in DL71 is unnecessary and generally leads to larger errors. The constant C in equation (2) has been shown (Jakeman 1970, Jakeman *et al* 1971a, Koppel 1971) to be expressible analytically in terms of the various experimental parameters and could therefore also be eliminated. In practice however the actual correlation measurement provides the best estimate of C, so that a two-parameter fit (JPS70, 71) is more realistic than a one-parameter fit (K71) and is used here in the detailed comparison of experiment with theory. The experimental variables which govern the error in the linewidth estimator are: k, the clipping level; T/τ_c, governing temporal integration, where τ_c ($= 1/\Gamma$) is the optical coherence time; A/A_c, governing spatial integration, where A is the solid angle subtended at the source by the detector and A_c is the solid angle of the first Airey disc formed by the source aperture at the detector (called the coherence area); $\bar{R}$, the counts per coherence time per coherence area; and m, the number of correlation channels.

In previous theoretical analyses the effects of clipping level have been investigated by DL71 and K71 who both considered all k values and JPS71 who only took $k = 0$. The effect of finite sample time has been treated by the latter authors. Several authors (Benedek 1968, Haus 1969, Cummins and Swinney 1970, DL71) have considered the effects of large detector area ($A \gg A_c$) while DL71 and JPS71 took small but finite detector areas ($A < A_c$). DL71, K71 and JPS71 all studied the effects of count rate and the number of correlation channels. However, detailed comparison can only really be made here with JPS71; where this theory is incomplete, computer simulation is adopted.

In this work twenty correlation channels were used throughout and the effects of the remaining variables investigated. In addition different data normalization schemes were compared since these also affected statistical accuracy. One, generally insignificant, difference between the theoretical analysis of JPS70, 71 and the experiments is that JPS70, 71 adopted optimum weighting in the least-squares fitting procedure, whereas experimentally and in computer simulation uniform weighting was adopted; however, this is usually close to optimum.

3. Computer simulation

In the situations where analytic methods of error estimation become excessively complicated computer simulation has proved particularly useful. This situation occurs for the case of high clip levels. This method uses the known properties of the radiation field and photodetectors to construct correlation functions which can then be analysed in the usual manner with a least-squares fitting procedure. If the optical field corresponds to a gaussian–lorentzian source then the positive-frequency component of the electric field, $\mathscr{E}^+$, is a complex variable whose real and imaginary parts are statistically independent

$$\mathscr{E}^+(t) = \mathscr{E}_r(t) + i\mathscr{E}_i(t). \tag{3}$$

Values of $\mathscr{E}_r$ and $\mathscr{E}_i$ at times t and $t-\tau$ are related by two first-order Markoff processes given by

$$\mathscr{E}_r(t) = \rho\mathscr{E}_r(t-\tau)+\alpha_1 \tag{4}$$

$$\mathscr{E}_i(t) = \rho\mathscr{E}_i(t-\tau) + \alpha_2 \tag{5}$$

where α_1 and α_2 are independent random numbers with gaussian probability distributions of equal variance about zero mean. If τ is the delay between successive samples then ρ is related to Γ by

$$\rho = e^{-\Gamma\tau}. \tag{6}$$

Since the photodetector responds to the square modulus of $\mathscr{E}^+(t)$ the sum of the squares of $\mathscr{E}_r(t)$ and $\mathscr{E}_i(t)$ gives the detected intensity. Using independent series of gaussian random numbers for α_1 and α_2 an intensity distribution can be simulated. The intensity at each sample is then used as the mean value of a Poisson random number generator whose output $n(t)$ is an integer corresponding to the photocounts occurring in a sample at time t. From the sequence of values of $n(t)$ a simulated photon-counting correlation function can be calculated. Sets of these correlation functions may be produced and analysed to determine the error in the fitted values of Γ, using the same computer programs as in the analysis of the experimental work.

The simulation corresponds to a slightly different sampling scheme from the actual experiments in that the sample time is assumed to be infinitesimally short. The effect of temporal integration only becomes apparent, as we shall see later, where high count rates with low clip levels are involved; in which case the exponential part of the correlation function is reduced significantly making the simulated error lower than the observed and theoretically predicted value.

4. Dependence on the method of data normalization

The operation of the single-clipping photon-counting correlator has been described elsewhere (Jakeman and Pike 1969, Foord *et al* 1969, 1970, JPS71). In particular JPS71 discuss in detail the effect of the normalization of the counts recorded in the correlator store. The data in the store must be normalized by dividing by the product $N\langle n\rangle\langle n_k\rangle$, where N is the total number of samples, $\langle n\rangle$ is the mean unclipped count rate per sample time and $\langle n_k\rangle$ is the mean count rate clipped at k. However we only have estimates of $\langle n\rangle$ and $\langle n_k\rangle$ and so normalization using these estimates results in a bias of order $1/N$ (JPS71) which is usually negligible compared with the statistical

fluctuations in the data points. It is apparent that this type of independent normalization has the desirable feature of removing some of the effects of drift in count rate from run to run. In addition it is found to remove some of the statistical fluctuations due to variations in the total photodetections recorded for the same mean count rate. In order to analyse our data we perform a large number of runs (eg 25) which are each independently normalized against their individual total clipped and unclipped counts to give biased estimates of the clipped correlation function. An alternative technique involving much smaller bias would be subsequently to normalize each run against the overall measured count rates. The effects of these two types of normalization will be compared. Other data processing schemes can be devised which improve on the subsequent normalization method (Chen *et al* 1972). These have the effect of introducing other types of bias into the correlation function estimator but do not in general achieve as great a gain in accuracy as the scheme outlined here.

Whichever normalization technique is adopted the normalized single-clipped correlation function for a gaussian–lorentzian source has the form

$$g_k^{(2)}(rT) = 1 + C\exp(-2\Gamma rT) + \mathrm{O}\left(\frac{1}{N}\right) \tag{7}$$

similar to equation (2) but including terms of order $1/N$ due to the biasing. From a series of experiments using a least-squares fitting procedure treating C and Γ as variables the value of the optical linewidth determined by each measurement and hence the mean value and standard deviation of the results can be obtained.

Initially, therefore, let us investigate the effect of the normalization method on the performance of the correlator. The incident count rate $\bar{R}$ (counts per coherence time, τ_c, per coherence area, A_c) was adjusted to a value of 2000. The detector area A was chosen to give a value of A/A_c of 0·01 and the sample time so that T/τ_c was 0·1. The mean counts per sample time ($\bar{n}$) were then equal to 2. A series of 25 runs each of 10^5 samples utilizing single clipping at $k = 0, 2, 4, 8$ were taken and the standard deviation calculated in each case. The analysis for each situation was performed using both independent and subsequent normalization. Computer simulation of the independent normalization results for the same conditions was also performed. The results are shown in figure 1 where smooth curves have been drawn through the data points for the sake of clarity. The independent normalization results are everywhere more accurate than the subsequent normalization ones. This is particularly apparent at $k = 0$ where the statistical fluctuations in the normalization constant are greatest using subsequent normalization. The agreement between simulated and experimental results is within a statistical uncertainty of approximately 20% in each. If we compare the minimum errors in each case occurring for $k \simeq \bar{n}$, of 3·3% (independent normalization) and 7% (subsequent normalization) we see that the independently normalized estimates are more accurate by about a factor of two than the subsequently normalized ones.

5. Dependence on the clip level

Though individual terms cannot be factorized when discussing the form of the correlation function (Jakeman *et al* 1971a) under many circumstances a reasonable approximation to the true errors can be obtained if this assumption is made. The effect of clip level on the errors proved too cumbersome to treat analytically, except for $k = 0$, so computer simulation and experimental results were compared, each analysed using the

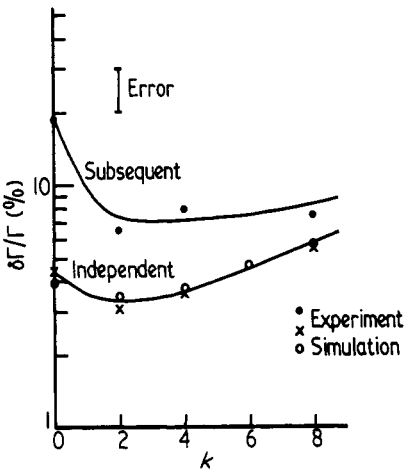

Figure 1. The dependence of the linewidth error on biasing. Single-clipping measurements and simulation are shown for $\bar{n} = 2$ and for clip levels $k = 0, 2, 4, 8$; $T/\tau_c = 0{\cdot}1$ and $A/A_c = 0{\cdot}01$. Both subsequent and independent normalization are used. A smooth curve is drawn through the data to aid interpretation. The results are for 10^4 coherence times.

independent normalization scheme. A series of measurements with $A/A_c = 0{\cdot}01$, $T/\tau_c = 0{\cdot}1$ and $\bar{R}$ having values of 350 ($\bar{n} = 0{\cdot}35$), 1000 ($\bar{n} = 1{\cdot}0$), 4000 ($\bar{n} = 4{\cdot}0$) and 10 000 ($\bar{n} = 10{\cdot}0$) were taken for various clip levels, k. For comparison computer simulation was carried out for the $\bar{n} = 1$ and $\bar{n} = 10$ data. The results are shown in figure 2 with smooth curves drawn through the data points as before. Since the individual points have an accuracy of $\pm 20\%$ the agreement is adequate. The simulated point

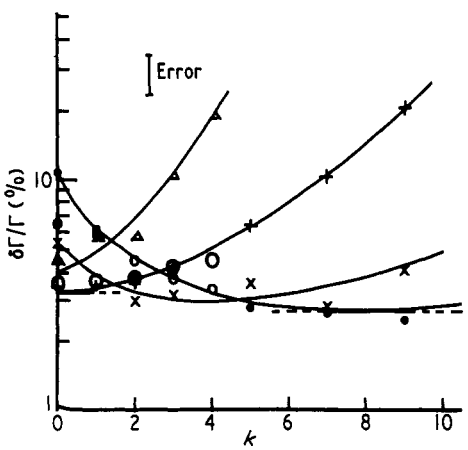

Figure 2. The dependence of linewidth error in single-clipped correlation on clip level as a function of the overall count rate $\bar{R} = 350$, 1000, 4000 and 10 000. Since $T/\tau_c = 0{\cdot}1$ and $A/A_c = 0{\cdot}01$ the count rates per sample time are $\bar{n} = 0{\cdot}35(\triangle)$, $1{\cdot}0(+)$, $4{\cdot}0(\times)$ and $10{\cdot}0(\bullet)$ respectively; simulated results for $\bar{n} = 1{\cdot}0(\bigcirc)$ and $10{\cdot}0(\circ)$ are included. Independent normalization is used throughout. The results for simulated full correlation when $R = 1000$ and 10 000 are shown for comparison as broken lines. Smooth curves are drawn through the data for clarity.

for $\bar{n} = 10$, $k = 0$ however shows a marked discrepancy of 2 standard deviations from the experimental result. This results from the above mentioned difference in the sampling schemes used for the simulation and for the experiment. The experiment uses a sample time equal to the separation of the samples whereas the simulation assumes infinitely short samples. This would yield a simulated value for $g^{(2)}(\tau)-1$ a factor of at least 60–70% higher (Jakeman *et al* 1971a). In addition the effect of the area correction has been ignored in the simulation which will again drop the observed value of $g^{(2)}(\tau)-1$ below the simulated. As the clip level increases this discrepancy will be reduced considerably, as is in fact observed.

From the form of the curves in figure 2 it seems that the optimum choice of clip level lies in the region of $k = \bar{n}$ though the function is in fact rather slowly varying with k. This is similar to the result of DL71. The comparative accuracy of single-clipped and ideal correlation can be obtained from figure 2 by comparing the accuracy at optimum k with that for the full correlator obtained by simulation at $\bar{n} = 1$ and 10. The close agreement between the single-clipped and the full correlation results shows that virtually no information has been lost in the clipping process. The theory of JPS71 predicts greater accuracy for the ideal correlator but this is probably due to the various approximations made in this work, in particular, to the neglect of correlation effects between the different coefficients.

6. Dependence on count rate, detector area and sample time

The linewidth accuracy can now be expressed as a function of the form

$$\frac{\delta\Gamma}{\Gamma}(A/A_c, T/\tau_c, \bar{R})$$

where the effect of clip level has been already taken out. For convenience of representation we shall consider the error in Γ as a function of pairs of these parameters with the effect of the third parameter made negligible, or at least factorizable in the small $\bar{n}$ limit.

(i) $\frac{\delta\Gamma}{\Gamma}(\bar{R}, T/\tau_c)$ where $A/A_c \ll 1$

(ii) $\frac{\delta\Gamma}{\Gamma}(\bar{R}, A/A_c)$ where $T/\tau_c \ll 1$

(iii) $\frac{\delta\Gamma}{\Gamma}(T/\tau_c, A/A_c)$ where $\bar{R} \ll 1$.

These dependences can now be investigated individually:

(i) $\frac{\delta\Gamma}{\Gamma}(\bar{R}, T/\tau_c)$ where $A/A_c \ll 1$.

This dependence was given in JPS71 for single clipping at $k = 0$. Experiments were performed with twenty correlation channels using a coherence area ratio $A/A_c = 0{\cdot}01$ with $\bar{R}$ taking values 10, 100, 1000 and T/τ_c varying between 0·01 and 0·5. The results

are shown in figure 3. Where the predicted accuracy lay outside the 10 % to 50 % range the number of samples taken was reduced by 10 or increased by 10 or 100 as required. Where more samples were taken due to low count rates it is permissible to renormalize

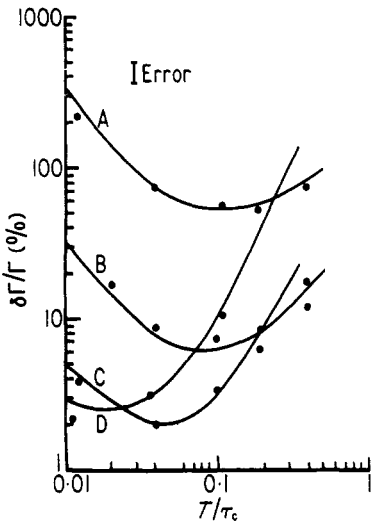

Figure 3. The dependence of the linewidth error on sample time for different overall count rates $\bar{R}$ = 10, 100, 1000 and 10 000 (curves A, B, C and D, respectively). A/A_c = 0·01 and T/τ_c varies between 0·01 and 0·5. The clipping was at k = 0 throughout. The results are for 10^4 coherence times.

the error assuming a straightforward dependence on $1/\sqrt{N}$. However this is not strictly true for high count rates though it has in fact been assumed here since the error introduced would be less than the experimental uncertainties. Agreement is seen to be adequate under all circumstances. As was shown previously in JPS71 and DL71 for low count rates ($\bar{n} < 0{\cdot}1$) the optimum choice of sample time is $T/\tau_c \simeq 0{\cdot}1$. At high count rates a lower sample time is preferred because of the clip level (k = 0) chosen. If this is allowed to vary so that $k = \bar{n}$ the same sample time as before, such that the correlator spans approximately two optical correlation times, would be optimum.

(ii) $\dfrac{\delta\Gamma}{\Gamma}(\bar{R}, A/A_c)$ where $T/\tau_c \ll 1$.

The condition $T/\tau_c = 0{\cdot}1$ found optimum in the previous section was adopted in this section. The dependence can be given analytically in two limits. One is where $\bar{n} \ll 1$ in which case clipped and unclipped correlation are essentially identical and the dependences on $\bar{R}$ and A/A_c can be factorized combining the results of JPS71, for $T/\tau_c = 0{\cdot}1$, $A/A_c = 0$ with varying $\bar{R}$, given in figure 3 with those of Scarl (1968) and Jakeman *et al* (1970a) for the detector area correction. The linewidth error can therefore be expressed as

$$\frac{\delta\Gamma}{\Gamma}(\bar{R}, A/A_c) = \frac{1}{f(A/A_c)} \frac{\delta\Gamma}{\Gamma}(\bar{R}). \tag{8}$$

This expression provides the continuous curve on the left of figure 4.

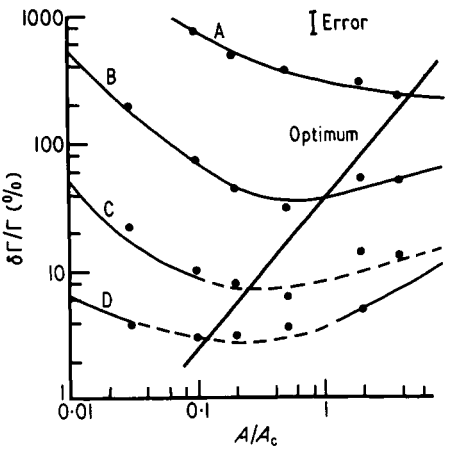

Figure 4. The dependence of the linewidth error on detector area for different overall count rates $\bar{R}$ = 0·1, 1·0, 10 and 100 (curves A, B, C and D, respectively). A/A_c varies between 0·01 and 4·0; T/τ_c = 0·1. Also shown is the optimum detector area setting for each count rate $\bar{R}$. The results are for 10^4 coherence times.

In the opposite limit where $\bar{n}$ and A/A_c are both much greater than one the error dependence for full correlation is

$$\frac{\delta\Gamma}{\Gamma}(\bar{R}, A/A_c) \simeq \left(\frac{1}{f(A/A_c)}\frac{1}{N}\frac{\tau_c}{T}\right)^{\frac{1}{2}} \tag{9}$$

for $\bar{n} \to \infty$ and $A/A_c \to \infty$. For large A/A_c $f(A/A_c) \propto 1/A$ and so the error becomes proportional to $\sqrt{A}$ as found by Degiorgio and Lastovka for the three-parameter fit. Assuming that the effect of clipping can be factorized as in figure 2, the asymptotic values for the large area will be $\delta\Gamma/\Gamma$ = 11·1%(A/A_c = 10) and 3·5% (A/A_c = 1·0). This asymptotic line is included on the right of figure 4. Intermediate regions are denoted by a smooth broken curve.

Series of measurements were made with T/τ_c = 0·1 ; $\bar{R}$ having values 0·1, 1, 10, 100 and A/A_c varying between 0·03 and 4·0. The experimental duration was chosen to give errors in the range 10%–50% as before and the results, given in figure 4, corrected to 10^4 coherence times. Agreement is again satisfactory. It can be seen that the result of increasing the detector area for low count rates is to improve the statistical accuracy of the data even though it reduces the exponential part of the correlation function. However once the counts per sample time, $\bar{n}$, exceed a certain value this statistical gain is overcome by the reduction in the exponential component. A line has been drawn through the approximate points in figure 4 where this occurs corresponding to count rates per sample time of $\bar{n}$ = 0·05, 0·1, 0·25 and 1·2 respectively. Optimum operating conditions for any other value of $\bar{R}$ could be deduced by interpolation along this line. However since the minima are very flat the adoption of A/A_c = 1 would be a suitable compromise.

(iii) $\dfrac{\delta\Gamma}{\Gamma}(T/\tau_c, A/A_c)$ where $\bar{R} \ll 1$.

Measurements were made for $\bar{R}$ = 0·1, T/τ_c = 0·1 and 0·46 and A/A_c was varied between

0·1 and 4·0. Under these conditions $\bar{n} \ll 1$ and so the components can be factorized as in full correlation. The linewidth error can then be expressed as

$$\left.\frac{\delta\Gamma}{\Gamma}\left(\frac{T}{\tau_c}, \frac{A}{A_c}\right)\right|_{\bar{R}=0\cdot1} = \frac{1}{f(A/A_c)} \left.\frac{\delta\Gamma}{\Gamma}\left(\frac{T}{\tau_c}\right)\right|_{\bar{R}=0\cdot1}. \tag{10}$$

The continuous curves in figure 5 have this form.

With the low count rates chosen many more than 10^4 coherence times were required; the data being renormalized as before. The agreement between theory and experiment is again satisfactory indicating the validity of the analysis.

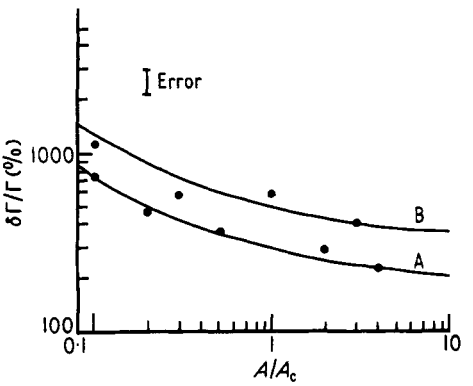

Figure 5. The dependence of the linewidth error on detector area for different sample times. $\bar{R} = 0{\cdot}1$, $T/\tau_c = 0{\cdot}1$ (curve A) and 0·46 (curve B) and A/A_c varies between 0·1 and 4·0. The results are for 10^4 coherence times.

7. Conclusions

In conclusion we summarize the operating principles for single-clipped photon correlation which may be deduced from the results of the previous sections.

(i) Each run should be normalized independently against its own total counts since this gives greater accuracy than subsequent normalization against a long term mean value. The best experimental technique uses an on-line computer to normalize a large number of short runs independently. The length of the individual runs can be reduced until the bias, of order $1/N$, affects the results measurably, that is for $N = 10^3$ say.

(ii) The clip level should be set equal to the mean counts per sample time in which case the loss in accuracy due to clipping is insignificant.

(iii) For a given number of channels the optimum choice of sample time is such that the correlation function extends for about two optical correlation times.

(iv) For a given cross section, that is, count rate per coherence area per coherence time $\bar{R}$, the detector area should be increased until the value of A/A_c reaches an optimum value which can be deduced from figure 4. Alternatively use with $A/A_c = 1$ is near optimum for a very wide range of scattering cross sections.

It must be realized that all the discussion and results presented here have applied to a single lorentzian spectral component. If spectra containing more than one component are encountered similar operating principles will apply but the accuracy required in individual points, and hence experiment duration, will be much greater.

Acknowledgments

The authors wish to thank P N Pusey of the Royal Radar Establishment for useful comments and discussion.

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