This item was submitted to Loughborough University as a PhD thesis by the author and is made available in the Institutional Repository (https://dspace.lboro.ac.uk/) under the following Creative Commons Licence conditions.

For the full text of this licence, please go to:
http://creativecommons.org/licenses/by-nc-nd/2.5/
Analogue Measurement of Scattered Light Fluctuations

by

Douglas Andrew Green

A Doctoral Thesis

Submitted in partial fulfilment of the requirements for the award of
Doctor of Philosophy of the Loughborough University of Technology

October 1997

© by D A Green 1997
Analogue Measurement of Scattered Light Fluctuations

Abstract

This thesis investigates two methods of optical analysis of multiphase fluids. These two methods are nephelometry and the statistical analysis of scattered light intensity fluctuations.

Nephelometry is an established technique for investigating particulate suspensions. In this work the basic technique is combined with neural network processing to develop a system capable of automatically distinguishing and quantifying different suspensions, in particular suspensions of oil. Evidence obtained in this study suggests that neural networks can distinguish the light scattering from suspensions of different size distributions and produce a more accurate estimate of volume fraction than commonly used turbidity measurements.

Non-Gaussian, fluctuating light intensities arise from the scattering of light from a varying population of suspended particles. Successful measurement of these intensity fluctuations makes feasible new instrumentation based on the statistical behaviour of the detected signal. Analyses that could prove possible include particle number, size, type and flow characteristics. Photon counting methods have traditionally been used to measure fluctuations from random media but the lower cost of analogue pin diodes makes them the preferred choice of detector if they can be applied usefully. A method of quantifying the effect of noise from the diode detectors and removing it from the statistics of the fluctuations is developed from a model of the pin diode detectors. Experimental results show that detector noise can be compensated for in the analysis of scattered light fluctuations. Results also indicate that the model used to describe the scattering process is valid and that further work can lead to a practical instrument for the study of suspensions.
Acknowledgement

I would like to thank my supervisor, Dr Peter Smith, for his help, guidance and encouragement in all aspects of work during the period of this study.

Thanks is due also to Dr Rambo Naimimohasses for his related work which included the training and testing of artificial neural networks. David Barnett also contributed directly with windows based data acquisition software. In addition to these specific contributions their general assistance in the running of the laboratory and thought provoking discussions have been most welcome. Likewise Gabriella Ruiz and the other members of the Optical Engineering Group, including project students, have all added variety and interest to my working days.

For his efforts in explaining some of the more advanced mathematics of the random walk models Dr Keith Hopcraft at the University of Nottingham has my thanks too.

This work has been supported by an EPSRC research grant GR/H45261.
## Contents

1. Introduction 4
   1.1. Equipment Considerations 5
   1.2. Current Instrumentation 6
   1.3. Research Objectives 10
   1.4. Outline of Thesis 12

2. Light Scattering Models 14
   2.1. Scattering Calculations 14
       2.1.1. Mie Theory 14
       2.1.2. Rayleigh Scattering 16
       2.1.3. Rayleigh-Gans Model 17
       2.1.4. Anomalous Diffraction and Ray Optics 18
       2.1.5. Application of Models to Systems of Objects 19
       2.1.6. Application of Neural Networks to the Inverse Problem 19
   2.2. Statistical Optics 20
       2.2.1. Random Walk Model 21
       2.2.2. Special Cases of the Random Walk Model 30
       2.2.3. Description of Distributions by their Moments and Important Relationships 39
       2.2.4. Finite N and Cross Correlations 42
   2.3. Chapter Summary 45

3. System for Nephelometric Experiments 47
   3.1. Flow system 47
   3.2. Nephelometer Sensor Heads 49
       3.2.1. Mk I Nephelometer 50
       3.2.2. Mk II Nephelometer 50
   3.3. Light Sources 56
   3.4. Source Drivers 59
   3.5. Detectors 60
   3.6. Synchronisation 62
   3.7. Experimental Systems 63

4. Discussion of Nephelometric Results 66
   4.1. Mk I Nephelometer 66
       4.1.1. Comparison of Light Sources 66
       4.1.2. Effect of Water Temperature 78
       4.1.3. Scattering from Different Oil Types 84
       4.1.4. Neural Network Analysis 105
       4.1.5. Summary of Mk I Nephelometer Study 108
   4.2. Mk II Nephelometer 109
       4.2.1. Effect of Flow Rate and Temperature 109
       4.2.2. Scattering from Different Oil Types 114
       4.2.3. Neural Network Analysis 124
   4.3. Summary of Nephelometric Study 126

5. System for Speckle Statistic Experiments 127
   5.1. Receiver Characterisation 127
   5.2. Speckle Characterisation 129
   5.3. Particle Size Distributions 136
6. Discussion of Speckle Statistical Results
   6.1. Receiver Characterisation
      6.1.1. Receiver Noise Model
      6.1.2. Comparison of Receiver Noise and Model
      6.1.3. Deconvolution of Receiver Noise
   6.2. Discussion of Point Statistics
      6.2.1. Empty Cuvette Experiments
      6.2.2. Tap Water Experiments
      6.2.3. Polystyrene Spherule Experiments
      6.2.4. Desert Dust Experiments
      6.2.5. Soluble Oil Experiments
      6.2.6. Correlation of Results
      6.2.7. Ballotini Experiments
      6.2.8. Reliability of Moments
   6.3. Discussion of Cross Correlations
      6.3.1. Empty Cuvette Experiments
      6.3.2. Tap Water Experiments
      6.3.3. Polystyrene Spherule Experiments
      6.3.4. Desert Dust Experiments
      6.3.5. Soluble Oil Experiments
      6.3.6. Ballotini Experiments
      6.3.7. Summary of Cross Correlations
      6.3.8. Normalised Intensity Cross Correlation Function
   6.4. Summary of Statistical Results

7. Conclusions
   7.1. Nephelometry
   7.2. Statistical Analysis

8. Discussion of Conclusions and Further Work
   8.1. Nephelometry
   8.2. Statistical Analysis
      8.2.1. Practical Considerations
      8.2.2. Development of Theory
   8.3. General Comment

9. References

Appendices
   A. Electrical Hardware
   B. Summary of Experimental Details
   C. Linescan Camera Software
1. Introduction

The content of this thesis is concerned with the analysis of multiphase fluids by light scattering techniques. Light scattered by suspended particles not only reveals the presence of those particles, but the way in which the light is scattered contains information about the particles. Applications for techniques which can analyse suspensions are widespread, the largest generic areas probably being industrial processes, environmental monitoring and biomedical studies. Electromagnetic scattering can also find uses in other fields such as the military and as a tool for scientific research. In this work it is applications in the fields of the environment and industrial processes that are addressed most directly.

The demand for new instrumentation to monitor the environment is being driven by public concern about health and safety in general and the effects that modern lifestyles are having on our environment. The amount of public concern has led governments to take a number of steps. Government is now funding more research into the state of the environment on a global scale and also on the effects of pollution. This has in turn led to an increase in the amount of scientific equipment being needed to make the necessary studies. Many of the studies being made are concerned with particulate matter; for example, smoke from industry, oil in water suspensions and silt transport [1.1][1.2][1.3][1.4]. Whilst research may provide a market for certain types of, often expensive, monitoring equipment, industry is being forced to make more widespread use of environmental monitoring. Industry is being made more responsible for the discharges it makes to the atmosphere and water courses and the threat of prosecution effectively means that industry must monitor its output. Prosecuting bodies such as the National Rivers Authority (now part of the Environment Agency) also require various monitoring tools to carry out their policing role.

The water industry is of particular interest because of directives made by the Economic Community (EC) on water quality for human consumption (directive 80/778/EEC). This directive states that particular materials in the water, such as pesticides and microbes, need to be measured and must not exceed certain levels of concentration. Microbiological content will scatter light propagating in the water and this is one application to which light scatter techniques could be applied. Proposals were made to enforce a maximum solids content in potable water which would be based on volume fraction rather than the current turbidity measurement. This is another area where new instrumentation could be needed. It is likely that legislation such as EC directives will be the driving force which actually makes industry use new monitoring equipment.
Process industries are interested in instrumentation for two reasons other than effluent monitoring. Monitoring a process should ensure that only products of the right quality are released to the market place. This quality assurance may protect a company’s reputation but greater financial incentives accrue if instrumentation feeds back into a control loop. Improved control can make a process more efficient in terms of materials used / wasted and energy requirements. Identification of changing process conditions can point to early warning of maintenance needs, leading to less down time and higher productivity. All these types of saving are an incentive to process industries to consider installing instrumentation that gives new information about the process without sampling and remote analysis. The types of processes that light scattering techniques would be applicable to are those which involve immiscible fluids or particles in suspension, e.g. crystallisation from solutions. Light scattering has also been used for some time for clean room monitoring in the electronics industry; if more information could be found about the dust particles it could identify sources of dirt in the production environment.

1.1. **Equipment Considerations**

For new instrumentation to be successful it must provide useful information at a cost which is less than the savings which that information can lead to. Information about suspensions that is useful are size distribution, type of suspended material or distribution of materials, concentration and shape. Size distribution and concentration are useful to know since they can indicate how advanced a process is towards completion and also check that the process is continuing correctly. Knowing the type of suspended material can be of use in environmental monitoring to trace a source of contamination. Shape can be another clue in identifying particles and also effects the way the particles behave e.g. flow characteristics and adhesion.

To gain maximum benefit from measurements it is desirable that they be made in-situ or on-line in a process. In-situ measurements circumvent a number of problems that laboratory based methods have. If measurements have to be made in a laboratory a sample must be taken, the significance of the measurement then being limited by the relation between sample and process stream. There are difficulties with obtaining representative samples and the sample may change state during transfer to the laboratory. Also, in-situ measurements do not suffer from the long time delays of laboratory analysis, allowing a faster response in a control loop. The drawback with in-situ methods is that they are more difficult to perform and hence such detailed analyses are not generally possible.
One of the major problems with current equipment is the cost of installation. Although this cost may be justifiable in some specialised applications, for widespread acceptance equipment must be cheap meaning only simple technologies may be used.

1.2. Current Instrumentation

What follows is a brief review of some of the optical particle sensing / sizing equipment that is currently in the market and the methods that they use. None of these instruments are used to identify the material of the suspended particles and some use apriori knowledge of the material to improve accuracy. Shape is another parameter that is not currently measured by these types of instrument, most assume spherical particles. All of them, in a strict sense, use light scattering although they measure different angular ranges and process the data differently.

The simplest instrument is the turbidity meter. This basically gives a measure of the clarity of a sample. Most modern turbidity meters measure the ratio of light intensity scattered at 90° to that transmitted through the sample. The measurement is presented in Nephelometric Turbidity Units (NTU) which is a measure based on a formazine standard. British BSI and American ASTM standards [1.5] [1.6] exist for the production of the formazine standard and the optical apparatus but few manufacturers follow them or appear to be aware that they refer to the optical design as well as formazine calibration. Even without the differences between manufacturers and the practical problems of using formazine, the turbidity measure is ambiguous because of its dependence on the suspended material. Size distribution, refractive index and shape all affect the amount and direction of light scattered in the suspension. An analysis of how the size distribution may affect the measurement can be found in [1.7]. A number of companies manufacture turbidity meters, Camlab supply laboratory based and portable models, Solomat and Grant include them on their sondes for environmental monitoring. Zullig and BTG produce models to be used in process monitoring and control. Of the instruments considered here, turbidity meters have become the most widely used in the process environment owing to their simplicity and the fact that the water industry is forced to measure turbidity by legislation.

More sophisticated turbidity type instruments are on the market that measure the scattered light intensity at a number of angles. These are being used where it is important to have some knowledge of particle size as in monitoring oil in water. VAF and ELE sell instruments that are targeted at this particular market. The Brewery Research Foundation has been investigating the yeast content of beer
after filtering. Sigrist is a leading manufacturer of these systems for use with gaseous or liquid samples. Like turbidity meters, these instruments will be somewhat sensitive to changes in particle size and type, but the extra information gained helps to produce a measure more closely related to overall volume of suspended matter. In essence the ratio of two angular measurements is taken to be approximately proportional to the size of the particles, larger particles scattering less light to the large angles.

The scattering techniques described above, although sensitive to particle size, do not give a definite size measure. Obscuration methods are the most simple used to measure size. As a particle moves through a beam of light, the light intensity falling on a line of sight detector falls and then rises again as the particle obstructs the beam and then moves away. Measuring the time taken for the particle to pass through the beam gives a measure of the particle's size. As more particles pass the sensor a distribution of sizes can be collated. For particles smaller than the beam size the amount of light obscured by the particle (scattering and absorption cross-section) gives a measure of size. Advantages of obscuration methods are their simplicity and wide range of operation (sensing particles in the range of 1μm to 5mm) but the need to count particles individually and know the flow speed has tended to restrict its use to the laboratory. UCC, however, produce a portable monitor which can link to pipelines without interrupting the flow.

A more sophisticated particle sizing method extends the idea of measuring scattered light at several angles. By using a coherent source and imaging the forward scattered light onto a detector array the position of diffraction maxima and minima can be found (figure 1.1). The diameter of these diffraction rings depend on the size of the particles and their refractive index. Prior knowledge of the suspended material helps to guarantee an accurate result. By using a lens to image at its focal distance the position of particles in the light beam does not affect the position of the diffraction rings (figure 1.2). This means that with some signal processing more than one particle can be in the field of view at a time, giving some advantage over obscuration methods. The disadvantage is the cost of these diffraction systems which tend towards £100 000. The cost of these systems is one reason why they do not yet have widespread use on-line. These types of instrument operate successfully over particle sizes (diameter) of approximately 1μm to 1000μm. Coulter extend the range of their instrument down to 0.1μm by adding a system based on the polarisation of light scattered at angles near to 90° and at different wavelengths. Malvern extend the range of their Mastersizer by rearranging the optical set-up, a technique they call reverse Fourier optics. The main competitor to Coulter and Malvern is the French company Cilas.

Lasentec have developed a technique of sizing that is in a way opposite to the obscuration method. The Lasentec instruments focus a laser just inside a sample jar or at the end of a probe. The laser beam is then scanned rapidly across any particles that may be in the focal plane and back scattered light is
Figure 1.1. Typical diffraction type particle sizer configuration.

Figure 1.2. Fourier lens imaging, light scattered at the same angle is focused onto an annulus in the focal plane.
detected by two detectors. The length of the pulses of backscattered light are measured and assumed to be proportional to the size of the particle. Only strong signals are processed, that is those returns from particles at the focal point of the laser. Weak scatter signals from particles away from the focal point are filtered out by a threshold method. As more particles are counted a statistical picture of the particle size distribution can be built up. This method relies on the particles being in motion so that the same few are not counted again and again. Although essentially a particle counting method, relatively high concentrations are needed to get a reliable response in a reasonable time.

There are other methods of particle sizing that use dynamic light scattering, that is information in the time dependence of scattered light is used as well as the intensity. Photon correlation spectroscopy (PCS) is used to measure small particles, from 1nm to 5μm. Small particles in suspension move with Brownian motion and as a particle drifts through the focal point of a laser, a detector (normally a photomultiplier) detects the scattered light. The scattered light measured fluctuates as particles diffuse in and out of the laser beam. The autocorrelation of this signal is related to the diffusion rate of the particles. Stated differently, the time it takes the autocorrelation function to decay is a measure of how fast a particle moves through the laser beam. Because the particles are being moved by random collisions with molecules, this diffusion speed is related to the momentum and size of the particles. PCS can be a useful tool because of its ability to measure down to molecular sized particles, but because it relies on diffusion there are problems applying it to situations where there is bulk flow of the material.

It is worth pointing out a particular difference between PCS and obscuration or the Lasentec instrument. Obscuration and the Lasentec instrument are both particle counters, sizing each particle that passes the sensor. PCS systems may illuminate more than one particle at a time and still produce a result that is relevant without resolving individual particles. In this respect it is similar to a turbidity meter in that the intensity of scattered light may give an idea of the particulate content but does not give a particle count.

There are a number of problems with using current technology for particle analysis, some of which have been noted above. Instruments that can measure particle size tend to be too expensive for widespread installation (diffraction, PCS, scanning laser) or there are problems such as robustness of the method and equipment when applying the technique to in-situ measurements (PCS, obscuration). Turbidity meters which are more affordable do not give size discrimination which is important for some processes. For example, if a turbidity reading were to increase whilst monitoring a crystallisation process, would it be because the volume of crystallised material had increased or because the crystals were breaking up into more smaller pieces? The correct control procedure may be different in each case.
A problem common to all methods is that of sensor fouling. In most situations optics will get dirty or micro-organisms will grow over windows and lenses. With methods that rely on light intensity measurements this is a serious problem since it reduces the signal to noise ratio and also makes it appear as if there is less light being scattered than there really is. Dynamic light scattering methods are less susceptible to fouling problems because they use temporal information which is not affected by long term drift in the intensities measured. Various techniques have been tried to reduce fouling effects. Hach make a turbidity meter where the light is scattered off the surface of the liquid. This seemingly works to some extent but still relies on having a smooth surface and no sedimentation of the suspension. Other manufacturers use a similar idea, having a column of liquid dropping like water from a tap. This is to avoid the need for optical windows in contact with the flow. Zullig, who produce turbidity meters for sludge, use a wiper to clean the optics. BTG hold a patent on soluble glass which slowly dissolves, taking away surface contamination as it does so [1.8]. All of these techniques aid in reducing the effect of sensor fouling but they are not ideal because they require maintenance or affect the quality of the optical coupling to the sample. Any system that is insensitive to fouling, such as dynamic light scattering, is to be preferred over straight intensity measurements in a process or field environment.

1.3. Research Objectives

It can be concluded from the above considerations that there are a number of problems associated with currently available instruments for particle analysis. These are summarised in the list below:

- Prohibitively expensive
- Do not discriminate shape
- Do not discriminate material of particles
- Do not measure volume fraction or number of scatterers
- Not suitable for in-situ use
- Prone to sensor fouling

Not all of these problems are relevant to all types of instrument on the market, but no single instrument available defeats all these problems. The objective of this research was therefore to further development of such instrumentation. This task is tackled in this work by two approaches:
1. Nephelometry and neural network signal processing

2. Statistical analysis of temporal fluctuations in scattered light intensity

The nephelometric technique studied is an extension of the multiple scattering angle measurements used in some commercial instruments. By using a greater number of angular detectors some redundancy is built into the system which will aid in overcoming problems of fouling. Neural network processing is used to attempt an inverse solution to the scattering and estimate the size, volume fraction and/or material type of the suspended media. The combination of the neural network processing and extra number of sensors means that the assumption does not have to be made that particle size is proportional to the ratio of light intensity scattered at two angles. The extra information obtained will also aid in discriminating the particulate material.

Particular reference is made to the use of neural networks to identify different oil samples since oils prove relatively easy to work with and there is a direct application to environmental monitoring. Identification of different oils is based on the assumption that they have different optical and hydrophobic properties, leading to differently sized oil droplets in suspension.

The statistical analysis of fluctuations in scattered light intensity is a new technique as applied to analysing suspensions. The intensity of light scattered from a collection of scatterers to any point in the far field is dependent on the scatterers and their relative positions. As the scattering media changes the observed light intensity changes. The distribution of the light intensities observed at any one point need not tend to that of a Normal distribution as is common in physical processes. The precise statistics of the observed light intensities will be related to the physical properties of the scattering media. It was the objective of the research in this area to demonstrate that different statistics could be measured for different suspensions.

Prohibitive cost is tackled in both of these methods by the simple nature of the measurements and the use of cheap analogue pin diode detectors. The average of the intensities measured in the statistical technique is the same measurement made for the nephelometry. This makes it feasible to combine both methods in a single instrument, providing the ability to gain more information for little extra cost.

The principles behind these methods of analysis are valid for all sizes of particles if a suitable wavelength of radiation is used to probe the sample. It is the size of the particles relative to the
wavelength that is important. Since this work used wavelengths in the visible region, the range of particle sizes of interest is approximately 1 to 100 μm.

1.4. Outline of Thesis

Chapter 2 introduces models commonly used to describe and predict the electromagnetic scattering from inhomogeneous media. The models are divided into two sorts, those based on deterministic calculations and those based on assumptions of randomness in the scattering process. Deterministic models are most appropriate to the nephelometric technique in which neural networks are being used to estimate an inverse solution to the question of how particles scatter light spatially. The statistical optics approach is relevant to the analysis of temporal light fluctuations from suspensions of particles. The important predictions of the theory with regard to single point measurements are presented before the technique is extended to include the joint statistics of spatially separated measurements.

Chapter 3 describes two nephelometers designed and built for the study and the overall experimental system in which they were used.

The nephelometric experiments and the results obtained are presented in chapter 4. Experiments using the nephelometer designed and built first are considered followed by the results of the neural network analysis on the obtained data. A similar presentation is then made of the experiments and data using the second nephelometer described in chapter 3.

Originally it was intended to use the nephelometric system in the investigation of statistical light fluctuations. However, this proved unsuccessful and a more controllable series of experiments had to be developed. The apparatus for these investigations into the scattered light intensity fluctuations is described in chapter 5.

Chapter 6 discusses the experiments performed to study light intensity fluctuations and the results obtained. Statistical characterisation of the detection system is firstly shown to be possible by comparing results from measurements of detector noise with a photodiode receiver model. A method of compensating for the effect of the detector noise is presented. A detailed analysis of the experimental
results follows starting with the point statistics. This is followed by an analysis of the cross correlation statistics obtained from the same sequence of experiments.

Conclusions from both the nephelometric and the light intensity fluctuation studies are stated in chapter 7.

Chapter 8 discusses the conclusions and other findings in more detail. Developments to the experiments and areas for further study are also considered.
2. Light Scattering Models

There are two different routes to predicting the type of scatter that will be obtained from an object or system of objects. One is to calculate the actual scattered field due to each object and the interactions between them. This in theory gives a precise result but is practically useful only for single or small numbers of particles. The second method is to work out what the scattered field will be like in a statistical sense. This is more applicable to systems of scatterers and scatter from unpredictable or random processes.

2.1. Scattering Calculations

The most commonly used theories to calculate light scattering from particles are well established. As evidenced from the number of commercially available instruments, (c.f. chapter 1) the predictions of these theories have been put to practical use for some time. Thorough reviews of the most popular theories mentioned here can be found in books such as Van De Hulst [2.1], Bohren and Huffman [2.2], and Bayvel and Jones [2.3]. The aim here is not to go in to depth on the mathematics of each model but to introduce the general ideas behind them. Most recent research in this area has concentrated on the inversion of the light scattering problem by various techniques such as multiwavelength methods [2.4] and empirical inversions [2.5][2.6]. Extension of the theory to non-spherical particles is also of much interest [2.7]. Continued research in the area of light scattering from aerosols and suspensions has led to continued development of multichannel [2.8] and imaging nephelometers [2.7].

2.1.1. Mie Theory

The most correct method for calculating the scattered light field is to solve Maxwell’s equations for the whole system. This is in practice not simple and is not normally tried for real systems. For some simple cases a solution can be found, as by Mie and Lorentz [2.9][2.10][2.11] for spherical particles in an incident plane wave. Solving Maxwell’s equations inside and outside the sphere and then matching the boundary conditions leads to a solution which can be expressed as an infinite series. This series converges and therefore a useful result can be obtained by truncating the series after the first few terms. Examples of the field scattered from three different spheres, as calculated by Mie theory, are shown in figures 2.1, 2.2 and 2.3. \( m \) is the relative refractive index and \( x \) is a size parameter. Points to note are
Figure 2.1. Mie theory plot for unpolarised light, $m = 1.2, x = 16$. Polar plot of scattered light, illumination from bottom of diagram.

Figure 2.2. Mie theory plot for unpolarised light, $m = 1.2, x = 2$. Polar plot of scattered light, illumination from bottom of diagram.
the relative amounts of light that get scattered to large angles. The larger particle (figure 2.1) has more intensity maxima and minima in the polar plot of scattered light and most of the energy is directed in the forward direction. The smaller particle (figure 2.2) creates a more isotropic scatter pattern. The higher refractive index of the third sphere (figure 2.3) makes it scatter the incident light more effectively, again giving more structure to the scatter pattern.

2.1.2. Rayleigh Scattering

The difficulty of solving Maxwell's equations means that approximate methods are often preferred when modelling even simple systems. The simplest of the approximations regularly used is that known as Rayleigh scattering. In this approximation the particles are small compared to the wavelength of the light, and the relative refractive index, m, is low. These conditions can be represented in the form:

\[
\frac{\text{particle diameter}}{\text{wavelength}} = x \ll 1
\]  

(2.1)

and 

\[x|m-1| \ll 1\]  

(2.2)
The condition that the particle is small compared to the wavelength means that the incident field can be assumed to be uniform over the particle. The constraint of low relative refractive index means that the phase can be assumed to be the same in the particle and outside. Under such conditions the particle will oscillate like a dipole, radiating isotropically in the plane perpendicular to the incident field’s polarisation and in a dumbbell shape in the plane of polarisation. If the incident field is unpolarised the resulting scattered field will be the sum of the two, figure 2.4. Figure 2.4 is only valid for particles with isotropic polarisability, if anisotropic particles are considered the polarisability should be resolved along the three Cartesian axes.

![Figure 2.4. Polar plot of Rayleigh scattered light.](image)

*The plot for unpolarised light is drawn as the average of the plots for plane and perpendicular polarised light. Illumination from bottom of diagram.*

2.1.3. Rayleigh-Gans Model

An approximation for larger particles that scatter weakly is the Rayleigh-Gans model or Born approximation. In this model it is assumed that the internal field is the same as the external, hence its application to weak scatterers only. These restrictions are a slightly different limit to the Rayleigh case and can be represented by
The volume of the scattering particle is divided into infinitesimal components each acting as a Rayleigh scatterer. Integrating the resultant fields over the volume of the whole particle and making use of approximations that follow from the above conditions gives the result of a scatter pattern that is due to the interference between individual volume elements. The accuracy of this approximation is limited, Bayvel and Jones [2.12] suggest that for spheres better than 10% accuracy is only obtained for $m < 1.2$ and this range reduces as $x$ increases. In any case $x$ should be less than 12. Further analysis of the useful limits for different approximate scattering models is given in [2.13].

2.1.4. Anomalous Diffraction and Ray Optics

The Rayleigh and Rayleigh-Gans models are two size limits with the approximation $m = 1$. A third limit exists where particle size is large compared to the wavelength, that is

\[ x \gg 1 \]  \hspace{1cm} (2.5)

and

\[ |m - 1| \ll 1. \]  \hspace{1cm} (2.6)

Large particles will generally reflect and refract light strongly. However, if the relative refractive index is close to 1 the scattering will be weak and diffraction effects will be of the same order of intensity as the refraction. The overall scattered field will then be an interference between diffracted light and refracted rays which for the weakly scattering case are almost without change of direction. The result is known as anomalous diffraction.

As the refractive index increases for large particles, the refractive part of the scatter pattern becomes more separable from the diffracted part which becomes more concentrated in the forward direction. Ray optics can then be used to predict the scattering obtained from such a particle. This is not quite so simple as it may at first appear since account needs to be taken of multiple reflections and phase shifts at interfaces.
2.1.5. Application of Models to Systems of Objects

The above review of calculating the scattered field when particles are illuminated by a plane wave has only been concerned with single particles. When more than one particle is present the calculations become more difficult. Of the methods noted above only Rayleigh and Rayleigh-Gans can be simply extended to multiple particles in close proximity. If, however, there are a number of particles far enough apart such that interference effects are not observed, and the particles are from a monodispersion, then the general pattern of scattered light will be the same for the ensemble of particles as it was for individual particles. This holds true in the single scattering regime, that is when the light scattered by one particle is not significantly scattered or absorbed by other particles. This is a reasonable assumption at low concentrations of suspended matter \[2.14\]. Under such conditions the rough pattern of scattered light may be predicted but the intensity will vary with the number of particles present. When particles are close enough such that interference effects can be resolved then to estimate the scattered field it is necessary to resort to a method such as Rayleigh-Gans. For systems that are changing with time, as suspensions do (due to thermal motion if nothing else), then it is practically impossible to analytically calculate what the scattered light field will be from one moment to the next.

2.1.6. Application of Neural Networks to the Inverse Problem

It can be seen that there are serious practical difficulties in trying to calculate the scattering from a system of objects which is changing with time. In using scattered light to analyse that system we are attempting to work in reverse and calculate the physical state of the scattering media from the observed scattering. This is even more difficult because of the greater degree of freedom and because it is not a well defined problem; more than one system of scattering objects may give rise to the same scattered field.

Using neural networks to analyse the data circumvents many of the problems involved in trying to calculate a solution. A neural network may be trained to classify data sets based on experience, the training algorithm developing the internal structure of the neural network such that a chosen error metric is a minimum. The trained neural network performs a non-linear mapping of an input onto an output. It does not calculate the inverse solution but determines relationships between the input data and the desired output. As long as the inputs leading to different results are suitably dissimilar the neural network should learn a mapping. Because a full mathematical model describing the scattering process is not calculated the computational processing power required to form a solution is not excessive.
Another advantage of neural networks is that because the internal processing is distributed between neurons in the network they are generally good at making use of redundancy and hence are robust to errors in input data. This will be of using in overcoming sensor fouling.

In applying neural networks to the inverse scattering problem it was thought that different suspension size distributions would lead to observed scattering that was dissimilar enough for a neural network to identify. It was further hoped that differences in the optical properties of the suspended material would lead to structure in the scattered field that could be identified. Although it is desirable to be able to separate out the suspension size distribution and material, in the application investigated of distinguishing oil types, it was the combination of suspension size and material together that was used to identify the different oils.

### 2.2. Statistical Optics

In circumstances where the scattering is from a distribution of particles it is possible to model the scattering as a random process. This is an extension of random walk models that were originally applied to rough surface scattering and random phase screens [2.15][2.16][2.17]. The scattering from such media causes speckle patterns to be formed in the far field, the spatial characteristics of which depend on the physical form of the media. By measuring the autocorrelation length of the speckle pattern a correlation length describing any underlying periodicity or scale of the scattering media can be calculated [2.18]. When the medium is varying useful statistical data can still be obtained even though the speckle is changing. As long as the statistical properties of the medium remain constant the statistics of the speckle pattern will stay constant. For instance, the sea surface is varying constantly but the same piece of a calm sea surface viewed at two close intervals will look similar in that the waves will be of about the same size with the same interval between them. As a result the field scattered by the sea could be expected to have similar properties at each time although the detailed pattern would be different.

Statistical fluctuations in the intensity of scattered light have been investigated since the 1960s. Theories to explain non-Gaussian observations were developed during the 1970s. Much of the theory developed since then has concentrated on looking at surface scattering with the emphasis on applications of radar scatter from the sea. An early paper investigating the point statistics of scattering from suspensions was by Pusey, Schaefer and Koppel [2.19]. This presented probability distributions
and expected moments for scattering from suspensions of identical particles, assuming a finite fixed number and also a Poisson distribution of the number of particles. The scattering was shown to be non-Gaussian for finite numbers of particles. Jakeman and Pusey demonstrated in [2.16] by two theoretical methods that non-Gaussian effects may still be significant for surface scattering when the effective number of scatterers is large (i.e. tending to $\infty$). A random walk approach, assuming a random phase disturbance by the phase screen / surface, and a facet model were shown to be equivalent. Results from light scattering experiments using liquid crystals are given by Jakeman and Pusey in a follow up paper [2.20]. This compares experimental data to the theoretical predictions for normalised second moments of intensity, spatial and temporal correlations and the experimental geometry. Another Jakeman and Pusey paper [2.16] introduces the K distribution for the first time by extending the random walk model to have variable number of steps. Since the K distribution was proposed as a description of the single interval intensity statistics, the theory has developed to include a Generalised K distribution. The Generalised K distribution was introduced by Barakat [2.21] and later derived in a less model dependent manner by Jakeman and Tough [2.22]. The Generalised K distribution arises from a model in which the random walk has a directional bias, a model for weak scattering.

The remainder of this chapter describes the random walk model for strong scattering used in the papers noted above and derives the major results for single interval statistics. A similar method is used to derive the Gamma distribution applicable to incoherent sources. The description of probability density functions by their moments is then explained and some relevant relationships between moments are given. Finally the effects of finite numbers of scatterers and spatial correlations are discussed.

2.2.1. Random Walk Model

As well as the spatial characteristics of the speckle being governed by the scattering medium, the way in which the intensity varies at an observation point as the medium changes also depends on the surface / medium and how it is changing. A good physical conception of how these point statistics arise with suspensions can be gained from considering the particles in figure 2.5(a) uniformly illuminated from the left with coherent radiation. Each particle will scatter some radiation towards the detector, the actual amount depending on the particle size, shape, refractive index, radiation wavelength and the relative position of the detector. For each particle / scattering radiation, the field at the detector can be represented by an amplitude and complex phase:
a) Each illuminated particle scatters a different power towards the detector

b) Different intensities and phases represented in 2D by length and angle of phasor. The sum of contributions from all scatterers is the measured field.

Figure 2.5. Coherent detection of scattered light.
The resultant field at the detector will be the coherent sum of the contributions from all \( N \) particles that scatter:

\[
E_j = A_j e^{i\Theta_j}
\]

The field measured is therefore a sum of random variables, which due to the complex nature of the exponent term is known as a random walk in the complex plane, figure 2.5(b). In analysing the consequences of using such a model it is normal to make three assumptions. Firstly that the \( A_j \) all come from the same distribution, secondly that the distance between particles is greater than the wavelength of the source and thirdly that the \( A_j \) and \( \Theta_j \) are independent for all \( j \). The first assumption means that the central limit theorem may be applicable to the amplitude random variable and the second means that the probability distribution of phases can be assumed uniform over \( 2\pi \) radians. The second and third assumptions together mean that the resulting random walk will have no bias as to its direction.

\[ \text{2.2.1.1. Variation in Length of Random Walk} \]

There is another parameter in equation (2.8) that may be considered as a random variable and this is the number of particles, \( N \). In any real suspension the particles will be moving and therefore the number of particles in the observation volume could be changing. Since the number fluctuations will be governed by the fluid dynamics of the system some consideration is given here as to how the number of scatter centres may vary. The first point to note is that \( N \) must be discrete since only whole particles are possible. A physical idea of how \( N \) changes may be gained by imagining a suspension flowing through an observation volume as in figure 2.6. At a time \( t=0 \) there may be say \( N \) particles in the observation volume. At an interval later, \( t=1 \), the bulk flow will have moved the particles in suspension part way through the observation volume. If this time interval is made small enough only one of three things could have happened to the number of suspended particles in the test volume. All the particles may
have moved part way through the volume but none far enough to move out of the field of view and none far enough to move into the volume. The number of particles therefore remains as $N$. A second possibility is that there is just enough time for one particle to move out of the test volume (emigration), the number of particles observed now is $N-1$. The third possibility is that no particles move far enough to leave the test volume but one more enters (immigration), the number of observable particles is then $N+1$. To summarise, the number of suspended particles can either remain the same, increase by one or decrease by one. This immigration/emigration process is a type of Markov chain, illustrated by figure 2.7 where $\nu$ is the immigration rate and $\gamma$ is the emigration rate. As long as the chance of an emigration is larger than the chance of immigration $N$ does not increase indefinitely and the process is stable. There remains a finite probability of $N$ not being zero at any given moment the resulting probability distribution for $N$ is a Poisson distribution. Poisson number distributions for suspensions have long been accepted as a reasonable model [2.23].

![Diagram of birth/death/migration process](image)

**Figure 2.6. Illustration of birth / death / migration process.**

Box with solid border defines particles in observation volume at time $t=0$. At $t=1$ a different group of particles is observed because of the fluid flow (box with dashed border). In this example one particle has moved out of the observation volume (a death) between $t=0$ and $t=1$. 

24
There is evidence that not all of the particles in the observation volume will scatter strongly enough to be detected, due for example to shadowing [2.24]. This effect is more likely with non-spherical particulates which may need to be oriented at a specific angle to the detector to give a strong signature. In such cases it is the $N$ strongly scattering particles that are important. As in the above model there will still be an overall immigration / emigration of particles, some proportion of which will scatter strongly, but now there is also another mechanism by which the number of particles observed can change. Particles in the field of view may move and reorient such that they change from weak scatterers in the direction of the detector to strong scatterers in the direction of the detector (births), or they may change from being strong scatterers to weak scatterers (deaths). The number of deaths is assumed proportional to the overall number of particles visible ($N$) and the number of births is assumed proportional to the population that gave rise to the birth ($N-1$). The model is refined further by recognising that as the population in the field of view increases, the rate at which the general flow moves particles out of the observation area also increases. The emigration rate is therefore dependent on $N$ and may be included with the deaths. Immigration is not dependent on the current state of the observed area and is therefore still considered as a separate process. This scenario is illustrated in figure 2.8 where the probability of a state change to or from state $N$ is resolved into components due to births, deaths and immigration. $v$ is the immigration rate, $\lambda$ is the birth rate and $\mu$ is the death rate.

The transitions to the state $N$ are:

I. $\lambda(N-1)$ births from state $N-1$

II. $v$ immigrants from state $N-1$

III. $\mu(N+1)$ deaths from state $N+1$
The transitions from state $N$ are:

I. $\lambda N$ births from state $N$ to state $N+1$

II. $\nu$ immigrants from state $N$ to state $N+1$

III. $\mu N$ deaths from state $N$ to state $N-1$

![Diagram of birth/death/immigration process](image)

Figure 2.8. Birth / death / immigration process.

At each transition in the state of the system the number of particles may remain the same, increase with a probability $\nu$ plus a factor proportional to the previous state, $\lambda$, or decrease with a probability proportional to the previous state, $\mu$.

A rate equation can be formed from these different transitions and the probability of being in the relevant state.

$$\frac{dP_N(t)}{dt} = \mu(N+1)P_{N+1} + (\lambda(N-1) + \nu)P_{N-1} - ((\lambda + \mu)N + \nu)P_N$$

(2.9)

To find the number distribution arising from such a process, the equilibrium solution of this rate equation needs to be found. The rate equation can be analysed with the aid of the probability generating function, pgf, or Z transform [2.25] defined as

$$Q(z,t) = \sum_{N=0}^{\infty} z^N P_N(t)$$

(2.10)

Differentiating with respect to time reintroduces the rate equation.

26
\[ \frac{\partial Q(z,t)}{\partial t} = \sum_{N=0}^\infty z^N \frac{\partial P_N(t)}{\partial t} \]

\[ = \sum_{N=0}^\infty z^N \left( \mu(N+1)P_{N+1} - N\mu P_N \right) + \]
\[ + \sum_{N=0}^\infty z^N \lambda(N)P_{N-1} - NP_N \]  \hspace{1cm} (2.12)
\[ - \sum_{N=0}^\infty z^N \nu(P_{N+1} - P_N) \]

Expanding these series gives

\[ \frac{\partial Q(z,t)}{\partial t} = \mu(1-z)\left( P_1 + zP_2 + 2z^2P_3 + \ldots + N^2P_N + \ldots \right) + \]
\[ - \lambda(1-z)\left( P_1 + zP_2 + \ldots + N^{N-1}P_N + \ldots \right) + \]
\[ - \nu(1-z)\left( P_0 + zP_1 + \ldots + zNP_N + \ldots \right) \]

(2.13)

Noting that

\[ \frac{\partial Q(z,t)}{\partial z} = \frac{\partial}{\partial z} \left( P_0 + zP_1 + z^2P_2 + \ldots + z^NP_N + \ldots \right) \]

\[ = P_1 + 2zP_2 + 3z^2P_3 + \ldots + N^{N-1}P_N + \ldots \]

(2.14)

(2.15)

equation (2.13) simplifies to

\[ \frac{\partial Q(z,t)}{\partial t} = (1-z)(\mu - \lambda z)\frac{\partial Q(z,t)}{\partial z} - \nu(1-z)Q \]

(2.16)

a partial differential equation with constraint

\[ \sum_{N=0}^\infty P_N(t) = 1 = Q(z=1,t) \]

(2.17)

and assuming the process starts in the state \( M \)

\[ Q(z,t=0) = z^M \]

(2.18)
This pde can be solved using the method of characteristics [2.26] by letting \( \partial Q/\partial z = 0 \) and then \( \partial Q/\partial t = 0 \) and rearranging the two results to get the simultaneous result:

\[
\int \frac{\partial t}{t} = \int \frac{\partial z}{(\lambda z - \mu)(1-z)}
\]

\[
= \int \frac{\lambda \partial z}{(\lambda - \mu)(\lambda z - \mu)} + \int \frac{\partial z}{(\lambda - \mu)(1-z)}
\]

Hence

\[
t = \frac{\ln(\lambda z - \mu) - \ln(1-z)}{\lambda - \mu} + c_1
\]

and

\[
\frac{1-z}{\lambda z - \mu} \exp((\lambda - \mu)t) = C_1
\]

where \( C_1 \) is a constant. There must also be a simultaneous solution to

\[
\int \frac{\partial z}{(\lambda z - \mu)(1-z)} = \int \frac{-\partial Q}{\nu(1-z)Q}
\]

\[
\int \frac{\partial z}{\lambda z - \mu} = \int \frac{-\partial Q}{\nu Q}
\]

So

\[
\frac{\ln(\lambda z - \mu)}{\lambda} = -\frac{\ln Q}{\nu} + c_2
\]

hence

\[
(\lambda z - \mu)^{\nu/\lambda} Q^{1/\nu} = C_2
\]

The intersection of the two surfaces \( C_1 \) and \( C_2 \) is the general solution found by finding a function \( F \) that satisfies the boundary conditions such that

\[
(\lambda z - \mu)^{\nu/\lambda} Q^{1/\nu} = F\left(\frac{1-z}{\lambda z - \mu} \exp((\lambda - \mu)t)\right)
\]

It follows that

\[
Q(z,t) = \left[F\left(\frac{1-z}{\lambda z - \mu} \exp((\lambda - \mu)t)\right)(\lambda z - \mu)^{-\nu/\lambda}\right]^{\nu}
\]

which using the initial condition for \( Q(z,0) \) leads to
\[
F\left( \frac{1-z}{\lambda z - \mu} \right) = (\lambda z - \mu)^{\frac{\lambda}{\mu}}^{M/\nu} \tag{2.28}
\]

Substituting \( x = \frac{1-z}{\lambda z - \mu} \) leads to an expression for \( F(x) \) of

\[
F(x) = \left( \frac{\lambda - \mu}{1 + \lambda x} \right)^{\lambda} \left( \frac{1 + \mu x}{1 + \lambda x} \right)^{M/\nu} \tag{2.29}
\]

which when substituted back into equation (2.27) gives

\[
Q(z,t) = \left[ \frac{\lambda - \mu}{1 + \lambda \left( \frac{1 - z}{\lambda z - \mu} \right) e^{(\lambda - \mu)t}} \right]^{\frac{\lambda}{\mu}} \left( \frac{1 + \mu \left( \frac{1 - z}{\lambda z - \mu} \right) e^{(\lambda - \mu)t}}{1 + \lambda \left( \frac{1 - z}{\lambda z - \mu} \right) e^{(\lambda - \mu)t}} \right)^{M/\nu} \left( \lambda z - \mu \right)^{-\frac{\lambda}{\mu}} \tag{2.30}
\]

which reduces to

\[
Q(z,t) = \left( \frac{\lambda - \mu}{\lambda z - \mu + \lambda (1-z) e^{(\lambda - \mu)t}} \right)^{\frac{\lambda}{\mu}} \left( \frac{\lambda z - \mu + \mu (1-z) e^{(\lambda - \mu)t}}{\lambda z - \mu + \lambda (1-z) e^{(\lambda - \mu)t}} \right)^{M} \tag{2.31}
\]

The equilibrium solution is found as \( t \to \infty \). For a stable solution the death rate must be larger than the birth rate, in which case \( e^{(\lambda - \mu)t} \to 0 \) as \( t \to \infty \).

\[
\lim_{t \to \infty} Q(z,t) = \left( \frac{\lambda - \mu}{\lambda z - \mu} \right)^{\frac{\lambda}{\mu}} = g(z) \tag{2.32}
\]

\( g(z) \) can be rearranged into the form

\[
g(z) = (\mu - \lambda)^{\frac{\lambda}{\mu}} (\mu - \lambda z)^{-\frac{\lambda}{\mu}} \tag{2.33}
\]

\[
= \left( 1 - \frac{\lambda}{\mu} \right)^{\frac{\lambda}{\mu}} \left( 1 - \frac{\lambda z}{\mu} \right)^{-\frac{\lambda}{\mu}} = p^a (1 - (1 - p)z)^{-a} \tag{2.34}
\]

which by comparison with Hastings and Peacock [2.27] is the pgf for a negative binomial distribution with parameters \( \alpha = \nu/\lambda \) and mean \( N = \bar{N} = \nu/\mu - \lambda \). The probability function is described by
This negative binomial distribution simplifies to a Poisson when the birth rate equals zero. The birth / death / immigration model for \( N \) is therefore a more general one than the Poisson process described earlier. The parameter \( \alpha \) is sometimes called a bunching factor since it determines the nature of the number fluctuations about the mean and any number fluctuations look like a local bunching together of particles.

### 2.2.2. Special Cases of the Random Walk Model

With the random walk model all the physics of the complex light scattering problem is tied up in the random variables \( A_j, \Theta_j, \) and \( N \). The particle size and refractive index distributions and fluid dynamics are all described in some statistical sense by these random variables and the resulting electric field variations at the detector. It is obvious that the precise statistical distribution of intensity observed at a point will depend on a number of parameters, but by considering some special cases it is possible to predict some characteristics that may result.

#### 2.2.2.1. \( N \) Large and Constant

The first special case to consider is the simplest, that is with \( N \) large and constant. The random walk with \( A_j \) and \( \Theta_j \) can also be expressed as the sum of components along the real and imaginary axes, \( E_r \) and \( E_i \). With \( N \) tending to infinity and being constant the probability distribution of the sum of \( E_r \) and the sum of \( E_i \) will both be Gaussian by the central limit theorem and of zero mean following from the uniform distribution of \( \Theta_j \). These distributions can be written

\[
P(E_r) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp \left[ -\frac{E_r^2}{2\sigma^2} \right]
\]
and \[ P(E_j) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp \left( -\frac{E_j^2}{2\sigma^2} \right) \] (2.38)

where \( \sigma \) is the standard deviation and will depend on the particular distribution of \( A_j \). The joint probability is

\[ P(E_r, E_i) = P(E_r)P(E_i) \] (2.39)

since \( E_r \) and \( E_i \) are independent, and following on

\[ P(E_r, E_i) = \frac{1}{2\pi\sigma^2} \exp \left( -\frac{(E_r^2 + E_i^2)}{2\sigma^2} \right) \] (2.40)

To progress further we transform this pdf back into polar co-ordinates, \( A \) and \( \Theta \). This is done using the substitutions \( E_r = Acos\Theta \) and \( E_i = Asin\Theta \) together with the Jacobian of the transform which is \( A \). Hence

\[ P(A,\Theta) = \frac{A}{2\pi\sigma^2} \exp \left[ -\frac{(A^2cos^2\Theta + A^2sin^2\Theta)}{2\sigma^2} \right] \] (2.41)

\[ = \frac{A}{2\pi\sigma^2} \exp \left[ -\frac{A^2}{2\sigma^2} \right] \] (2.42)

Integrating with respect to \( \Theta \) over the range 0 to \( 2\pi \) gives the probability density function for the amplitude to be the Rayleigh distribution:

\[ P(A) = \frac{A}{\sigma^2} \exp \left[ -\frac{A^2}{2\sigma^2} \right] \] (2.43)

In most practical situations it is the intensity, \( I \), rather than the amplitude of a wave that is measured. The pdf of the intensity is found starting from \( P(A) \) as follows.

\[ P(I) = P(A^2) = P(A(I)) \frac{dA}{dl} \] (2.44)

Substituting in equation (2.43) \( A = \sqrt{I} \) leads to

\[ P(I) = \frac{\sqrt{I}}{2\sigma^2\sqrt{I}} \exp \left[ -\frac{I}{2\sigma^2} \right] \] (2.45)
which is a negative exponential distribution where the angle brackets denote the mean value.

The simple case of an isotropic random walk with a large, fixed number of steps, so called Gaussian statistics, leads therefore to a Rayleigh distribution of amplitudes and a negative binomial distribution of intensity. Physically this could model the scattered light from a large, fixed number of monodisperse particles in a uniform coherent field measured at a single point. Examples of Rayleigh and negative exponential distributions are shown in figure 2.9.

2.2.2.2. *N Large and Varying*

When *N* is varying the central limit theorem does not hold and a different analysis is required. Following the method of Jakeman and Pusey [2.28] we start by finding the characteristic function of an auxiliary random variable *U*, defined as [2.29]

\[
C(U) = \langle e^{iU} \rangle
\]  

such that *E* and *U* form a Fourier transform pair with

\[
E = E_r + iE_i = A\cos\Theta + iA\sin\Theta
\]  

and

\[
U = |U|\cos\Psi + i|U|\sin\Psi
\]  

It follows that the scalar product of *E* and *U* is

\[
E \cdot U = A\cos\Theta|U|\cos\Psi + A\sin\Theta|U|\sin\Psi
\]

\[
= |U| \sum_{j=1}^{N} A_j \cos(\Theta_j + \Psi)
\]
Figure 2.9. Examples of Rayleigh and negative exponential distributions. $y$ is the random variable, $P(y)$ the pdf, $\mu$ the mean and $\sigma$ the standard deviation.
The characteristic equation is then

\[ C_N(U) = \left\langle \exp \left( iU \sum_{j=1}^{N} A_j \cos(\Theta_j + \Psi) \right) \right\rangle \]  

(2.52)

\[ = \left\langle \prod_{j=1}^{N} \exp \left( iU |A_j| \cos(\Theta_j + \Psi) \right) \right\rangle \]  

(2.53)

\[ = \prod_{j=1}^{N} \left( \exp \left( iU |A_j| \cos(\Theta_j + \Psi) \right) \right) \]  

(2.54)

since all the random variables are independent. This characteristic equation includes an average over the phase distribution and amplitude distribution. The phase can be averaged since it is uniformly distributed over 2\(\pi\) and

\[ \left\langle e^{i\Theta} \right\rangle_\Theta = \int_0^{2\pi} \exp \left( iU |A_j| \cos(\Theta_j + \Psi) \right) P(\Theta) d\Theta \]  

(2.55)

\[ = J_0(U |A_j|) \]  

(2.56)

using the identity

\[ e^{i\alpha \cos \Phi} = J_0(\alpha) + 2 \sum_{k=1}^{\infty} i^k J_k(\alpha) \cos k\Phi \]  

(2.57)

where \(J_k(\alpha)\) is the Bessel function of order \(k\). Substituting (2.56) back into the characteristic equation

\[ C_N(U) = \prod_{j=1}^{N} \left( J_0(U |A_j|) \right)_{A_j} \]  

(2.58)

If the \(A_j\) are all from the same distribution then

\[ C_N(U) = \left( J_0(U |A_j|) \right)_{A_j}^N = \left( \int_0 \left[ J_0(U |A_j|) P(A_j) dA_j \right] \right)^N \]  

(2.59)

Assuming that the number fluctuations are negative binomial distributed, as the model introduced earlier predicted, the characteristic equation of the mean of \(N\), \(\bar{N}\), can be calculated:
To solve this $\langle J_0(|U|A_j) \rangle_{A_j}$ needs to be evaluated. This can be done using the Frobenius series [2.30] if mean $N$ tends to infinity. A simpler solution is found if $A_j$ is scaled by $\sqrt{N}$ to give a standard
deviation of $\langle A_j^2 \rangle$ as opposed to $\overline{N}\langle A_j^2 \rangle$. With these conditions the Bessel function averaged over all $A$
becomes

\[
\left\langle J_0(|U|A_j) \right\rangle_{A_j} = \int_0^\infty P(A_j) dA_j - \frac{1}{4} \int_0^\infty \frac{|U|^2 A_j^2}{N} P(A_j) dA_j + \frac{1}{2^6} \int_0^\infty \frac{|U|^4 A_j^4}{N^2} P(A_j) dA_j - \ldots
\]

\[
= 1 - \frac{|U|^2 \langle A_j^2 \rangle}{4N} + \frac{|U|^4 \langle A_j^4 \rangle}{2^6 N^2} - \ldots
\]

(2.65)

which is independent of the pdf of the amplitudes $P(A)$. Substituting equation (2.65) back into (2.64) gives for the characteristic function

\[
C_N(U) = \left[ 1 + \frac{\overline{N}}{\alpha} \left( 1 - \frac{|U|^2 \langle A_j^2 \rangle}{4N} + \frac{|U|^4 \langle A_j^4 \rangle}{2^6 N^2} - \ldots \right) \right]^{-\alpha}
\]

(2.66)

Which in the limit $\overline{N} \to \infty$
The pdf of $E$ can now be found using the inverse Fourier transform

$$P(E) = \frac{1}{(2\pi)^2} \int_{\tilde{U}} e^{-iE\tilde{U}} C(U) dU$$

$$= \frac{1}{(2\pi)^2} \int_{-\pi}^{\pi} \int_{0}^{\infty} \exp(-iE\tilde{U}) \frac{\tilde{U}}{1+|\tilde{U}|^2 \left\langle \frac{A_j^2}{4a} \right\rangle} |\tilde{U}| d\tilde{U} d\tilde{Y}$$

$$= \frac{1}{2\pi} \int_{0}^{\infty} \frac{\tilde{U} L_0(|\tilde{U}|A)}{1+|\tilde{U}|^2 \left\langle \frac{A_j^2}{4a} \right\rangle} d\tilde{U}$$

Integrating this function with respect to $U$ results in

$$P(E) = \frac{A^{a-1}}{2^{a-2}\Gamma(a)} \left( \frac{4a}{\left\langle A_j^2 \right\rangle} \right)^{\frac{1+a}{2}} \text{K}_{a-1} \left( A \left( \frac{4a}{\left\langle A_j^2 \right\rangle} \right)^{\frac{1}{2}} \right)$$

where $K$ is a modified Bessel function and $\Gamma$ is the Gamma function. The pdf of the amplitude can now be obtained by integrating over all values of the phase $\Theta$ and since the amplitude and phase are independent $P(A)$ is simply

$$P(A) = \int_{0}^{2\pi} AP(E) d\Theta = 2\pi AP(E)$$

By substituting

$$b = \left( 4a/\left\langle A_j^2 \right\rangle \right)^{\frac{1}{2}}$$

$$P(A) = \frac{A^a b^{i+a}}{2^{a-1} \Gamma(a)} K_{a-1}(bA)$$

$$= \frac{2b}{\Gamma(a)} \left( \frac{bA}{2} \right)^{\alpha} K_{a-1}(bA)$$
which has the form of a $K$ distribution and not Rayleigh even though the mean value of $N$ is infinite. The pdf of the intensity is found by the same method as used for transforming between amplitude and intensity with fixed $N$. Performing this transformation leads to

$$P(t) = \frac{b}{\Gamma(\alpha)} \left( \frac{b}{2} \right)^\alpha \sqrt{\frac{\alpha - 2}{\alpha}} K_{\alpha - 1}(b\sqrt{t})$$  \hspace{1cm} \text{(2.76)}$$

Examples of these distributions are given in figure 2.10. It can be seen that as the parameter $\alpha$ tends to infinity, the $K$ distribution tends to the Rayleigh distribution. This is clear empirically since as the bunching of particles becomes less severe, the number fluctuations will reduce until the stage is reached at $\alpha = \infty$ when the number of particles is constant and Gaussian statistics are recovered.

2.2.2.3. Scattering using an Incoherent Source

So far consideration has been given to the case of coherent illumination only. If an incoherent source is used the type of statistics predicted are different. This difference stems from the different model that needs to be used. Where before the measured intensity was the square of the coherent sum of complex amplitudes, with incoherent radiation the measured intensity is the sum of individual intensities

$$I = \sum_{j=1}^{N} A_j^2$$  \hspace{1cm} \text{(2.77)}$$

This is a real random walk and since $I$ is always positive the characteristic equation for this process is effectively the Laplace transform

$$C_N(s) = \int_I e^{st} P(t) dt = \langle e^{lt} \rangle$$  \hspace{1cm} \text{(2.78)}$$

$$= \prod_{j=1}^{N} \exp(sA_j^2)$$  \hspace{1cm} \text{(2.79)}$$

$$= \left( \exp(sA_j^2) \right)^N$$  \hspace{1cm} \text{(2.80)}$$

Following the same method as for the coherent case and assuming $N$ is negative binomial distributed

$$C_{\bar{N}}(s) = \sum_{N=0}^{\infty} P_N C_N(s) = \frac{1}{\left(1 + \bar{N}/\alpha - \bar{N}/\alpha \left( \exp(sA_j^2) \right) \right)^\alpha}$$  \hspace{1cm} \text{(2.81)}$$

37
Figure 2.10. Examples of K distributions and associated intensity distributions. 
y is the random variable, P(y) the pdf and α the bunching factor.
The mean of the exponential may be expanded as a series

$$\langle \exp(sA_j) \rangle = 1 + s\langle A_j^1 \rangle + \frac{s^2}{2!}\langle A_j^2 \rangle + \ldots + \frac{s^n}{n!}\langle A_j^n \rangle + \ldots$$  \hspace{1cm} (2.82)

Normalising such that $A_j \rightarrow A_j/\sqrt{\bar{N}}$ leads to

$$C_{\bar{N}}(s) = \left(1 + \frac{s\langle A_j^2 \rangle}{\alpha} + \frac{s^2\langle A_j^4 \rangle}{2\alpha\bar{N}} + \ldots\right)^{-\alpha}$$  \hspace{1cm} (2.83)

$$= \left(1 + \frac{s\langle A_j^2 \rangle}{\alpha}\right)^{-\alpha}$$  \hspace{1cm} (2.84)

as $\bar{N} \rightarrow \infty$. The pdf of $I$ is found by taking the inverse Laplace transform, resulting in [2.31]

$$p(I) = \frac{I^{\alpha-1}e^{-\alpha I}}{\Gamma(\alpha)\langle A_j^2 \rangle^\alpha} \exp\left(-\frac{\alpha I}{\langle A_j^2 \rangle}\right).$$  \hspace{1cm} (2.85)

This is the Gamma distribution. It is the distribution intensity fluctuations are expected to match under the current assumption of $\bar{N} \rightarrow \infty$ with a negative binomial distribution of $N$. When the parameter $\alpha$ equals 1, the Gamma distribution reduces to the negative exponential associated with Gaussian statistics.

### 2.2.3. Description of Distributions by their Moments and Important Relationships

Probability distributions may be described by their moments, providing a more practical method of comparing distributions than fitting to pdfs when real data are involved. To fully describe a distribution would take an infinite number of moments but a number of the lower order moments can describe the basic form of the distribution. The $r$th moment about the origin may be calculated as the mean of the random variable raised to the power of $r$:

$$M^r = \langle y^r \rangle = \int_{-\infty}^{\infty} y^r p(y)dy$$  \hspace{1cm} (2.86)

The first order moment will be recognised as the mean. It is sometimes useful to take moments about the mean:
which, when \( r=2 \), is the variance with \( \bar{y} \) equal to the mean or first moment. Another method of obtaining a description of the distribution unbiased by the mean, is to normalise the moments with respect to the mean.

\[
y^{(r)} = \frac{\langle y^r \rangle}{\langle y \rangle^r}
\]

The relationships that exist between the normalised moments for different distributions will be referred to later so it is worthwhile introducing those relationships for the distributions already encountered. Although the moments may be calculated as stated above, it is often easier to use the moment generating function.

\[
\Phi(t) = \int_{-\infty}^{\infty} e^{ty} p(y) dy
\]

The \( r \)th moment may be obtained by evaluating \( \frac{d^r \Phi}{dt^r} \) at \( t=0 \). Take as an example the Normal distribution with pdf

\[
p(y) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{(y-\bar{y})^2}{2\sigma^2}\right)
\]

and moment generating function

\[
\Phi(t) = \exp\left(\bar{y}t + \frac{\sigma^2 t^2}{2}\right)
\]

Differentiating with respect to \( t \)

\[
\Phi'(t) = (\bar{y} + \sigma^2 t) \exp\left(\bar{y}t + \frac{\sigma^2 t^2}{2}\right)
\]

\[
\Phi''(t) = \sigma^2 \exp\left(\bar{y}t + \frac{\sigma^2 t^2}{2}\right) + (\bar{y} + \sigma^2 t)^2 \exp\left(\bar{y}t + \frac{\sigma^2 t^2}{2}\right)
\]

\[
\Phi'''(t) = \left(\sigma^2 (\bar{y} + \sigma^2 t) + 2\sigma^2 (\bar{y} + \sigma^2 t)^2 + (\bar{y} + \sigma^2 t)^3\right) \exp\left(\bar{y}t + \frac{\sigma^2 t^2}{2}\right)
\]

and evaluating at \( t=0 \)
\( \Phi(0) = M^1 = \bar{y} \)  \hspace{1cm} (2.95)

\( \Phi'(0) = M^2 = \sigma^2 + \bar{y}^2 \)  \hspace{1cm} (2.96)

\( \Phi''(0) = M^3 = 3\sigma^2 \bar{y} + \bar{y}^3 \)  \hspace{1cm} (2.97)

These moments when normalised are

\[
y^{[2]} = 1 + \frac{\sigma^2}{\bar{y}^2}
\]

\( y^{[3]} = 1 + \frac{3\sigma^2}{\bar{y}^2} \)  \hspace{1cm} (2.99)

It can be noted that there exists a particular relationship between the second and third normalised moments which holds for any Normal distribution, that is

\[
y^{[3]} = 3y^{[2]} - 2
\]  \hspace{1cm} (2.100)

Other distributions also have particular relationships between moments. For the \( K \) distribution with parameter \( \alpha \) the \( r \)th moment is

\[
y^{[r]} = \frac{r! \Gamma(r + \alpha)}{\alpha^r \Gamma(\alpha)}
\]  \hspace{1cm} (2.101)

and the relationship between normalised second and third moments turns out to be

\[
y^{[3]} = 3\left(y^{[2]} - y^{[1]}\right)
\]  \hspace{1cm} (2.102)

which is dependent only on the parameter \( \alpha \). For the Gamma distribution the normalised moments are

\[
y^{[r]} = \frac{\Gamma(r + \alpha)}{\alpha^r \Gamma(\alpha)}
\]  \hspace{1cm} (2.103)

and \( y^{[3]} = 2\left(y^{[2]}\right)^2 - y^{[2]} \)  \hspace{1cm} (2.104)

which again is only dependent on \( \alpha \).
An interesting relationship between moments exists for the negative exponential distribution which has normalised moments

\[ y^{[r]} = r! \]  

(2.105)

The normalised moments of the negative exponential distribution are therefore fixed. This is reflected in the pdf being parameterised in terms of the mean only. Intensity moments from a Gaussian scattering process will always have second and third normalised moments of 2 and 6 respectively.

The relationship between normalised second and third moments for the negative binomial distribution does depend on a process parameter. Using the notation of Hastings and Peacock, this relationship is

\[ y^{[3]} = (1+\eta)(y^{[2]})^2 \]  

(2.106)

or in the notation used earlier in relation to birth / death / migration

\[ y^{[3]} = \left(1+\frac{\lambda}{\mu}\right)(y^{[2]})^2 \]  

(2.107)

These relationships between moments will be used later to judge how closely experimental measurements match theory, in particular equations (2.100), (2.102), (2.104) and (2.105).

2.2.4. Finite N and Cross Correlations

The models described show that even though the mean number of scatter centres is on average infinite, if the number is varying then non-Gaussian statistics will arise. The precise statistics observed will depend on the process by which the number fluctuations occur but the simple model of birth / death / migration indicates that the K and Gamma distributions are of particular interest. Not covered so far is the more realistic case of a finite average number of particles. In that case non-Gaussian statistics are even more likely, the higher order terms in the expansion of the characteristic equation remaining relevant. The intensity pdfs that arise from negative binomial number fluctuations are called the generalised K for the coherent source, and the (tentatively named) modified Gamma distribution for the incoherent source. Complete treatments can be found in the work of Hopcraft [2.32] but an interesting result is derived here for the simpler incoherent source case. If the mean number of particles is large but not infinite then the terms up to the order of \( s^2 \) in (2.83) will be relevant. The inverse Fourier or inverse Laplace transforms of this characteristic equation are not easily obtained. A complication is
added by the need to include a delta function to account for the finite probability of no particles being present. Even though the pdf is not simply obtained, the moments may be obtained from (2.83) using the method (c.f. (2.89) to (2.97))

\[
\langle \eta \rangle = \left. \frac{1}{i^\eta} \frac{dC(t)}{dt} \right|_{t=0} \quad (2.108)
\]

Since in (2.83) \( s \) includes the complex term \( s = -it \) it follows that

\[
\frac{dC_N(t)}{dt} = -\alpha \left( 1 - \frac{i\langle A_j^2 \rangle}{\alpha} + \frac{i^2 \langle A_j^4 \rangle}{2N\alpha} \right) \left( \frac{-i\langle A_j^2 \rangle}{\alpha} + \frac{t \langle A_j^4 \rangle}{N\alpha} \right) \quad (2.109)
\]

and evaluated at \( t=0; \)

\[
\langle t \rangle = \langle A_j^2 \rangle \quad (2.110)
\]

\[
\frac{d^2 C_N(t)}{dt^2} = \alpha(\alpha + 1) \left( 1 - \frac{i\langle A_j^2 \rangle}{\alpha} + \frac{i^2 \langle A_j^4 \rangle}{2N\alpha} \right) \left( \frac{-i\langle A_j^2 \rangle}{\alpha} + \frac{t \langle A_j^4 \rangle}{N\alpha} \right)^2 - \alpha \left( 1 - \frac{i\langle A_j^2 \rangle}{\alpha} + \frac{i^2 \langle A_j^4 \rangle}{2N\alpha} \right) \frac{\langle A_j^4 \rangle}{N\alpha} \quad (2.111)
\]

\[
\frac{d^2 C_N(t)}{dt^2} \bigg|_{t=0} = \frac{-(\alpha + 1)\langle A_j^2 \rangle^2}{\alpha} - \frac{\langle A_j^4 \rangle}{N} \quad (2.112)
\]

The normalised second moment is therefore

\[
\langle t^2 \rangle = \left( 1 + \frac{1}{\alpha} \right) + \frac{\langle A_j^4 \rangle}{N \langle A_j^2 \rangle^2} \quad (2.113)
\]

The amplitude statistics in the second term of (2.113) are a function of the angle of observation whilst in the first term \( \alpha \) is a parameter of the number fluctuation process and is thus isotropic, assuming that observations at different positions view the same scattering volume.

At the beginning of this chapter it was pointed out that the amount of light scattered in any particular direction depended on a number of parameters such as wavelength, particle size, relative refractive index etc, and that a knowledge of the scattering in several directions could lead to some estimate of the scattering system's physical condition even though a full inverse solution is not practical. Mapping out the scattered intensity around a scattering volume and comparing relative intensities at different angles is a type of (non-normalised) correlation which assumes that the amount of light scattered in all
directions is linked. This idea can be extended to look at the correlations between angles in a more statistical sense at different angles. Extending the incoherent source model with large but finite number of particles to study this correlation uses a joint variable characteristic equation.

\[ C_N(t_1, t_2) = \left\langle \exp\left[ it_1 A_1^2(\theta_1) + it_2 A_2^2(\theta_2) \right] \right\rangle \]

\[ = \left\langle \prod_{j=1}^{N} \exp\left[ it_1 A_j^2(\theta_1) + it_2 A_j^2(\theta_2) \right] \right\rangle \]

Using the same method as earlier the characteristic equation for a mean \( N \) is found to be

\[
C_N(t_1, t_2) = \left( 1 - \frac{it_1 \langle A_1^2(\theta_1) \rangle + it_2 \langle A_2^2(\theta_2) \rangle}{\alpha} \right)
+ \frac{t_1^2 \langle A_1^2(\theta_1) \rangle + t_2^2 \langle A_2^2(\theta_2) \rangle + 2t_1t_2 \langle A_1^2(\theta_1) A_2^2(\theta_2) \rangle}{2N\alpha} \right)^{\alpha}
\]

Joint moments are now found by taking the partial derivatives:

\[ \left\langle I^m(\theta_1) I^n(\theta_2) \right\rangle = \frac{1}{(m+n)!} \frac{\partial^{m+n} C(t_1, t_2)}{\partial t_1^m \partial t_2^n} \bigg|_{t_1=t_2=0} \]

Following this method to obtain \( \left\langle I(\theta_1) I(\theta_2) \right\rangle \) gives

\[
\left. \frac{\partial C_N(t_1, t_2)}{\partial t_1} \right|_{t_1=0} = -\alpha C_N(t_1, t_2) C_N(t_1, t_2) \left( \frac{-i\langle A_1^2(\theta_1) \rangle}{\alpha} + \frac{t_1 \langle A_1^2(\theta_1) \rangle + t_2 \langle A_2^2(\theta_1) A_2^2(\theta_2) \rangle}{N\alpha} \right)
\]

\[
\left. \frac{\partial^2 C_N(t_1, t_2)}{\partial t_1 \partial t_2} \right|_{t_1=0} = \alpha(\alpha + 1) C_N(t_1, t_2) C_N(t_1, t_2) \left( \frac{-i\langle A_1^2(\theta_2) \rangle}{\alpha} + \frac{t_1 \langle A_1^2(\theta_2) \rangle + t_2 \langle A_2^2(\theta_1) A_2^2(\theta_2) \rangle}{N\alpha} \right)
\times \left\langle \frac{\langle A_1^2(\theta_2) \rangle}{\alpha} + \frac{t_2 \langle A_1^2(\theta_2) \rangle + t_1 \langle A_2^2(\theta_1) A_2^2(\theta_2) \rangle}{N\alpha} \right\rangle
\]

Evaluated at \( t_1 = t_2 = 0 \) this simplifies to

\[
\left. \frac{\partial^2 C_N(t_1, t_2)}{\partial t_1 \partial t_2} \right|_{t_1=t_2=0} = \alpha(\alpha + 1) \frac{i\langle A_1^2(\theta_1) \rangle i\langle A_1^2(\theta_2) \rangle}{\alpha^2} - \alpha \langle A_2^2(\theta_1) A_2^2(\theta_2) \rangle \frac{1}{N\alpha} \]

(2.120)
Thus

\[
\langle I(\theta_1) I(\theta_2) \rangle = \frac{(\alpha + 1) \langle \mathcal{A}_1^2(\theta_1) \mathcal{A}_2^2(\theta_2) \rangle}{\alpha} + \frac{\alpha \langle \mathcal{A}_1^2(\theta_1) \mathcal{A}_2^2(\theta_2) \rangle}{N} \tag{2.121}
\]

Normalising leads to a type of normalised correlation function

\[
N^{[2]} = \frac{\langle I(\theta_1) I(\theta_2) \rangle}{\langle I(\theta_1) \rangle \langle I(\theta_2) \rangle} = \left(1 + \frac{1}{\alpha} \right) + \frac{\langle \mathcal{A}_1^2(\theta_1) \mathcal{A}_2^2(\theta_2) \rangle}{N \langle \mathcal{A}_1^2(\theta_1) \rangle \langle \mathcal{A}_2^2(\theta_2) \rangle} \tag{2.122}
\]

This result is similar in form to (2.113) consisting of an isotropic part and a "scattering fluctuation" part. The numerator of the fluctuating part can not be measured directly since the individual \( \mathcal{A}_j \) are not separable at the detector, but as \( \theta_1 \) and \( \theta_2 \) become widely separated then the fluctuating term is expected to decorrelate and tend in value to the order of \( 1/N \):

\[
\theta_1 \gg \theta_2 : \quad \frac{\langle \mathcal{A}_1^2(\theta_1) \mathcal{A}_2^2(\theta_2) \rangle}{N \langle \mathcal{A}_1^2(\theta_1) \rangle \langle \mathcal{A}_2^2(\theta_2) \rangle} \to O(1) \tag{2.123}
\]

This approximation does not lead to a set of equations solvable by measuring the intensity correlations and point statistics, there are too many unknowns, but does give a useful approximation for \( N \) when the number fluctuations are Poisson. In the Poisson limit of the negative binomial distribution (birth rate equal to zero) \( \alpha \) tends to infinity. Combining this with the approximation (2.123) suggests that for Poisson number fluctuations

\[
N^{[2]} \approx 1 + \frac{1}{N} \tag{2.124}
\]

Under the specific conditions of Poisson number fluctuations an estimate of the mean number of particles can therefore be made and the point statistics can then be used to investigate the physical nature of the scattering media.

### 2.3. Chapter Summary

The electromagnetic scattering from a single particle situated in an incident plane wave may be calculated using Mie theory. For some conditions approximations are valid which make simplified calculations possible. When extended to the calculation of scattering from multiple particles in motion, both Mie theory and the simplified models become too complicated for practical use. The inverse
problem, determining what system of particles gave rise to an observed scattered field, is even more
difficult.

In dilute suspensions the scattering is said to be in the single scattering regime. This is because the
separation between individual particles is large enough such that interference between the fields
scattered from each individual particle is not resolved. The detector therefore measures the sum of
intensities scattered from each particle. As a result the intensity detected depends almost linearly on the
number of scattering particles present. If the particles are monodisperse the spatial scattering pattern
will also be the same for the ensemble scattering as it is for the individual particles. In the single
scattering regime it is expected that artificial neural networks will have some success in providing an
inverse solution, characterising light scattering media in terms of its concentration, size distribution and
material.

Because of the complexity of calculating the field scattered from a complex varying system it is simpler
to describe the scattering in a statistical sense. This may be done using random walk models of the
scattering process. These models lead to predictions of the observed light intensities being random of
particular probability distributions. The most important predictions with regard to this research are

1. A coherent source illuminating a dense but varying suspension will produce scattered light
   intensities matching a K distribution.

2. An incoherent source illuminating a dense but varying suspension will produce scattered light
   intensities matching a Gamma distribution.

3. An incoherent source illuminating a varying suspension of finite number of particles will produce
   scattered light which may be described by the combination of an isotropic parameter, dependent on
   the variation of the suspension, and a non-isotropic parameter, dependent on the mean number of
   particles and the physical properties of those particles.

Statistics of experimental data may be compared to the theoretical predictions using the relationships
between moments for different distributions given in equations (2.100), (2.102), (2.104) and (2.105).
3. System for Nephelometric Experiments

The system built for performing on-line experiments consists of several components. As well as the actual pipeline flow system on which measurements were made there are the sensors and the optoelectronics. The optoelectronics can be split into the light sources and drivers, and the light detectors and preamplifiers. Power supplies, data acquisition and synchronisation electronics are also needed to complete the system. All of these components were designed and built during the course of this research. A description of these components is given.

3.1. Flow System

The pipeline flow system on which experiments were performed is pictured in figure 3.1. The pipeline is made from PVC tubing with an internal diameter of approximately 37mm. The external diameter is 50.8mm (2 inches). The wall thickness is greater than commonly used to allow the walls to have screw threads tapped if required. The pipeline is divided into a number of sections joined by unions which can be unscrewed by hand. This makes it particularly easy to add and remove sensor modules which have been designed to be of a fixed length (160mm prior to the unions being fitted) making them fully interchangeable. A heavy duty central heating pump pumps water around the system. This pump has three speed settings and a ball valve situated after the pump allows fine control of the flow speed. The flow speed can be monitored using a Kytola variable area flowmeter. Two meters which can be interchanged are needed to cover the full operating range of the system. One operates over the range 10 litres/minute to 50 litres/minute, the other from 50 litres/minute to 300 litres/minute. To swap the flowmeters the glass injection tube section first needs to be removed from the upflow section to give extra space for slipping out the flowmeter which has a nut extending beyond its body section. The glass injection tube is a section with a blown glass centre and branch at 90° to the flow. At the end of this side branch is a rubber Subaseal through which contaminants (fine suspensions or liquids) can be injected. The T port valves allow the flow to be either sent up into the header tank and across the crosspiece or to kept in a closed flow just around the lower loop. An arrow embossed on the valves indicates which ports are open, flow is in the direction of the arrow and will be forced to turn 90° at the valve, hence the crosspiece is always connected to either the header tank circuit or the closed loop.
Figure 3.1. Flow system for on-line experiments.
circuit. When the arrow points towards the supporting frame all three ports are open. The bottom T port valve provides a means of draining the system.

The best way of filling the system was found to be to fill up the header tank until the water starts to over-flow and then to turn off the tap. The top right valve is then turned such that water flows through the cross-piece and down the right hand down tube. When the water has finished draining from the top left side of the header tank the top left valve is half opened, so that the arrow points towards the frame, to release the air block in the flow loop. The top right valve is then opened slightly to allow water to slowly fill the flow loop. Towards the end of the filling process the valve may be opened fully. With the flow speed control valve wide open the pump is then started to pump the water around and expel any air blocks into the header tank. The flow speed is then reduced and the top left valve closed to force the flow through the cross-piece. Finally the top right valve is closed to seal off the closed loop circuit. Draining the system is achieved by opening up the two top valves and then opening the drain valve and letting the water flow out. To aid in cleaning, the header tank is normally topped up with clean water first and the route of the clean water as it drains through the system is varied to flush out the cross-piece.

The sections of the pipeline housing the sensors are in the downpipe where the flow is expected to be the least turbulent. It also ensures that the pipe is full at the sensor positions, a horizontal section could possibly only be partially full of water if there was air trapped in the system.

The capacity of the closed flow loop was measured to be 5.83 litres. This figure comes from an average of five individual measurements of capacity made using calibrated flasks and a measuring cylinder. The individual measurements were 5.820 litres, 5.803 litres, 5.812 litres, 5.837 litres and 5.863 litres, a spread of approximately 1% of the capacity.

3.2. Nephelometer Sensor Heads

Two nephelometer sensor heads have been designed and built to fit into the pipeline flow system. Both are fibre optic based to provide some flexibility in the sources and detectors that can be used.
3.2.1. *Mk I Nephelometer*

The first to be built is shown in figure 3.2. There are six detection angles azimuthally around the pipeline: 45°, 90°, 135°, 180° or line of sight, 240° and 300° as measured from the launch fibre. Eska plastic optical fibres are used to collect scattered light and carry it to the detectors. The fibres are connected to the pipeline using SMA connectors and sealed using silicon rubber compound. Care needs to be taken when fitting the fibres to keep the silicon rubber off the fibre ends. The fibres themselves are glued into the SMA connectors using epoxy resin; cyanoacrylate (superglue) is soluble. The fibre optics have a large numerical aperture, 28° half angle, and a large core of 0.998 mm diameter so they can collect a large proportion of the light scattered. For the same reason they are not very directional. This is good in the sense that mechanical alignment is not critical, but is a drawback in that they will not provide measurements of angular scattering to any great degree of resolution.

Light is launched into the sensor head via a fibre optic. A micro lens (Omron part E39-F1) is used to give some collimation, a matching gel is placed between the fibre end and the micro lens to reduce the reflection at the interface and to stop condensation forming on the lens surface. A diagram of the launch assembly is shown in figure 3.3.

3.2.2. *Mk II Nephelometer*

The second nephelometer design is more sophisticated, including optics on the launch and receive ports. The aim of using optics is to give better control over the collimation and focusing of the source and to improve the directivity of the receivers. This second design is shown in figures 3.4 and 3.5. As with the first design the detection angles are placed symmetrically around the pipeline although in this instance the angles are 55°, 125°, 180° (line of sight) and 270°. The reduced number of detection angles is necessary because of the physical size of the optics.

The optics are built as removable probes which fit into the sensor section through Cajon UltraTor compression fittings (part number SS-8-UT-1-6) which have been drilled out to the desired internal diameter of 12 mm. The end of the fittings have been spot faced so that glass windows can be glued in place if wanted. To date this has been done with the launch port only, using 13 mm diameter glass cover slips. These cover slips will take the pressure of a full head of water in the flow system but care needs to be taken when inserting the optical probes since the cover slips are easily broken. Care should also
Figure 3.2. MkI nephelometer design.

Figure 3.3. Launch fibre assembly for MkI nephelometer.
Figure 3.4. MkII nephelometer design.
Figure 3.5. MkII nephelometer showing removable probes.

Figure 3.6. Fixed focal length optical probe.
be taken to try and avoid condensation forming on the glass. A technique which has proved useful is to leave the water circulating in a closed loop for a period prior to inserting the launch probe, allowing the water to reach a more stable temperature before starting measurements.

Various light sources can be used with or without launch optics. Laser diodes can be used with commercially available optics to control the focal distance as long as the diameter of the unit is 12mm or slightly less. A fixed focal length launch probe, shown in figure 3.6, has been built which can be used to launch light from a fibre into the sensor. This probe uses two plastic aspheric lenses to focus light from a fibre terminated with a SMA connector. The lenses are manufactured by COIL (Combined Optical Industries Ltd). The probe design is such that the tip of the fibre is at the focal length of one lens, such that the source light forms a parallel beam to the second lens. The second lens then focuses the beam at its focal length. The focal length is suitable to focus light at the centre of the sensor. The gain of the probe in air is 2.08, that is if a fibre of core diameter 1mm is used as a source, an image 2.08mm in diameter will be formed at the focal length of the probe.

The receiver optics are more complicated because it was decided that being able to vary the numerical aperture would be desirable. The design uses three lenses (Melles Griot part numbers 01LPX037, 01LPX009 and 01LPK004), two being held at a fixed distance from each other and the collecting fibre, and the third being movable to vary the overall gain / numerical aperture of the probe. A glass or acrylic window in the end of the probe and a rubber O ring seals the unit to water ingress. Rubber O rings are also used to protect the lenses from damage by the clamping rings. Figure 3.7 shows the design. An optical fibre with SMA connector screws onto the end of the probe. The fibre tip is at the focal distance from lens C (15mm) so that light rays parallel to the probe axis will be focused onto the fibre. The focal length of the probe overall then depends on the relative positions of lenses A and B, the magnification of the lens system being equal to the ratio of front and back focal lengths or numerical apertures [3.1]. Lens A has a focal length of -15mm and lens B has a focal length of 25.4mm. The amount of travel allowed to lens A in the design should make the focal distance of the combination variable between 10mm and infinity in front of lens A.

Once built the receiver optics were found to give the desired wide operating range of numerical aperture. However, the amount of light collected by the receivers in nephelometric experiments was less than that collected by the bare fibres, despite having a larger aperture of 5mm diameter. This is probably due to the following reasons. Firstly, to keep the cost low, lenses without any anti-reflection coatings were used. The three lenses and glass window give a total of one water/plastic interface, one air/plastic interface and seven air/glass interfaces. This compares with the one water/plastic interface for the bare fibre. Each of these interfaces will produce a loss due to Fresnel reflections of approximately 4% of the incident power [3.2]. After seven transitions from air to glass or glass to air
Figure 3.7. Receiver optics.
only about 75% of the initial light will still be transmitted. Although the numerical aperture of the probes can be varied over a large range, there is little variation in the amount of light received as the numerical aperture is changed. This could well be due to lens misalignment and reflections off the brass construction of the probes. Alignment is only by placing the lens in a chamfered hole to encourage it to centre itself and there are no pupils to block reflected light. The design was not computer aided but made by hand using the combination lens equation

\[(3.1) \quad f = \frac{f_A f_B}{f_A + f_B - d}\]

where \(f_A\) and \(f_B\) are the focal lengths of two lenses a distance \(d\) apart, \(f\) the combination focal length. A full ray tracing analysis was not made to check where all light from a diffuse source would propagate to. Performance could possibly be improved by adding pupils to the design but it is hard to come to any firm conclusions without a full analysis by a ray tracing program.

The receiver probes succeed in one of their aims at least, that is to reduce the angular range of scatter that is detected by a single fibre, enabling measurements to a greater angular resolution to be made.

### 3.3. Light Sources

There are a number of considerations to be made when choosing what type of light source to use in any optical application. These are:

1. Output power
2. Coherence
3. Ease of coupling
4. Wavelength
5. Optical bandwidth
6. Electrical bandwidth
7. Matching with spectral response of detector
8. Ease of modulation
9. Cost
The most relevant of these points with regard to this research are mentioned below.

For the experiments described in this thesis a high optical source power was desired to give the best possible signal to noise ratio. However, it is not really the source power that is important, but how much of that power is coupled to the detector. This depends on how the light is emitted spatially from the source and on wavelength dependent factors. A light source with a low numerical aperture is generally easier to couple to the desired area.

Absorption is wavelength dependent in water and in the optical fibres used to carry light to and from the nephelometer sensor. Choosing a wavelength that is not absorbed strongly will obviously increase the optical power at the detectors. Graphs of the absorption at different wavelengths for both water and the optical fibres used are shown in figure 3.8. Data for the absorption in distilled water came from Sullivan [3.3]. Other studies extending into the infra red and ultra violet regions have been made. [3.4][3.5][3.6]. This last reference also indicates some variation with temperature. Data for the fibre comes from the manufacturer's data sheet.

![Figure 3.8. Attenuation of light in plastic fibre and distilled water.](image)
The absorption spectrum of the fibre bears some resemblance to that of water. This is probably due to water in the fibres having a major influence on the transmission performance; glass fibres are usually made in dry conditions to improve attenuation characteristics. In any case, due to the light's path length being shorter in the water than in the fibre, it is the attenuation in the fibre that is more important. The best transmission performance would be obtained at a wavelength of about 550nm, although any wavelength under 660nm is transmitted fairly well. There is also a narrow transmission window at around 760nm. Using the longer wavelengths is normally desirable since it becomes harder to find high power sources at short wavelengths.

In scattering applications the size of the scatterers in relation to the wavelength determines the amount of light scattered in each direction. If the wavelength is relatively long compared to the particle size the scattering will be Rayleigh and information about the size of the particles, other than that they are small, will not be recoverable. Shorter wavelengths may give a more structured scatter pattern allowing a better estimate of particle parameters.

One of the objectives of the experiments was to demonstrate the use of pin diode detectors in a field that has traditionally used photomultipliers. Solid state detectors tend to become gradually less sensitive to shorter wavelengths and have a cut-off at longer wavelengths where photon energy is less than the material bandgap.

It is often useful to be able to modulate a light source. Modulation of the source is not strictly necessary but switching a source on and off does allow instrument offsets to be monitored in real time so that they do not affect the result.

A consideration which in some applications is more important than the power is whether a coherent or incoherent source is required. A requirement for a coherent source effectively means a laser is needed, although LEDs can show a coherence length of a few microns. In this research there was no real preference for either a coherent or incoherent source.

The choice of light sources used for experiments was to a large extent determined by what was available without large capital expenditure. This basically limited the options to tungsten, mercury or sodium lamps, HeNe laser, diode lasers and LEDs. Of these the lasers and LEDs are the preferred choices for their spectral output, ease of use and in the case of lasers, their power.
A HeNe laser, a 10mW CW laser at 633nm, was the highest intensity source available in the visible region.

Several laser diodes were available, the best compromise between power and wavelength appearing to be a 30mW device at 780nm, the Sharp LT024MDO.

Various LEDs were tried but the best were FFT2000BHRs packaged by Fibre Data which are high power devices centred at 660nm and fitted into bulkhead housings with SMA connectors. The use of LEDs is an interesting study from an engineering point of view since if they can be successfully used there are savings to be made compared to laser based systems.

Sources finally used for experiments were the HeNe laser, LT024MDO laser diode, FFT2000BHR LED and a tungsten white light source.

3.4. Source Drivers

Each of the source types used needs its own type of drive electronics. The HeNe laser has its own commercial mains power supply and since it is a CW laser it cannot be modulated electrically. The tungsten lamp runs off mains power and therefore needs no special power supply.

The current sources for the LEDs and laser diodes were constructed specifically for use in the research work.

The driver initially used for LEDs consisted of a timer circuit switching a transistor. DC power was obtained from a bench top supply. The timing circuit was based on a dual monostable, one monostable triggering the other and visa versa. The lengths of the output pulses from each monostable are controlled by a variable RC circuit external to the chip enabling control of both period and duty cycle. An output from one of the monostables is buffered before being used to drive the base of a transistor. This transistor is used as a switch to pulse the LED. LED current is controlled by a variable resistor which limits the voltage drop across the LED. A 1 ohm resistor in series with the LED is used to monitor the current with an oscilloscope. This driver circuit was used in a number of experiments.
although it has now been superseded by the laser diode driver which can be used to drive LEDs as well as lasers.

The laser diode driver is based on a commercial PCB made by Oxford Optronix. The board performs several functions. Four outputs are provided to drive diode lasers, two of which use pin diode detectors built into the laser package to provide feedback for constant power operation. These constant power drives are intended for CW operation. The other two drives work in constant current mode and may be modulated between two power levels set by potentiometers. The modulated constant current drive works by starting with a constant current source which supplies the laser. A modulation signal then controls the proportion of that constant current that is sunk down a load parallel to the laser, using this method keeps a constant current load on the power supply. Drive currents can be monitored on an LCD included on the Oxford Optronics LDTC4000.

Interlock inputs and modulation inputs are electrically isolated from the laser driver board using 6N137 opto-isolators. This provides overvoltage protection and allows floating earth signals to be used as inputs to the laser driver. Noise immunity is improved because earthing problems between different pieces of equipment are avoided. The opto-isolators have a digital output so that they may only switch the lasers on or off, analogue modulation is not possible.

3.5. Detectors

There are several qualities desired in a detector for light. These are:

1. A high quantum efficiency.

2. High sensitivity / responsivity.

3. Good dynamic range.

4. Low noise.

5. Spectral response matching wavelengths of interest.


7. Ease of use.

8. Low cost.
The choice of silicon pin diode detectors for use with the nephelometers was made for the following reasons. Their spectral response matched the sources that were to be used, they have a large dynamic range, dark current is low and they can be obtained in suitable housings. Their ease of use (no high voltage supply, cooling not essential), robustness and low cost make them ideal for industrial applications if they can be engineered to work satisfactorily in applications that traditionally have used photon counting techniques.

A detector and receiver unit was designed to interface simply with the rest of the system, in particular the data acquisition board that was to be used, a National Instruments NB-MIO-16X. Light detection is by pin diodes, reversed biased to 15 Volts (photoconductive mode) with a load resistor of 1MΩ as shown in figure 3.9. The pin diode acts as a current source and the voltage dropped across the load is amplified to produce a signal suitable for input to the data acquisition board. A higher bias voltage would give a higher sensitivity but would also lead to a larger dark current. A 1MΩ load resistor was chosen as a compromise between sensitivity, thermal noise and linearity. The large value of resistance leads to increased thermal noise but this is offset to some extent by the increased sensitivity from a large voltage drop across the resistor. The large resistor value also guarantees a linear voltage response with intensity. Each channel has a variable gain for the preamplifier, set individually using a selector switch within the unit. Selectable gains are 1, 10, 100, 200, 300, 500, 600, 700 and 800. Dynamic range on the output is from -12.5 Volts to +12.5 Volts.

![Figure 3.9. Pin diode receiver circuit.](image-url)
The pin diodes are FDR850 IRLCs supplied by Fibre Data in metal SMA housings. These have a maximum responsivity at 850nm.

3.6. Synchronisation

Two different data acquisition systems have been used in this research. One is the National Instruments NB-MIO-16X board installed in a Macintosh computer used with the first designed nephelometer and the other is a National Instruments Lab-PC+ installed in an IBM compatible and used with the second nephelometer. In some experiments the source was modulated on and off to enable measurement of receiver offsets and any stray background light. In these experiments it was necessary to synchronise the sampling of the detected signal with the modulation of the source.

The Macintosh system was used with LabView which did not use any special synchronisation hardware. Software was written with LabView to control the data acquisition board and work out the average values of the scattered light intensities. The data acquisition board takes a block of samples from all six channels at a rate of 1kHz per channel. The software then scans the data from line of sight channel (the channel with the best signal to noise ratio) to find a positive edge in the received signal. This edge corresponds to the point at which the LED was switched on. An average of two hundred and fifty samples taken before the edge gives a measurement of the background and amplifier offsets and an average of two hundred and fifty samples taken after the edge gives a measurement of the scattered light. The LED was modulated at a 50% duty cycle with a period of 600ms for these experiments and enough samples were taken in each block to ensure that an edge could be found that had 250 samples before it in the same block.

The PC system software was a general data acquisition program written in C++ which does need some purpose built hardware to synchronise the data acquisition and light modulation. For the on-line experiments the Lab-PC+ board is triggered with an external signal which is synchronised with the light modulation so that measurements of background and electronic offsets can be made. This is done by arranging for each channel to be sampled firstly when the source is off and then when the source is on. All four channels are sampled as close together in time as possible and then there is a pause until the source has changed state. Limited bandwidth of the receivers means that there is a finite rise and fall time of the detected signal when the light source is switched. To allow time for the detected signal to rise or fall to its full value there is a delay between the switching of the source and the sampling of the channels.
3.7. Experimental Systems

The components introduced above were used to build the experimental systems for the two nephelometers. The first nephelometer was used in the flow system with an Apple Macintosh computer and National Instruments NB-MIO-16X data acquisition card. The pin diode receiver unit was used to detect and amplify the scattered light. Most of the experiments were made with the dual monostable LED driver and 660nm LED. Power came from bench top supplies. Some experiments were made using the HeNe laser by focusing the laser onto the end of a fibre and trailing the fibre across the laboratory to the flow rig. The fibre had to be run from the optical table to the flow system so that the laser, lens and fibre could be fixed stably and because the laser cannot safely be used in the open. The length of the fibre meant that much of the power was absorbed but this was still the most intense source available, coupling approximately 2mW to the nephelometer. A schematic of the system set-up is shown in figure 3.10.

The second nephelometer design was used with a PC and a National Instruments Lab-PC+ data acquisition board. The source used was the 780nm sharp diode laser housed in an Access Pacific laser collimator SK9620 with modulation timing from the synchronisation circuit. The receiver optics were set-up by launching light through them in reverse and adjusting them until the crispest image of a disc of light was formed on a screen placed at the centre of the nephelometer. This was in fact close to a parallel beam from the optics, a half angle measured as 2°. The complete system set-up is shown in figure 3.11.

The software for the second nephelometer system was written using Visual C++ as a windows application. Several parameters can be set in software such as the number of channels, the gain on each channel (that is the gain of the Lab-PC+ board, not the receiver preamplifiers), number of samples to collect and on board or external triggering can be specified. This software calculates the higher moments of a sample set as well as the mean and stores these statistical measures to disk. Processing and disk speed limit the rate and in what size blocks data can be acquired. As a result only the statistics of the data are stored, not the data samples themselves.

All experiments, using either nephelometer, were conducted in the same manner. Experience showed that the most repeatable results were obtained when the flow speed was kept steady and at a rate such that cavitations did not occur. Stopping and starting the pump gives rise to outgassing or cavitations.

---

Software written by D M Barnett
that can take some minutes to clear. Various different pollutants were tested, most of them oils because of a particular interest in oil pollution monitoring and because they form suspensions easily. Oils were injected into the flow system using Hamilton microlitre syringes. These syringes are quite delicate and the instructions supplied with them should be observed. The viscosity of the oils used makes it hard to fill the syringes and some practice helps. Fine solids can also be injected into the flow system using larger plastic syringes. The method used was to place the required amount of powder in a plastic syringe, insert the syringe through the Subaseal and extract an amount of water from the flow system. The syringe can then be removed and shaken to form a suspension which is then injected back into the flow system. To allow samples to disperse the system was left for a period of a few minutes, typically ten after injection, before taking measurements. With suspensions of oils the droplet size distribution was not controlled directly but left to reach a natural emulsion for the flow conditions. By using the
same flow speed for each experiment and allowing similar dispersion times between adding the oil and making measurements it was hoped that repeatable experiments would be possible. It was also hoped that this would allow the different physical natures of each oil to be reflected in the different suspensions formed. Size distributions of the oils at the end of some experiments were measured using a Coulter counter.

Figure 3.11. MkII nephelometer experimental system.
4. Discussion of Nephelometric Results

Some results from the experiments made using the on-line nephelometers are presented in this chapter with some discussion about the subsequent neural network processing of the obtained data. A more detailed and extensive discussion of the neural network aspect of the work can be found in the PhD thesis of R Naimimohasses [4.1]. Results and findings made using the first designed nephelometer are presented first. A table of the experimental conditions is given in Appendix B.

4.1. Mk I Nephelometer

4.1.1. Comparison of Light Sources

A comparison of different light sources was one of the first sequences of experiments to be made. Sources used were the HeNe laser, tungsten lamp and FFT2000BHR LED. All the experiments were made using soluble oil in tap water. Soluble oil is a ready made mixture of oil and a dispersant used in machining for the cooling and lubrication of cutting tools.

4.1.1.1. HeNe Laser

The HeNe laser was launched through a long fibre with a power at the fibre end measured as 2.8dBm using an Avo Meggar power meter. This meter is not calibrated for the laser wavelength and so the 2.8dBm reflects a real power of about 2.5mW, the meter being approximately 75% efficient at 633nm. The gains on each of the receiver channels were set as follows to make best use of the available dynamic range: 45°=500, 90°=800, 135°=600, 180°=1, 240°=700 and 300°=800.

Raw data in the form of voltages measured by the data acquisition board are shown in figure 4.1. Five points are plotted on top of each other at each concentration which is described in terms of μl of oil per litre of water. The 90° and 300° data are hard to distinguish due to their similarity of value. Over the range tested the intensity of light detected at each angle increases as the concentration of oil increases.
except for the line of sight detector which shows the opposite trend. This reflects increased angular scattering as more oil droplets appear in the light beam. This leaves less power in the beam itself and so the intensity measured by the 180° sensor decreases. Increased absorption also adds to the decrease in the 180° sensor reading. If the experiment had been continued to higher concentrations of oil, absorption and multiple scattering would start to affect all the sensors and the measured intensities would start to drop again as found in [4.2].

Figure 4.1 shows that each of the sensors indicates a different initial intensity with clean water. This is due in part to different amounts of light scattered by the water molecules, partly due to the beam profile and partly due to amplifier offsets. The line of sight sensor obviously has a high initial intensity because it detects the direct transmitted intensity. The 45° sensor has a high starting intensity because of an apparent reflection of the input beam off the opposite wall of the pipeline.

It is easier to make direct comparisons between the different angles using figure 4.2 which is a plot of the difference between the measurements made at each oil concentration and the mean measurement at 0 µl/l, that is

\[
\Delta S_\theta(c) = S_\theta(c) - \langle S_\theta(0) \rangle
\]  

(4.1)

where \( S_\theta(c) \) is the measured signal at angle \( \theta \) and oil concentration \( c \). This clearly shows that the greatest increase in scattered light is in the forward direction, 135°, followed by 240° and then 90° and 300°. The 45° direction apparently has the smallest increase in scattered intensity but it has to be remembered that the gains on each receiver channel are different. Dividing each of the data sets in figure 4.2 by the appropriate channel gain leads to the plot of figure 4.3 which more correctly indicates the change in intensities during the experiment. This figure indicates that the biggest changes in scattered light intensity are in the forward direction but that the scattering is more isotropic at large angles.

Another method of presenting the data which has proved useful for neural network processing is to normalise the data to the transmission measurement and the initial values. The first step in this normalisation process is to divide the angular measurements by the line of sight measurement at the same concentration:

\[
N_\theta(c) = \frac{S_\theta(c)}{S_{180}(c)}
\]  

(4.2)
Figure 4.1. Raw data from experiment using HeNe laser and soluble oil.

Figure 4.2. ΔS for experiment using HeNe laser and soluble oil.
Figure 4.3. Gain corrected $\Delta S$ for experiment using HeNe laser and soluble oil.

Figure 4.4. dB normalised data from experiment using HeNe laser and soluble oil.
The aim of taking this ratio is to compensate for the absorption taking place as the concentration of pollutant increases and to account for any variations in the power of the light source. The second stage of the normalisation process uses the unpolluted sample as a reference in order to reduce the effects of any changes in the set-up or water from day to day. This second step can be described by the formula,

\[ I_\theta(c) = 20\log_{10}\left(\frac{N_\theta(c)}{N_\theta(0)}\right) \] (4.3)

This final measure, \(I_\theta(c)\), is a decibel scaled measure of a relative change in signal at each angle. Figure 4.4 shows the result of this normalisation with the data from the HeNe laser and soluble oil experiment. It can be seen that most of the curves are closer together indicating that the increase in scattered power in each direction with increasing concentration of oil is in proportion to the relative amounts scattered at the start of the experiment, i.e. a single scattering regime exists over the range of concentrations investigated. The 45° sensor is an exception to this, probably due to the high initial measure caused by a reflection.

4.1.1.2. Tungsten White Light Source

Two experiments were made using the tungsten white light source, one at the minimum power setting and one at the maximum power setting. Soluble oil was again used as the pollutant.

Figures 4.5 to 4.8 show the data obtained using the low power setting in a similar format to the HeNe experimental data. Gains on the receivers were set to 100 for the 45° sensor, 800 for 90°, 100 for 135°, 1 for line of sight, 200 for 240° and 500 for the 300° sensor channel. These gains which are different from the HeNe experiment explain why the raw data have quite different relative sizes even though the pollutant is the same. The changes in signal size corrected for the different gains, figure 4.7, lead to a set of data points similar to those obtained using the HeNe laser. One point worth noting from figure 4.7 is that the changes in signal are approximately three times the size of those obtained using the HeNe laser indicating that more power is being scattered from the test volume. From the raw data of the 180° sensor in figure 4.5 it appears that the light power launched into the cell is about double that of the HeNe experiment. This apparently higher scattering efficiency, three times the scattering power with twice as much input power, could be due to the tungsten light source containing shorter wavelengths which will be scattered more effectively by the oil droplets or to effects of water temperature as discussed below. The normalised data given in figure 4.8 shows a similar trend in data to that obtained using the HeNe laser source.
Figure 4.5. Raw data from experiment using low power white light and soluble oil.

Figure 4.6. $\Delta S$ for experiment using low power white light and soluble oil.
Figure 4.7. Gain corrected $\Delta S$ for experiment using low power white light and soluble oil.

Figure 4.8. dB normalised data from experiment using low power white light and soluble oil.
An important discovery from this experiment was the effect of temperature on the measurements made. All of the figures 4.5 to 4.8 indicate an initial rise in the transmitted light power and an initial decrease in the scattered power at the 45° and 135° angles. This is the opposite of what is expected and could be due to either variations in the source power or changes in the transmission characteristics of the water. The beam of light becomes more focused on the line of sight detector indicating that the effect is not simply a change of source power since this would effect all channels similarly. Electronic effects are unlikely since the use of optical fibres makes all the electronics remote. This leads to the supposition that as the experiment takes place the water temperature increases and the absorption characteristics and refractive index of the water change. The effect is only apparent at the start of the experiment where the temperature variation is most rapid and where the increase in scattering with time is slowest because of additions of oil being made in small steps. Experiments to check this hypothesis and quantify the effect of temperature were performed and are described later. Those experiments suggest that the temperature rise over the period of the tests using the white light source was approximately 10°C.

The second experiment using the white light source produced similar results to the first but as expected the greater source power led to larger changes in the measured signals as indicated by figures 4.9 and 4.10. An increase in the transmitted power is again observed as the water temperature increases but the effect is not as great as in the previous experiment. The normalisation process in this experiment has a marked effect on the 300° channel. In this case a low initial measure on that sensor leads to the normalised values being larger than in the previous experiments. The reason for the initial

![Figure 4.9](image-url)  
**Figure 4.9.** Raw data from experiment using high power white light and soluble oil.
Figure 4.10. Gain corrected $\Delta S$ for experiment using high power white light and soluble oil.

Figure 4.11. dB normalised data from experiment using high power white light and soluble oil.
low reading is not known but it has been determined that the initial temperature will affect the measure as would any damage to the optical fibres. Although the normalisation process is not as successful at compensating for differing initial conditions as one would like, it has proved the best technique for later neural network processing.

4.1.1.3. LED Source

Figures 4.12 to 4.14 show results from an experiment with a LED as the source and soluble oil as the pollutant. The power from the LED driven at 50mA is less than that from the tungsten and HeNe laser sources and so the receiver gains are set higher, all were set to 800 except the line of sight receiver which was kept at a gain of 1. The data of figures 4.12 to 4.14 have had receiver offsets removed by the process of modulating the LED and measuring the amplified dark current.

The results clearly show a lower power signal due to the lower source power. The change in the signals with pollutant level show similar trends to the other types of source and some variation because of a temperature effect is also evident on the 180° sensor. The normalised data also follow similar trends to the lower power white light source experiment and the HeNe laser experiment. There is little change in the normalised values compared to earlier experiments because of the lower source power. The normalisation procedure therefore works well in that it compensates for varying input powers.

![Figure 4.12. Raw data from experiment using LED and soluble oil.](image)
Figure 4.13. Gain corrected $\Delta S$ for experiment using LED and soluble oil.

Figure 4.14. dB normalised data from experiment using LED and soluble oil.
4.1.1.4. Summary of Comparing Light Sources

A conclusion which can be drawn from this sequence of experiments is that the more light power is launched into the nephelometer, the more is scattered and detected as expected. Bearing this in mind, a higher power source should give a better signal to noise ratio and therefore a lower detection limit. Data for oil concentrations under 1μl/l were not obtained in the experiments using the tungsten lamp and so a minimum limit of detection for this system cannot be stated. However, the gas laser experiment did obtain data from lower concentrations. Figure 4.15 is as figure 4.4 but showing only the lower concentration range. From this it can be seen that under 0.5μl/l the spread in the measurements at each concentration becomes significant compared to the signal amplitude. The underlying trend continues and so further averaging of the detected signal could reduce the spread in points but as it stands the limit for making a reasonable estimate of concentration is around 0.3 to 0.4μl/l using the HeNe laser. This would be expected to improve with better coupling or using the tungsten lamp.

![Graph showing relative change in signal (dB) vs ppm by volume](image)

**Figure 4.15.** dB normalised data from experiment using HeNe laser and soluble oil.
It also appears that the soluble oil scatters the white light more efficiently than the red light of the HeNe laser and LED. Theory would suggest that it is the shorter wavelengths present in the white light are scattered more efficiently by the oil droplets.

An ideal light source for these experiments would therefore be one with a high power and of shorter wavelength than 633nm.

Another observation is that the water temperature has an effect on the measurements. Repeatability is obviously affected by this and an attempt to quantify the significance of the temperature effect was made by performing two different experiments as discussed in the next section.

### 4.1.2. Effect of Water Temperature

Experiments to compare the effectiveness of different light sources had led to the discovery that the measured light intensities apparently varied with the temperature of the water. Two experiments were made to check that the cause was the water temperature changing and quantify what effect it would have on measurements. In one experiment the transmission of light through water was measured as temperature varied to determine if the temperature coefficient of absorption was positive or negative and the other was an experiment using the flow system in which no pollutant was added to quantify the effect on the nephelometer and check repeatability.

#### 4.1.2.1. Variation of Absorption with Temperature

This experiment used the apparatus of figure 4.16. The HeNe laser was shone through a cuvette 60mm long and onto a receiving fibre. This fibre was connected to the receiver unit, the output of which was monitored on a digital multimeter. A beam splitter was placed before the cuvette to allow a second fibre and receiver to monitor the input power. This channel was also monitored using a digital multimeter. The filter was to reduce the laser power to a level that did not saturate the receivers. Water from a boiled kettle was poured into the cuvette and the digital multimeter readings were noted at various times as the water cooled. Temperature was not measured directly since a numerical value for the temperature coefficient of absorption was not required, it was the underlying trend that was being investigated. The obtained data are given in table 4.1. It is clear without plotting a graph that as time progresses and the water cools that the light power transmitted through the cuvette decreases whilst the
source power remains almost constant. It is therefore concluded that water absorbance at 633nm has a negative temperature coefficient. This was just a quick experiment to investigate how absorption varies with temperature and no special attempt was made to make the most accurate measurements possible. For example, the boiled water was not filtered to remove particulates but the slow response of the digital multimeters should average out dynamic effects such as scattering from contamination.

<table>
<thead>
<tr>
<th>time (minutes)</th>
<th>transmitted power measure (Volts)</th>
<th>laser power measure (Volts)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.88</td>
<td>5.78</td>
</tr>
<tr>
<td>1</td>
<td>0.84</td>
<td>5.78</td>
</tr>
<tr>
<td>3</td>
<td>0.83</td>
<td>5.78</td>
</tr>
<tr>
<td>5</td>
<td>0.80</td>
<td>5.79</td>
</tr>
<tr>
<td>8</td>
<td>0.73</td>
<td>5.79</td>
</tr>
<tr>
<td>10</td>
<td>0.72</td>
<td>5.79</td>
</tr>
<tr>
<td>15</td>
<td>0.69</td>
<td>5.79</td>
</tr>
<tr>
<td>20</td>
<td>0.66</td>
<td>5.80</td>
</tr>
<tr>
<td>25</td>
<td>0.64</td>
<td>5.79</td>
</tr>
<tr>
<td>30</td>
<td>0.60</td>
<td>5.79</td>
</tr>
<tr>
<td>36</td>
<td>0.59</td>
<td>5.80</td>
</tr>
<tr>
<td>40</td>
<td>0.58</td>
<td>5.80</td>
</tr>
<tr>
<td>45</td>
<td>0.57</td>
<td>5.79</td>
</tr>
<tr>
<td>50</td>
<td>0.57</td>
<td>5.82</td>
</tr>
<tr>
<td>55</td>
<td>0.56</td>
<td>5.82</td>
</tr>
<tr>
<td>60</td>
<td>0.56</td>
<td>5.82</td>
</tr>
</tbody>
</table>

Table 4.1. Variation of attenuation as temperature changes with time.
4.1.2.2. Effect of Temperature on Mk I Nephelometer

In the second experiment water was simply pumped around the flow system and measurements of scattered light were made with the nephelometer. As the experiment progressed the water heated up because of heat transfer from the pump to the closed water circuit. Water temperature was measured using a thermocouple inserted through the pipeline wall. This experiment was repeated and results from both experiments are shown in figures 4.17 to 4.22. These experiments were made using the HeNe laser, the highest power monochromatic light source available. Using a monochromatic source prevents any wavelength dependent effects from confusing the result. Gains on the receivers were 200 for the 45° channel, 1 for the 180° channel with the others set to 800. Good repeatability is shown between the two sets of results with only a small difference in the initial measurements made at the start of the experiment. The most obvious feature is the increase in transmitted intensity as the temperature increases. This increase indicates that the laser beam is either being attenuated less or becoming more focused on to the receiving fibre. The signal detected at the 45° detector, which is believed to consist in part of a reflection, decreases as the temperature increases. If the only effect was a lower absorption as
Figure 4.17. Raw data from temperature variation experiment using HeNe laser.

Figure 4.18. Raw data from repeat temperature variation experiment using HeNe laser.
Figure 4.19. Gain corrected $\Delta S$ for temperature variation experiment using HeNe laser.

Figure 4.20. Gain corrected $\Delta S$ for repeat temperature variation experiment using HeNe laser.
Figure 4.21. dB normalised data from temperature variation experiment using HeNe laser.

Figure 4.22. dB normalised data from repeat temperature variation experiment using HeNe laser.
temperature rose then this signal would increase as the line of sight measurement does. It can therefore be concluded that a change in focus of the laser is responsible for at least part of the variation seen with temperature. The 240° sensor shows an interesting effect in that the intensity at that angle starts by increasing with the temperature and then drops again. The initial increase could be due to better transmission or by more light reaching that detector direct from the launch fibre. The following decrease must be due to either less scattering or to less stray light reaching the detector from the launch. There is no reason to suspect that the scattering should change significantly and so it seems probable that stray light is the cause of this peculiar effect. Because of the wide angle from the line of sight and because the same effect is not seen at 135°, it is suspected that a sidelobe of the main beam is close to this detector which moves away as the temperature of the water increases and the focusing of the light beam changes. (Small lenses do not focus all the incident light onto the focal point, some is focused to points off axis. The effect is similar to that of a large particle scattering light). The remaining sensors at 90°, 135° and 300° all display an increase in intensity as the temperature rises. This seems to indicate a general increase in the amount of light being detected all round the sensor as though there is less absorption of the scattered light at higher temperatures. A combination of changing absorption and beam profile appears to be responsible for the detected variations with temperature. The precise mechanisms of how the light scattering and absorption change with temperature remains debatable since the literature (e.g. [4.3]) reports only a slight variation of refractive index with temperature. The results obtained in this work indicate a more significant effect. However, the important result is that these temperature effects are repeatable and so some type of correction could be made to the data if the temperature was monitored. In later experiments the temperature was monitored using the thermocouple but no processing of the data was attempted to correct for temperature changes.

4.1.3. Scattering from Different Oil Types

Another sequence of experiments investigated scattering from different types of oil in suspension. These experiments were made using a LED source with offset subtraction. Although the LED is a relatively low power source, the repeatability is good because of the stable power output and automatic offset subtraction possible with electronic modulation. In all of the following experiments the LED drive current was 50mA and all receiver gains were set to 800 except the line of sight receiver which was set to unity.

The water was circulated for a period before measurements were started. This was to try and reduce the temperature changes during the experiment by waiting until the greatest temperature gradient was over.
The oils used were two types of crude, one from a laboratory chemical supplier and one a North Sea crude supplied by the Elf oil company. Hereafter these are referred to as crude 1 and crude 2 respectively. Also used were two refined oils, an oil used in motorcycle forks and an engine oil (Castrol GTX). Soluble oil was also used, the results of one experiment already presented in section 4.1.1.3.

4.1.3.1. Crude 1 Experiments

The first experiment using crude 1 gave rise to the results of figures 4.23 to 4.25. The water temperature when measurements were started was 29.4°C and at the end of the experiment was 32.9°C. Comparing these results to those using soluble oil (figures 4.12 to 4.14 above) it can be seen that there is only about one twentieth the variation in detected signal with crude 1. It therefore follows that there is less scattering of the light. This is reflected in the normalised data being noisier because of a lower signal to noise ratio. There is also evidence of temperature affecting the result in that the 45° sensor drops in voltage which has been seen to be typical of an increasing water temperature. The variations on the other channels are not in the same proportion as in figures 4.18 and 4.22 (variation with temperature data) and so it is believed that not all of the variation is due to temperature changes. The low level of scattered light suggests that the crude oil droplets are either poor scatterers or that there are only a few in suspension. When injecting the oil it was noticed that large drops formed at the end of the syringe needle which were eventually pulled off by the flow. The crude oil not mixing easily with water causes two problems. Firstly larger droplets are formed and so there is less scattering to detect, and secondly, should some oil come into contact with the pipeline wall and adhere to it, there is a relatively large proportion of the oil not actually in suspension. This leads to inaccurate estimates of the oil concentration.

In an attempt to mix the crude oil better and to get more scattering a repeat experiment was made but the flow control valve was opened at the time of injecting the oil to speed up the flow. With this higher flow rate oil drops were seen to break off the tip of the syringe needle in smaller blobs, approximately 0.25mm to 0.5mm diameter. After the oil had been injected the flow was reduced back to its original speed and left to settle for a few minutes. Measurements at different oil concentrations were made at ten minute intervals. The results from this experiment are shown in figures 4.26 to 4.28. The starting temperature was 29.0°C and the water temperature at the end was 32.3°C. The initial voltages recorded are similar to those of the previous experiment but as oil is added to the system the amount of scattering detected is significantly larger, approximately three times as great. It is possible that this increased scattering is due to a greater number of oil droplets formed by better mixing or it could be due to the increased speed causing turbulence and outgassing which does not settle out before measurements are made.
Figure 4.23. Raw data from experiment using LED and crude1.

Figure 4.24. Gain corrected ∆S for experiment using LED and crude1.
Figure 4.25. dB normalised data from experiment using LED and crude.

Figure 4.26. Raw data from experiment using LED and crude, flow rate increased for injection of oil.
Figure 4.27. Gain corrected $\Delta S$ for experiment using LED and crude1, flow rate increased for injection of oil.

Figure 4.28. dB normalised data from experiment using LED and crude1, flow rate increased for injection of oil.
4.1.3.2. Soluble Oil / Effect of Flow Speed

To obtain an idea of how the changing flow rate affects the scattering a second soluble oil experiment was performed. The size of soluble oil droplets should be affected little by the higher flow rate because it has a dispersant to break the oil up anyway. Results from this second soluble oil experiment are given in figures 4.29 to 4.31. Less variation in signal level is detected in this second soluble oil experiment (c.f. figures 4.12 to 4.14) but this could be due to either less scattering, higher initial background scatter masking the oil scattering or different experimental conditions. The initial measures are almost the same in each of the two soluble oil experiments so there is no obvious difference in the initial scattering and conditions. The temperature range in the second experiment is greater than in the first and this is likely to be the cause of the smaller variations in the measured scattering. In any case it can be concluded that the faster flow speed has not led to a measurable increase in scattering.

A conclusion drawn from this result is that using a faster flow speed as the oil is injected does not produce scattering from water turbulence that is measurable after a settling period. This in turn implies that the increased scattering detected in the second crude I experiment as discussed in 4.1.3.1. was due to better dispersion of the oil.

Figure 4.29. Raw data from experiment using LED and soluble oil, flow rate increased for injection of oil.
Figure 4.30. Gain corrected $\Delta S$ for experiment using LED and soluble oil, flow rate increased for injection of oil.

Figure 4.31. dB normalised data from experiment using LED and soluble oil, flow rate increased for injection of oil.
4.1.3.3. Crude 2 Experiments

In light of the above conclusion, experiments with crude 2 used a higher flow speed as the oil was injected into the flow system.

Results from three experiments using crude 2 are plotted in figures 4.32 to 4.40. These show a similar trend in each experiment with most scattering in the forward (135°) direction. The scattering in the 90° and 240° directions appears to be roughly equal but the 90° channel is noisier as best seen in the dB normalised data. To make the data trends easier to see figures 4.41 to 4.43 show quadratic curve fits to the data. An obvious feature in the third experiment is a drop in all of the measurements in the 40 to 50 μl/l concentration range. This decrease in measured light power is not just due to momentary noise because the measurements are averages and because the effect is shown in several consecutive measurements. The likeliest cause is fouling of the launch fibre reducing the amount of light power reaching the test volume. This fouling clears itself during the period that the measurements at 60 μl/l were taken and the measured intensities then return to the trend that is typical of the earlier two experiments.

![Figure 4.32. Raw data from experiment using LED and crude2, flow rate increased for injection of oil.](image-url)

91
Figure 4.33. Raw data from 1st repeat experiment using LED and crude2, flow rate increased for injection of oil.

Figure 4.34. Raw data from 2nd repeat experiment using LED and crude2, flow rate increased for injection of oil.
Figure 4.35. Gain corrected ΔS for experiment using LED and crude2, flow rate increased for injection of oil.

Figure 4.36. Gain corrected ΔS for 1st repeat experiment using LED and crude2, flow rate increased for injection of oil.
Figure 4.37. Gain corrected $\Delta S$ for 2nd repeat experiment using LED and crude2, flow rate increased for injection of oil.

Figure 4.38. dB normalised data from experiment using LED and crude2, flow rate increased for injection of oil.
Figure 4.39. dB normalised data from 1st repeat experiment using LED and crude2, flow rate increased for injection of oil.

Figure 4.40. dB normalised data from 2nd repeat experiment using LED and crude2, flow rate increased for injection of oil.
Figure 4.41. Quadratic fit to dB normalised data from experiment using LED and crude2, flow rate increased for injection of oil.

Figure 4.42. Quadratic fit to dB normalised data from 1st repeat experiment using LED and crude2, flow rate increased for injection of oil.
4.1.3.4. Motorcycle Fork Oil Experiments

Two experiments using motorcycle fork oil (figures 4.44 to 4.49) lead to results similar to those for the crude oils. The 90° channel is again the noisiest and most scattering is once again in the forward direction. The size of the measured signals are also similar to the crude oils implying that there was a similar amount of power scattered by the suspension. In these experiments the flow speed was increased at the time of injection as in the crude oil experiments.

4.1.3.5. Castrol GTX Experiments

Castrol GTX gave rise to less intense scattering as shown in figures 4.50 to 4.52. Again the 90° channel is noisy compared to the other channels and the relative intensities similar to earlier experiments.

Figure 4.43. Quadratic fit to dB normalised data from 2nd repeat experiment using LED and crude2, flow rate increased for injection of oil.
Figure 4.44. Raw data from experiment using LED and motorcycle oil, flow rate increased for injection of oil.

Figure 4.45. Raw data from repeat experiment using LED and motorcycle oil, flow rate increased for injection of oil.
Figure 4.46. Gain corrected $\Delta S$ for experiment using LED and motorcycle oil, flow rate increased for injection of oil.

Figure 4.47. Gain corrected $\Delta S$ for repeat experiment using LED and motorcycle oil, flow rate increased for injection of oil.
Figure 4.48. dB normalised data from experiment using LED and motorcycle oil, flow rate increased for injection of oil.

Figure 4.49. dB normalised data from repeat experiment using LED and motorcycle oil, flow rate increased for injection of oil.
Figure 4.50. Raw data from experiment using LED and Castrol GTX, flow rate increased for injection of oil.

Figure 4.51. Gain corrected $\Delta S$ for experiment using LED and Castrol GTX, flow rate increased for injection of oil.
4.1.3.6. Properties of Oil Suspensions

The sequence of experiments discussed in section 4.1.3. indicates that the soluble oil has quite different scattering properties from the other oils tested. The most obvious difference is in the amount of light scattered. More light is scattered by the soluble oil which theory suggests is due to the smaller droplet size obtained using soluble oil. Figure 4.53 shows a soluble oil droplet size distribution as obtained using a Coulter counter. For comparison the size distribution obtained from a sample of crude 2 taken from the flow system at the end of an experiment (figure 4.54) shows a wider distribution centred around a larger mean size.

Smaller droplets are to be expected from the soluble oil because of the dispersant included to break up the oil. The more isotropic scattering obtained with soluble oil is also evidence of a smaller droplet size.
Figure 4.53. Size distribution of soluble oil droplets measured using Coulter counter.

Figure 4.54. Size distribution of crude2 oil droplets measured using Coulter counter.
The essentially similar nature of scattering obtained using all the other oils suggests that their properties are similar. Measurements of the refractive index of each oil were made using an Abbe '60' refractometer to compare their optical properties. Results are given in table 4.2 which show that the refractive indices of all the oils were in the region of 1.48. Soluble oil is mid range of the oils used with Castrol GTX having the highest index and crude 2 the lowest. Since Castrol GTX and crude 2 showed similar scattering and the soluble oil is mid range, particle size must have a greater influence on the scattering than optical properties for the materials tested. Another observation made was that when left unsealed overnight the properties of a sample of crude 2 changed. The different refractive index is noted in the table but the oil was also visibly more tar like after being left. This is an important factor to note if experiments using oils are to be repeated some time after the sample was originally opened.

<table>
<thead>
<tr>
<th>oil type</th>
<th>refractive index</th>
</tr>
</thead>
<tbody>
<tr>
<td>soluble</td>
<td>1.48215</td>
</tr>
<tr>
<td>motorcycle fork</td>
<td>1.47769</td>
</tr>
<tr>
<td>Castrol GTX</td>
<td>1.48551</td>
</tr>
<tr>
<td>crude 1</td>
<td>1.48304</td>
</tr>
<tr>
<td>crude 2 (fresh)</td>
<td>1.47785</td>
</tr>
<tr>
<td>crude 2 (aged)</td>
<td>1.47105</td>
</tr>
</tbody>
</table>

Table 4.2. Refractive indices of different oils.

Since particle size appears to be more important in determining the scattering than refractive index it seems reasonable to conclude that the similarity between experimental results is due to roughly similar oil droplet size distributions. This is not necessarily unexpected since the oils were mixed in similar flows in each experiment. The slight differences between experiments would therefore be because of slightly different size distributions, due to differing hydrophobic nature of the oils, and the less important small differences in refractive index.
The main object of the experiments was not to visually compare scattering from different materials but to obtain data in order to see if neural processing could automatically identify different pollutants and their concentration. To this end the data obtained from the experiments were separated into training and test data. Training data were used in the process of training neural networks and test data were kept separate and only used after training was complete to test the performance of the neural networks. In testing neural network performance it is important to have data obtained from more than one experiment to make sure that the neural network is responding to the intended variable and not identifying other experimental conditions such as temperature or cleanliness of the sensor etc.

The problems of identifying oil type and estimating concentration were initially investigated by the author in references [4.4] and [4.5]. The type of neural networks used in this work were multilayer perceptrons (MLP) using the common logistic activation function and trained using backpropagation [4.6]. Reference [4.4] concentrates on the problem of estimating volume fraction using nephelometry, showing how a reasonable estimate of concentration could be made and demonstrating some immunity to sensor fouling. Reference [4.5] details how a second neural network with five input neurons and twenty hidden neurons was trained to identify different oils with data from the mkI nephelometer. Data normalised as in (4.2) and (4.3) were input to the neural network and the output was trained such that each oil type would activate a separate output neuron, providing a type of classifier where the largest output indicated the most likely oil type. This method was found to work reliably for the motorcycle fork oil, the soluble oil and crude I, but was not capable of distinguishing the other two oils tested.

Figure 4.55 shows the output value at each of the three output neurons across a range of concentrations for the network trained to classify soluble, crude I and motorcycle fork oil. The way this network was trained meant that even at zero concentration the network had to classify the suspension to be one of the three oils which explains the apparent recognition of oil type before any oil is added.

A more complete study of concentration estimation and classification was made in [4.7]. Here the oil classification network was trained not to give an output at zero concentration and the output of the classifier was thresholded and then fed as an extra input into the concentration estimator network. Using the oil type classification as an extra input led to a better performance in estimating the concentration. This was expected since each oil type gives slightly different scattering intensities for the same concentration; knowing which oil type is present therefore gives a guide to the relationship between concentration and scattered intensity which is most appropriate. The overall neural network used was thus a partially concatenated MLP with the classifier part using five normalised inputs, seven hidden layer neurons, three outputs which feed into a comparator device (with an output value of 1, 2 or
Figure 4.55. Artificial neural network oil type classifier performance.

Output at each of the three network outputs for oil concentrations 0 to 100 µl/l of soluble oil, fork oil and crude 1, indicating correct classification.

3 depending on which of the classifier outputs is the larger) which then acts as an input to a concentration estimator MLP with six inputs (the same five as the classifier plus the classifier output), twenty hidden layer neurons and a single output neuron giving an output value proportional to the concentration. The number of neurons used in the hidden layers was obtained using the optimal brain damage technique [4.8]. Using more neurons did not lead to a network that was successful in classifying more oil types or significantly better at estimating concentration. Figure 4.56 shows the network performance in classifying the three oil types (soluble, crude 1 and motorcycle fork oil) and figure 4.57 shows the concentration estimator performance. Correct classification of oil type over the range 20 to 100 µl/l was achieved for all three oils and over the full range (0 to 100 µl/l) for soluble and motorcycle fork oil. At low concentrations identification of crude oil is unreliable but not in the sense that other oils are wrongly identified instead. Error in the concentration estimation is dominated by errors at low concentrations where the signal to noise ratio is poorest.
Figure 4.56. Second ANN oil type classifier performance, trained to give no output at zero concentration.

Output at each of the three network outputs for oil concentrations 0 to 100 µl/l of soluble oil, fork oil and crude 1 indicating correct classification at concentrations greater than 20µl/l.

Figure 4.57. Concatenated ANN concentration estimation performance.

Output of network for oil concentrations 0 to 100 µl/l of soluble oil, fork oil and crude 1.
Increasing the complexity of the neural network did not aid in identifying more oil types. It follows therefore that the scattering from the different oils is too similar or too noisy for the neural network to distinguish between more than three oils. This is certainly a limit with this system in the application to identifying different oils but since it is believed that the scattering is similar because the size distributions of oils are similar it is quite possible that this system could find application as a cheap particle sizer. Further experimentation using graded suspensions of solids would be required to test this hypothesis.

4.1.5. Summary of Mk I Nephelometer Study

Soluble oil forms a finer suspension than the other oil samples when dispersed in the flow system. The soluble oil forms a suspension with the majority of droplets less than $1\mu$m in size and nearly all less than $10\mu$m. The other oil samples are harder to emulsify and form suspensions of a broader size range but mostly about $10\mu$m diameter. The refractive indices of the oils are similar.

The different size distribution of the soluble oil means that it scatters more light and in a different angular pattern than the other samples. Because the scattering is clearly different a neural network may be trained to distinguish soluble oil from the other types. Because the size distributions of the other oil suspensions tested are similar, the scattering is also similar. Accordingly it is harder to train a neural network to classify the crudes, motor oil and motorcycle oil although a network was successfully trained to distinguish the motorcycle fork oil and crude 1.

A neural network may be trained to estimate the volume fraction of oil added to the experimental system. The estimate tends to be less accurate at low concentrations, as expected, because of the lower signal to noise ratio.

The temperature of the water appears to affect the measurements. Once temperature effects were noted the temperature was monitored during experiments. Calibration of the sensor would allow measurements to be corrected for changing temperatures but this was not attempted with the experimental data.
4.2. Mk II Nephelometer

To try and develop a system with greater sensitivity to suspension type the second nephelometer was designed and built as described in chapter 3. The lower numerical aperture of the receivers should be more selective in the angular scattering they detect which in turn should produce a system more sensitive to small changes in the angular distribution of scattered light.

4.2.1. Effect of Flow Rate and Temperature

The first experiments using the mkll nephelometer were to try and characterise its response to experimental conditions of temperature and flow rate. For these tests the light source used was a CW FFT2000BHR LED launched through the fixed focal length probe.

Results from the flow rate experiment are plotted in figure 4.58 showing an increase in scattering as the flow rate increases and a lower transmitted power. This is concordant with cavitations or air bubbles scattering light away from the line of sight detector and towards the other sensors. Figure 4.59 shows the same data as a change from the average measure at a flow rate of $10 \text{ l/min}$. From this it can be seen that there is little effect until the flow rate exceeds $20 \text{ l/min}$. Also evident is an increase in the standard deviation of the measurements as the flow rate is increased, implying that the distribution of scattering centres is not uniform.

Although the experiments were made in as short a time as possible there was still some variation in temperature. The starting temperature was $21.1\,\text{C}$ and the temperature at the end of the experiment was $23.7\,\text{C}$. To judge if the temperature variations effected the results observed in this flow rate experiment the results of the temperature experiment need to be considered. These are given in figures 4.60 and 4.61 as raw data and as a change from an average initial measurement. Temperature effects were studied at flow rates of $10 \text{ l/min}$ and $20 \text{ l/min}$ and hence the two sets of results. The first observation to be made from these graphs is that they are similar in form for both flow rates but the $20 \text{ l/min}$ test is less effected by temperature. There is no evidence from these two experiments to counter the conclusion that at flow rates of under $20 \text{ l/min}$ the water turbulence is a minor influence on the background scatter. Using curve fits to the $10 \text{ l/min}$ data of figure 4.61(a) and compensating for temperature effects on the flow rate experiment gives an indication of how much of the observed variation in figure 4.59 was due to temperature and how much was due to scattering. Compensation was performed simply by subtracting from the raw data the voltage change indicated by the curve fit at each temperature the flow
Figure 4.58. Variation of raw data with flow speed using LED source.

Figure 4.59. $\Delta S$ for variations in flow speed using LED source.
Figure 4.60. Variation of raw data with temperature using LED source.
Figure 4.61. ΔS for variations in temperature using LED source.
rate measurements were made at. These temperatures were recorded by hand during each experiment. Compensated data is given in figure 4.62 which shows that most of the variation in the forward direction was due to scattering and that a significant proportion of the variations at 90°(=270°) and 55° was due to temperature.

A second observation made from figures 4.60 and 4.61 is that the effect of varying temperature is the opposite of that for the mkI nephelometer in that at higher temperatures there is less light transmitted and more detected off-axis. This indicates that either the beam profile is being radically altered by temperature or that there is more scattering. It was observed when the launch probe was removed after the experiment that there was some condensation on the glass window and it is probably this which is the cause of greater scattering to the angular receivers and less transmission to the line of sight receiver. This would also explain the two tests giving rise to different sized variations because the formation of condensation would not be repeatable. Because of the formation of condensation it is not possible to use these measurements to calibrate the sensor for temperature changes and compensate data obtained from other experiments. In later experiments attempts were made to stop condensation forming by not inserting the launch probe until the water had warmed and measurements were ready to commence. This was an attempt to reduce the temperature gradient across the window before blocking the circulation of air with the launch probe. Taking this precaution condensation on the glass window was not noticed again.

Figure 4.62. Temperature compensated ΔS for variations in flow speed using LED.
4.2.2. Scattering from Different Oil Types

Experiments with oil pollution made use of the OEG laser diode driver and sharp laser LTO24MDO. For automatic offset subtraction the laser had to be modulated through its lasing threshold. This shortens the life of the device which is why a CW LED was used for investigating temperature and flow effects. However, no problems with device reliability were found during the laser diode experiments. The drive current for the laser was switched between 72.0mA and 0.0mA using the constant current drive of the laser driver. Since the mkI nephelometer had demonstrated the ease of detecting and identifying soluble oil from the other oil types, the mkII experiments investigated only the crude oils and Castrol GTX. All receiver gains other than the line of sight detector were set to 800. The line of sight detector needed to be attenuated before reaching the pin diode to stop the receiver saturating. Sea water was used in these experiments to see if it led to any characteristics different to those of tap water.

Some results from these experiments are given in figures 4.63 to 4.68 for data obtained in separate trials as summarised in table 4.3.

<table>
<thead>
<tr>
<th>Oil Type</th>
<th>Figure Nos.</th>
<th>Flow Speed (l/min)</th>
<th>Starting Temp. (°C)</th>
<th>Finishing Temp. (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Castrol GTX</td>
<td>4.63(a)-(c)</td>
<td>20</td>
<td>30.3</td>
<td>35.4</td>
</tr>
<tr>
<td>Castrol GTX</td>
<td>4.64(a)-(c)</td>
<td>20</td>
<td>30.0</td>
<td>33.8</td>
</tr>
<tr>
<td>crude 1</td>
<td>4.65(a)-(c)</td>
<td>20</td>
<td>30.0</td>
<td>33.9</td>
</tr>
<tr>
<td>crude 1</td>
<td>4.66(a)-(c)</td>
<td>20</td>
<td>31.0</td>
<td>34.7</td>
</tr>
<tr>
<td>crude 2</td>
<td>4.67(a)-(c)</td>
<td>20</td>
<td>30.0</td>
<td>34.6</td>
</tr>
<tr>
<td>crude 2</td>
<td>4.68(a)-(c)</td>
<td>20</td>
<td>30.0</td>
<td>33.9</td>
</tr>
</tbody>
</table>

Table 4.3. Summary of experiments using mkII nephelometer.

These results clearly show difficulty in obtaining repeatable results and also a general similarity between all three oils in the angular nature of the scattering produced. Also evident from the results are the large variations using the dB scale which arise from small voltage variations. The small voltage variations indicate only a small amount of scattered light power reaching the detectors. Since the initial voltage measures at zero pollutant concentrations are also small there will be a large variation in power terms which leads to the high dB measurements. The low powers being detected also explain the apparent noisiness of the data. The electronic noise (after a gain of 800 and some averaging) is only
Figure 4.63. Results of experiment using laser diode and Castrol GTX.
Figure 4.63 continued. Results of experiment using laser diode and Castrol GTX.

Figure 4.64. Results of repeat experiment using laser diode and Castrol GTX.
Figure 4.64 continued. Results of repeat experiment using laser diode and Castrol GTX.
Figure 4.65. Results of experiment using laser diode and crude1.
(c) dB normalised data

Figure 4.65 continued. Results of experiment using laser diode and crude1.

(a) Raw data

Figure 4.66. Results of repeat experiment using laser diode and crude1.
Figure 4.66 continued. Results of repeat experiment using laser diode and crude1.
Figure 4.67. Results of experiment using laser diode and crude2.
Figure 4.67 continued. Results of experiment using laser diode and crude2.

Figure 4.68. Results of repeat experiment using laser diode and crude2.
Figure 4.68 continued. Results of repeat experiment using laser diode and crude2.
about 10mV which appears large because of the low signal level. More light power is being launched into the nephelometer in these tests than in the mkI experiments as is evident from the need to attenuate the line of sight receiver. The low detected powers must therefore be due to a lack of scattering or the scattered light not reaching the pin diode detectors. The direct laser diode launch does have a smaller focus than the fibre launched methods so that fewer scatterers would be illuminated but the difference is not that great and the higher power of the laser diode should compensate for this. It is likely therefore that the major problem is the combination of the lower numerical aperture receiver probes collecting less light and the attenuation caused by the series of lenses in the probes as discussed in chapter 3.

Given that only low light powers are being detected it is not surprising that the results are not accurately repeatable. Several factors will influence the repeatability of the experiments. The dB scaling has some sensitivity to the initial values and these will change from trial to trial as the launch probe is now refitted between trials to prevent and check on the formation of condensation. Also there is still the dependence of suspension size on how the oil is injected and although the chosen flow rate should have little effect, temperature effects are not clearly defined. All of these factors influence the repeatability of the experiments, and although in a system with more power reaching the detectors they may be insignificant, with the current low efficiency receivers they become more important because of the poor signal to noise ratio.

4.2.3. Neural Network Analysis

Despite the poor quality of the data obtained a neural network was trained with some success to estimate oil concentration and oil type. A similar concatenated type of MLP as before was used to provide an oil type input to the concentration estimator. Inputs to the concentration estimator were normalised as in (3.2) and (3.3) but an improved performance of the classifier was obtained using the normalisation:

\[ I_\theta(c) = \frac{S_\theta(c) - \langle S_\theta(0) \rangle}{\sigma_\theta(0)} \]  

(4.4)

where \( \sigma_\theta(c) \) is the standard deviation of the signal at angle \( \theta \) and concentration \( c \). There is no specific reason for using this type of normalisation other than it improves performance, it is in essence a measure of the signal to noise ratio which should become larger as the oil concentration increases. More complete details of the neural network and training can be found in references [4.1] and [4.9].
Figure 4.69. Concatenated ANN concentration estimation performance.

Output of network for oil concentrations 0 to 100 µl/l of crude 1, crude 2 and Castrol GTX. Two independent trials for each oil type shown in individual concentration registers.

Figure 4.70. ANN oil type classifier performance.

Output at each of three network outputs for oil concentrations 0 to 100 µl/l of crude 1, crude 2 and Castrol GTX indicating incorrect classification of crude 2 as GTX for one set of data. Two independent trials for each oil type shown in individual registers.
As would be expected the poor signal to noise ratio of the data compared to that obtained with the first nephelometer leads to a higher error in the concentration estimation. The mean error averaged over all test data in the concentration range 0 to 100µl/l was 28.4% which is large compared to the 5.4% mean error achieved with the mkI nephelometer but this result described graphically in figure 4.69 shows that the network recognises the general trend and that a large proportion of this average error is due to one (the final or second GTX) test set. The oil type classifier is successful 73% of the time. Again this error is due almost entirely to one test being misclassified as indicated by figure 4.70 where the third test set is classified exclusively as GTX when it was in actual fact crude 2. With this one exception the neural network does appear in general to recognise differences between the experiments. However, it is not clear that the network is responding to different scattering from oil to oil or if it is recognising individual experiments, i.e. experimental conditions. When an oil is present the network always makes a guess at it being one of the three oils and only on one occasion does it guess an oil type at zero concentration. The network therefore appears to recognise the presence of oil which lends credence to the idea that the network is responding to different scatter patterns. Because the test data is taken from the same experimental runs as the training data it is still possible that the network is recognising the particular data set that the data comes from instead of generalising trends. To clarify this uncertainty would require more experiments with which to train the network and other independent data for use in testing. One conclusion that can be drawn is that the neural network can distinguish between quite similar data sets with substantial noise on the inputs. This suggests that if enough reliable data could be obtained the neural network method would provide a useful tool in sizing and classifying different suspensions.

Using sea water instead of tap water had no observed influence on the scattering or performance of the nephelometer.

4.3. Summary of Nephelometric Study

Evidence from the nephelometric experiments suggests that different suspensions of oil scatter light with differing efficiencies and with different angular character. The difference in scattering appears to be more dependent on suspension size than the optical properties of the suspended oil. If the size distributions of a number of suspensions are sufficiently different then a neural network may be trained to recognise the different scatter patterns and identify the different suspensions. The mk I nephelometer with neural network processing was able to classify three different types of oil; soluble oil, crude 1 and a motorcycle fork oil.
The mk II nephelometer was designed to be more specific in the angular scattering it detected. This was to see if better definition of the angular scatter pattern aided the classification process. Some success was obtained in discerning suspensions of similar oils but the result is not as clear cut as with the mk I nephelometer. Further work is needed to confirm that the neural network was identifying different oils and not just different experimental conditions. The author feels that improvements to the mk II nephelometer optics should be made before continuing this investigation.

An estimate of the concentration of a suspension may be made from the scattered intensities detected by the nephelometers. There was enough information in the angular measurements to enable neural networks to adjust the concentration estimate according to the size of the suspensions. This is a significant result because current nephelometric techniques assume a linear relationship between the relative intensity of light scattered to different angles when estimating volume fraction. With neural network processing this assumption is not made, the network is left to form its own internal representation of the relationships between angular scattering and particle size.

Temperature has been observed to effect the measured intensities of light. For experimental apparatus repeatability is more important than absolute accuracy but the effects of temperature should be considered. This is likely to be of more concern if a sensor is to be used in a real application where there is less control or monitoring of temperature.
5. System for Speckle Statistic Experiments

Light intensity data from the nephelometric experiments showed no indication of being of any probability distribution other than Gaussian. To investigate the possible use of statistical optics further experiments were made off-line. Performing experiments on an optical table enables greater control of conditions than with the flow system.

Two separate apparatus were used to study different aspects of the scattering. The first experiments were to statistically characterise the receivers and check that the noise model was accurate. The other test looked at the spatial correlation of the speckle pattern as well as the point statistics. These different experiments are described in this chapter with results and analysis following in chapter 6.

5.1. Receiver Characterisation

The analogue detection system will add noise to the signal of interest affecting the statistics of that signal. To quantify the effect of this noise it is necessary to be able to measure the properties of the noise or to have a good noise model. Measurements were made with the pin diode receiver unit to compare the detector response to that of a noise model discussed in the next chapter.

The experimental procedure was to illuminate the pin diode detectors with a range of light powers and measure the statistical parameters of the output signal. Ideally for a steady illumination the output would be a dc signal. It was known that the average receiver output would not be zero with no illumination because of dark current and amplifier offset. The effect these have on the measured statistics expressed as normalised moments are of particular interest since it is proposed to use moments to identify different scattering processes.

Figure 5.1 shows a schematic of the apparatus used for this experiment. The power supplies were bench top supplies with variable output levels. Supply number 2 was used to vary the LED intensity by changing the supply voltage. A light emitting diode was used as the source because LEDs are the most
The general purpose data acquisition software written by D Barnett for the mkII nephelometer was also used since this measures the background (dark current and offsets) and the moments of the data which is all the information required. The LED was coupled to a plastic fibre which was used to deliver the light via the two lens nephelometer launch probe. The probe was placed inside a blackout box and pointed towards another fibre mounted on a linear positioning stage. By moving the fibre closer to or further from the focal point of the launch probe the amount of light coupled could be varied. The collected light was split using a homemade fibre coupler, one branch going to the racking receiver and the other to a commercial power meter, a Newport 835. The fibre coupler has a poor performance in terms of its efficiency (see table 5.1) but as it is used here this is an advantage since it helps to produce low light.

Figure 5.1. Apparatus for receiver characterisation.
powers, the region where the detector performance is of most interest. The reason for splitting the light was to enable the linearity and responsivity of the receivers to be measured.

<table>
<thead>
<tr>
<th>input</th>
<th>output</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>1</td>
<td>X</td>
</tr>
<tr>
<td>2</td>
<td>10.5</td>
</tr>
<tr>
<td>3</td>
<td>7.9</td>
</tr>
</tbody>
</table>

Table 5.1. Loss matrix (dB) for fibre coupler measured at 850nm.

Statistical data were calculated from 15000 samples, 7500 with the source off to measure the background and 7500 with the source on to measure the receiver response. The optical power incident on the detectors was varied over a range from approximately 10nW to under 0.05nW. Two channels of the receiver unit were tested, one with a positive offset and one with a negative offset. Results of this experiment are presented in the following chapter, figures 6.1 and 6.3.

5.2. Speckle Characterisation

Experiments to investigate the dynamic speckle were performed using a linescan camera to obtain spatially resolved measurements of the scattered light. This camera is described as a photodiode array but the output is integrated and read as with a CCD. The 1024 element camera [5.1] was taken from an Oriel spectragraph II spectrometer which could be controlled from a PC. Thus the camera had two important features which are closely spaced, small detection areas and the ability for software to control the array and perform some of the processing, avoiding the need to save large amounts of individual data.

The linescan camera experiment was set up as shown in figure 5.2. The HeNe source was the most powerful available and the easiest to focus. The lens and pin hole arrangement was to provide some spatial filtering of the laser beam in order to reduce fine structure observed in the beam profile. A cuvette was used to hold the samples and was set at an angle to the laser beam so that reflections off the cuvette walls did not interfere with the light scattered towards the detector. A continuously variable speed stirrer was used to agitate the sample with a paddle made from a brass rod and small washer twisted into a shallow propeller shape. The camera itself was positioned about a metre away (sufficient
Figure S.2. Apparatus for linescan camera experiments.
distance to be able to spatially resolve the speckle) from the cuvette and in the forward direction where scattering is strongest. A range of angles from approximately 5° to 15° from the laser beam axis is covered by the array of detectors. A cardboard light baffle painted black on the inside was placed over the optics, camera and cuvette to block out reflections and as much extraneous light as possible. Two holes were cut in the baffle, one through which the stirrer was inserted and the other through which the laser was launched. A shutter was placed between the laser and the entry hole in the baffle so that the source could be modulated mechanically. The shutter was driven by a stepper motor which was triggered from the PC. The drive electronics for the stepper motor are described in appendix A.

The linescan camera is controlled from a PC with a special ISA bus card inserted. Software can be run on the PC to communicate with the camera through this board and several signals are accessible through a connector on the board. The software for the experiment, written in a form of Basic, is reproduced in appendix C. When the motor drive circuit returns a trigger pulse a single scan is taken from the camera. Readings are taken from the photodiode array with the minimum integration time possible of 20ms in an attempt to reduce time averaging of the speckle. Any averaging of the speckle will reduce the size of the observed fluctuations and hence the size of the moments will tend to those of a Gaussian distribution. Measurements of the background are taken first to check the offset levels of the array and to check that the noise is Gaussian as expected for thermal noise in the electronics. A number of measurements are usually taken in a row, calculating cumulants as the experiment progresses. Once the shutter has opened and the trigger signal has been taken high a series of measurements of the scattered light intensities are made. During these measurements of the scattered light, the array is checked to see if any of the elements have been saturated. If it is found that the array has been saturated a subroutine is called to make a note of the number of times such an event occurs. A later addition to the program was to store the minimum value of intensity measured at each element across the array. This was to give some idea of what any static speckle was like, it assumes that the total measured intensity is the sum of the static speckle intensity and dynamic speckle intensity, that is there is no interference between the two. This will only be the case if the source of static speckle is far enough away from the scatterers such that the interference is too small to resolve.

After the program has looped the required number of times such that a minimum number of samples have been collected, a test is made on the size of the moments in comparison to the number of samples to see how reliable they are. The test is based on the fact that if the moments are reliable then the variance of the moments will be small compared to the square of the moments themselves. It should be pointed out that this is a necessary condition and not a sufficient condition so it provides a guide to reliability rather than a definitive measure. The argument behind the test is as follows. Consider the $M$ sets of $L$ samples.
and the combination set
\[ Y = \{ X_1, X_2, \ldots, X_M \} \]  

The \( r \)th moment of the set \( X \) is
\[ \bar{X}_r = \frac{1}{L} \sum_{i=1}^{L} x_{ri} \]  

and that of the set \( Y \) is
\[ \bar{Y}_r = \frac{1}{LM} \left( \sum_{i=1}^{L} x_{ri} + \sum_{i=2}^{L} x_{ri} + \ldots + \sum_{i=M}^{L} x_{ri} \right) = \left( \bar{X}_r \right) \]  

It is necessary to calculate a limit for the value of \( \left( \bar{X}_r^2 \right)^2 \) as follows.
\[ \left( \bar{X}_r \right)^2 = \frac{1}{L^2} \sum_{i=1}^{L} x_{ri} \sum_{j=1}^{L} x_{rj} \]  
\[ = \frac{1}{L^2} \left( \sum_{i=1}^{L} x_{ri}^2 + 2 \sum_{i<j} x_{ri} x_{rj} \right) \]  

For simplicity substitute \( x_{ri} = w_{iz} \) and \( w_{iz} = \bar{w}_z + v_{iz} \) where \( \bar{w}_z \) is the mean of \( w_{iz} \) and \( v_{iz} \) is a random variable of zero mean. Thus
\[ \left( \bar{X}_r \right)^2 = \frac{1}{L^2} \left( \sum_{i=1}^{L} x_{ri}^2 + 2 \sum_{i<j} (v_{ia} + \bar{w}_z)(v_{ja} + \bar{w}_z) \right) \]  
\[ = \frac{\bar{X}_r^2}{L} + \frac{2}{L^2} \sum_{i<j} (v_{ia} v_{ja} + v_{ia} \bar{w}_z + v_{ja} \bar{w}_z + (\bar{w}_z)^2) \]  
\[ = \frac{\bar{X}_r^2}{L} + \frac{2(L-1)}{L^2} (\bar{w}_z)^2 + \frac{2}{L^2} \sum_{i<j} v_{ia} v_{ja} \]
since \( v_{iz} \) has zero mean. The last term in (5.9) may be expanded to give

\[
\left( \bar{X}_z' \right)^2 = \frac{X_{zr}^2}{L} + \frac{2(L-1)}{L^2} \left( \bar{X}_z' \right)^2 + \frac{1}{L^2} \left( \sum_{i=1}^{L} v_{iz} \sum_{j=1}^{L} v_{jz} - \sum_{i=1}^{L} v_{iz}^2 \right)
\]  

(5.10)

which reduces, again because \( v_{iz} \) has zero mean, to

\[
\left( \bar{X}_z' \right)^2 = \frac{1}{L} \left( \frac{X_{zr}^2}{L} + 2(L-1) \left( \bar{X}_z' \right)^2 - \bar{v}_z^2 \right)
\]  

(5.11)

This can be rewritten as an inequality

\[
\left( \bar{X}_z' \right)^2 \leq \frac{1}{L} \left( \frac{X_{zr}^2}{L} + 2 \left( \bar{X}_z' \right)^2 - \bar{v}_z^2 \right)
\]  

(5.12)

After rearranging (5.12) becomes

\[
\left( \bar{X}_z' \right)^2 \leq \frac{X_{zr}^2 - \bar{v}_z^2}{L-2}
\]  

(5.13)

which is a maximum when \( \bar{v}_z^2 \) is zero, hence

\[
\left( \bar{X}_z' \right)^2 \leq \frac{X_{zr}^2}{L-2}
\]  

(5.14)

If the sets \( X_z \) are representative of the set \( Y \), then the variance of the moments of \( X_z \) will be small compared to the mean squared value of the moments as noted earlier. That is

\[
\text{var}(\bar{X}_z') \ll \left( \bar{X}_z' \right)^2
\]  

(5.15)

where

\[
\text{var}(\bar{X}_z') = \left( \left( \bar{X}_z' \right)^2 \right) - \left( \bar{X}_z' \right)^2
\]  

(5.16)

Substituting for \( \left( \bar{X}_z' \right)^2 \) from (5.14) leads to the inequality

\[
\text{var}(\bar{X}_z') \leq \left( \frac{X_{zr}^2}{N-2} \right) - \left( \bar{X}_z' \right)^2
\]  

(5.17)

the maximum value of which may be substituted in (5.15) so that

\[
\frac{X_{zr}^2}{L-2} - \left( \bar{X}_z' \right)^2 \ll \left( \bar{X}_z' \right)^2
\]  

(5.18)
which may be written in terms of \( Y \) using (5.4)

\[
L \gg \frac{\bar{Y}^{2r}}{2^{\frac{1}{r}}} + 2 = L_{\text{crit}} \tag{5.19}
\]

\( L_{\text{crit}} \) is a number relating to the quantity of samples required for a reliable measurement of the \( r \) th moment. To obtain this number it is necessary to calculate the \( 2r \) th moment. What number of samples is actually needed for an accurate measurement of moments is undefined but a minimum of 10 times \( L_{\text{crit}} \) seems a reasonable estimate. Figures 5.3 and 5.4 show \( L_{\text{crit}} \) as calculated for Gamma and \( K \) distributions. For more skewed distributions \( L_{\text{crit}} \) is correspondingly larger. The expression used in the software for calculating \( L_{\text{crit}} \) differs from (5.19) slightly in that just the ratios of the cumulants are calculated as in (5.20). This is just to save some processing. If this test indicates that the number of samples collected is sufficient then the programme continues and data is saved to disk. If the test suggests that more samples should be taken, the ratio of the cumulants is printed to the screen as a factor describing the reliability of the measured statistics. The operator can then decide whether to stop the experiment and save the data or to continue by doubling the length of the experiment.

\[
\frac{\sum_{i=1}^{N} Y_i^{2r}}{\left( \sum_{i=1}^{N} Y_i \right)^{\frac{2r}{2}}} = LM \frac{\bar{Y}^{2r}}{\left( \bar{Y}^{r} \right)^{2}} \tag{5.20}
\]

At the end of the experiment the cumulants are scaled into moments by dividing by the number of samples taken. The calculation of the correlations are also completed by dividing the cumulant calculated earlier by the square root of the product of second moments so that the final correlations are calculated as

\[
\tau = \frac{\langle \mathcal{A}_1 \mathcal{A}_2 \rangle}{\sqrt{\langle r^2 \rangle} \langle r^2 \rangle} \tag{5.21}
\]
Figure 5.3. $L_{\text{crit}}$ for Gamma distributions.

Figure 5.4. $L_{\text{crit}}$ for K distributions.
5.3. Particle Size Distributions

A number of different types of test particles were available for use in these off-line experiments. Those of most use were polystyrene calibration spheres, glass spheres (ballotini), soluble oil and "desert dust". The calibration spheres were of a monodispersion of 2.93\( \mu \)m. The ballotini were larger and of a broader size distribution. Soluble oil forms a suspension in water, the size of the oil droplets depending to an extent on how the suspension is mixed but tending to be mostly less than 2\( \mu \)m. Figures 5.5 and 5.6 show size distributions of soluble oil suspensions as measured using a Coulter Counter. Figures 5.7 to 5.9 show the size distributions of the polystyrene spheres, ballotini and the desert dust, a sandy type of soil.

![Size distribution of soluble oil measured using Coulter Counter.](image)
Figure 5.6. Size distribution of soluble oil measured using Coulter Counter.

Figure 5.7. Size distribution of polystyrene calibration spheres.
Figure 5.8. Size distribution of Ballotini measured using Coulter Counter.

Figure 5.9. Size distribution of desert dust measured using Coulter Counter.
6. Discussion of Speckle Statistical Results

6.1. Receiver Characterisation

The results from the experiments to characterise the detectors and receivers should be viewed in relation to the type of noise expected and how this will affect the measure of moments expected from a scattering process. It will be convenient later to refer to the scattering models introduced in chapter 2 but first a model for the statistics of the detector signal and noise is developed.

6.1.1. Receiver Noise Model

The photon generated reverse current $i_{ph}$ is estimated via the relationship \[6.1\]

$$i_{ph} = \eta P q = RP$$

where $q$ is the charge on an electron, $\eta$ is the quantum efficiency of the diode, $R$ the responsivity and $P$ is the incident optical power. There are two types of noise generated in the receiver, shot noise due to the discrete nature of the detection process, and Johnson noise arising from thermal effects in the load resistor. These two noise currents, $i_{sh}$ and $i_{th}$, have mean square values of

$$\langle i_{sh}^2 \rangle = 2q(i_{ph} + i_d)B$$

(6.2)

and

$$\langle i_{th}^2 \rangle = \frac{4KTB}{R_{load}}$$

(6.3)

where $i_d$ = dark current, $B$ = bandwidth, $K$ = Boltzman constant and $T$ is the absolute temperature. The shot noise current is Poisson distributed and the Johnson noise is Gaussian distributed. The moment generating function of a Gaussian or normal distribution is as stated in (2.91) and thus the moment generating function for the thermal noise distribution is
\[ \Phi_{ph}(t) = \exp \left( i_{oa} t + \frac{\langle i_{oa}^2 \rangle t^2}{2} \right) \] (6.4)

where \( t_{oa} \) is the mean noise current or offset caused by amplifier bias currents and referred to the input. The moments of the shot noise current pdf must be calculated by a transformation of variables from discrete numbers of photons to current. The mean shot noise current \( \langle i_{sh} \rangle = qB\langle n \rangle \), and all associated current moments may be generated from

\[ \Phi_{sh}(t) = \exp \left( -\langle n \rangle + \langle n \rangle e^{qBt} \right) \] (6.5)

\( \langle n \rangle \), the mean number of electrons, and hence \( \langle i_{sh} \rangle \) may be calculated from (6.2) and the derivatives of (6.5).

If the scattered signal of interest is also considered as a type of noise source with a moment generating function \( \Phi_{sc}(t) \), then the composite distribution of shot noise, thermal noise and detected photocurrent may be obtained by the convolution theorem,

\[ \Phi(t) = \Phi_{sc}(t) \Phi_{sh}(t) \Phi_{ph}(t) \] (6.6)

The moments of the combination may therefore be calculated thus

\[ \Phi'(t) = \frac{d(\Phi_{sh}(t)\Phi_{ph}(t))}{dt} \Phi_{sc}(t) + \Phi_{sh}(t)\Phi_{ph}(t) \frac{d(\Phi_{sc}(t))}{dt} \] (6.7)

\[ = \left( \langle n \rangle qBe^{qBt} + i_{oa} + \langle i_{oa}^2 \rangle \right) \Phi_{sh}(t) \Phi_{ph}(t) \Phi_{sc}(t) + \Phi_{sh}(t) \Phi_{ph}(t) \Phi_{sc}'(t) \] (6.8)

Evaluated at \( t=0; \)

\[ \Phi'(0) = \langle i \rangle = \langle i_{sh} \rangle + i_{oa} + \Phi_{sc}'(0) \] (6.9)

\[ = \langle i_{sh} \rangle + i_{oa} + \langle i_{ph} \rangle \] (6.10)

which is the first moment. Higher moments may be found by differentiating further, giving after some manipulation

\[ \langle i^2 \rangle = \left( \langle i_{ph}^2 \rangle + 2[\langle i_{oa} + \langle i_{sh} \rangle] \langle i_{ph} \rangle + [\langle i_{oa} + \langle i_{sh} \rangle]^2 + \langle i_{oa} \rangle qB + [\langle i_{oa} \rangle^2 \right) \] (6.11)
\[ \langle i^3 \rangle = \langle i_{ph}^2 \rangle + 3\langle i_{ph} \rangle \langle i_{th} \rangle \langle i_{ph} \rangle + 3\langle i_{ph} \rangle^2 \langle i_{th} \rangle + \langle i_{ph} \rangle^3 \]
\[ + 3(i \langle i_{th} \rangle qB + \langle i_{th}^2 \rangle + qB^2 \langle i_{th} \rangle) \]  
(6.12)

In the case of a scattered light intensity that does not fluctuate, that is the receivers being illuminated by a constant power, \( \langle i_{ph}^2 \rangle = \langle i_{ph} \rangle^2 \) and \( \langle i_{ph}^3 \rangle = \langle i_{ph} \rangle^3 \) so that the normalised moments are

\[ f^{[2]} = \frac{\langle i_{ph}^2 \rangle + 2\langle i_{ph} \rangle \langle i_{th} \rangle \langle i_{ph} \rangle + \langle i_{ph} \rangle^2 \langle i_{th} \rangle}{\langle \langle i_{ph} \rangle^2 + \langle i_{ph} + \langle i_{th} \rangle \rangle^2 \rangle} + \frac{\langle i_{th} \rangle qB + \langle i_{th}^2 \rangle}{\langle i \rangle} \]  
(6.13)

\[ = 1 + \frac{\langle i_{th} \rangle qB + \langle i_{th}^2 \rangle}{\langle i \rangle} \]  
(6.14)

and

\[ f^{[3]} = \frac{\langle i_{ph}^3 \rangle + 3\langle i_{ph} \rangle \langle i_{th} \rangle \langle i_{ph} \rangle^2 + \langle i_{ph} \rangle \langle i_{ph} \rangle^2 \langle i_{th} \rangle + \langle i_{ph} \rangle^3 \langle i_{th} \rangle + 3(i \langle i_{th} \rangle qB + \langle i_{th}^2 \rangle + qB^2 \langle i_{th} \rangle)}{\langle \langle i_{ph} \rangle^2 + \langle i_{ph} + \langle i_{th} \rangle \rangle^2 \rangle} + \frac{3(i \langle i_{th} \rangle qB + \langle i_{th}^2 \rangle + qB^2 \langle i_{th} \rangle)}{\langle i \rangle^3} \]  
(6.15)

\[ = 1 + 3 \frac{\langle i_{th} \rangle qB + \langle i_{th}^2 \rangle}{\langle i \rangle^2} + \frac{qB^2 \langle i_{th} \rangle}{\langle i \rangle^3} \]  
(6.16)

The normalised third moment may be written in terms of the second as

\[ f^{[3]} = 3f^{[2]} - 2 + \frac{qB^2 \langle i_{th} \rangle}{\langle i \rangle^3} \]  
(6.17)

which shows the moments to be similar to those of a Gaussian distribution with an extra term added due to the shot noise. This extra term tends to be very small due to the small value of \( q^2 \). Measurements made of the receiver characteristics bear this out as indicated in figure 6.1, the normalised moments closely adhering to the linear relationship for Gaussian distributions.

From equations (6.14) and (6.16) it can be seen that the values of the normalised moments depend on the mean shot noise, rms thermal noise, mean total signal and the bandwidth of the receivers. The bandwidth can be fixed by design to a suitable value and the rms thermal noise will be a constant for a defined temperature. The shot noise and mean measured signal however vary with photocurrent and offset so that even if the incident light power is constant the measured moments of the receiver output will vary with that power. A constant incident power of course looks like an offset so the offset of the receivers also affect the measured moments. Figure 6.2 shows how the normalised second moment is expected to vary with power at various values of receiver offset. Higher order moments follow a
Figure 6.1. Relationship between normalised moments for pin diode receiver.

\[ y = 3.0005x - 2.0005 \]
\[ R^2 = 1 \]

Figure 6.2. Theoretical variation of normalised moment with optical power.
similar trend as can be deduced from (6.17) for the normalised third moment. In calculating the graph of figure 6.2 the following parameters were used with equations (6.14), (6.1), (6.2), (6.5) and (6.3); $R=0.44 \text{ AW}^{-1}$, $i_d=0.5 \text{ nA}$, $B=17.4 \text{ kHz}$, $R_{\text{load}}=1\text{ M} \Omega$, $T=273\text{ K}$. The value of dark current was taken from the data sheet for the IRS50LC pin diodes used, the load resistance is the same as used in the receivers and the bandwidth and responsivity are as measured for the receivers. With no offset the normalised moments are expected to increase as the power decreases. This is due to the denominator in (6.14) tending to zero. At low optical powers the moments are sensitive to offset but above a threshold power, in this case about 2nW, the normalised moments tend towards the value of one expected for a non-fluctuating source. Positive offset limits the extent to which the denominator of (6.14), and of (6.16) and similar terms for higher orders, can approach zero and so limits the maximum value of the normalised moments. With a negative offset there will be a power at which the photocurrent equals the offset and the mean detected current will be zero. The normalised moments will then tend towards infinity as figure 6.2 shows at a power of approximately -7dBm. At low powers the absolute error in the measured moments can be huge suggesting a lower limit of optical power for which the receivers can be used to make useful measurements. This limit can be pushed to lower powers if the offsets can be measured.

6.1.2. Comparison of Receiver Noise and Model

Figure 6.3 shows data obtained using two channels of the receiver unit which indicate a reasonable fit to the theoretical expectations. When the measured offsets are subtracted the data for both channels approximately fits the theoretical curve for no offset. It is believed that the fit could be improved by adding a noise figure for the amplifiers which have so far been ignored. The noise model refers only to the photodiode and load resistor.

6.1.3. Deconvolution of Receiver Noise

(6.14) and (6.16) were calculated assuming a non fluctuating source. If for the time being it is assumed that the source is fluctuating according to a Gamma distribution, the normalised second and third moments may be predicted by using in (6.6) and subsequent equations the moment generating function

\[
\Phi_{sc}(t) = \left(1 - \frac{i_{\text{ph}}}{\alpha}\right)^{-\alpha}
\]  

(6.18)
Using the same method as before, the normalised second and third moments can be predicted to be

\[ J_2 = 1 + \frac{\langle i_{sh} \rangle qB + \langle i_{ph} \rangle^2}{\langle i \rangle^2} + \frac{\langle i_{ph} \rangle^2}{\alpha \langle i \rangle^2} \]  
\[ J_3 = 1 + \frac{\langle i_{sh} \rangle qB + \langle i_{ph} \rangle^2}{\langle i \rangle^2} + \frac{(qB)^2 \langle i_{sh} \rangle}{\langle i \rangle^3} + \frac{3 \langle i_{ph} \rangle^2}{\alpha \langle i \rangle^2} + \frac{2 \langle i_{ph} \rangle^3}{\alpha^2 \langle i \rangle^3} \] 

These are the same as the normalised moments for the Gamma distribution on its own with added terms for the noise of the receiver system. It is clear by inspection that for a scattering process with a large enough mean the noise terms will be negligible. What is not obvious is how the value of the normalised moments will change at low mean powers and how this will effect the relationship between \( J_2 \) and \( J_3 \).

In general it is the statistics of the scattering alone that are of interest rather than the combination of the scattering and electronic noise. A method of observing the desired pdf in isolation is to remove the effects of offset and noise from the measured moments directly. An analytic method for this process may be derived starting from (6.6). Introducing the shorthand notation \( J'(\Phi_{sh}(t)\Phi_{ph}(t)) = \Phi_{sc}(t) \) the scattering moment generating function can be differentiated to any order using the Leibnitz theorem:
\[
\frac{\partial^m \Phi_{xc}}{\partial t^m} = \Phi^{(n)} \Phi_e + n \Phi^{(n-1)} \Phi_e^{(1)} + \cdots + \frac{n!}{r!(n-r)!} \Phi^{(n-r)} \Phi_e^{(r)} + \cdots + \Phi^{(n)} e^{(r)}
\]

(6.21)

where
\[
\Phi^{(n)} = \frac{\partial^n \Phi(i)}{\partial t^n}
\]

(6.22)

From (6.21) the \(n\)th order moment can be obtained. Using (6.2) to (6.5) as before, the first three scattering moments are found to be

\[
\langle i_{ph} \rangle = \langle i \rangle - i_{os} - \langle i_{sh} \rangle
\]

(6.23)

\[
\langle i_{ph}^2 \rangle = \langle i^2 \rangle - 2 \left[ i_{os} + \langle i_{sh} \rangle \right] \langle i \rangle + \left[ i_{os} + \langle i_{sh} \rangle \right]^2 - \langle i_{sh} \rangle \langle qB \rangle
\]

(6.24)

\[
\langle i_{ph}^3 \rangle = \langle i^3 \rangle - 3 \left[ i_{os} + \langle i_{sh} \rangle \right] \langle i^2 \rangle + 3 \left[ i_{os} + \langle i_{sh} \rangle \right]^2 \langle i \rangle - \langle i_{sh} \rangle \langle qB \rangle \langle i \rangle + \left[ i_{os} + \langle i_{sh} \rangle \right] \left[ 3 \left( \langle i_{sh} \rangle + \langle qB \rangle \right) - \left[ \langle i_{sh} \rangle + \langle qB \rangle \right]^2 \right] - \langle i_{sh} \rangle \langle qB \rangle^2
\]

(6.25)

which is simply a rearrangement of (6.10-6.12). Normalising with respect to \(\langle i_{ph} \rangle\) gives finally

\[
\frac{I_{ph}^{[2]}}{I_{ph}^{[2]}} = 1 + \frac{\langle i^2 \rangle - \langle i \rangle^2 - \langle i_{sh} \rangle qB + \langle i_{sh}^2 \rangle}{\langle i_{ph} \rangle^2}
\]

(6.26)

\[
\frac{I_{ph}^{[3]}}{I_{ph}^{[3]}} = 1 + \frac{\langle i^3 \rangle - \langle i \rangle^3 - 3 \langle i_{os} + \langle i_{sh} \rangle \rangle \langle i^2 \rangle - \langle i \rangle^2 - 3 \langle i_{sh} \rangle qB + \langle i_{sh}^2 \rangle + \langle i_{ph} \rangle \langle qB \rangle^2}{\langle i_{ph} \rangle^3}
\]

(6.27)

When there is no scattering source, that is with only the instrument noise present, it is to be expected that all normalised, deconvolved moments will be indeterminate due to the vanishing first moment.

When scattering sources are present the mean photocurrent is finite and the subsequent moments are those of the source without noise. The size of change in moments due to this noise deconvolution varies with the signal to noise ratio of the original data. With a high signal to noise ratio the shift is relatively small but as the signal to noise ratio reduces the shift becomes larger because the noise contributes more to the overall signal statistics.

The results of the receiver characterisation experiments indicate that the noise added to the signal by the receivers is Gaussian in nature, figure 6.1, and that the shot noise is not significant. In this case equations (6.23) to (6.25) simplify to

\[
\langle i_{ph} \rangle = \langle i \rangle - i_{os}
\]

(6.28)
\[
\left\langle i_{\mu}^2 \right\rangle = \left\langle i^2 \right\rangle - 2i_{\mu} \left\langle i \right\rangle + 2i_{\mu}^2 - \left\langle i_{\mu}^2 \right\rangle \tag{6.29}
\]

\[
\left\langle i_{\mu}^4 \right\rangle = \left\langle i^4 \right\rangle - 3\left\langle i^2 \right\rangle i_{\mu} + 3\left\langle i \right\rangle \left(2i_{\mu}^2 - \left\langle i_{\mu}^2 \right\rangle \right) + 3i_{\mu}^2 \left\langle i_{\mu}^2 \right\rangle - 4i_{\mu}^2 \tag{6.30}
\]

By modulating the light source during an experiment and measuring the statistical behaviour of the instrumentation whilst the source is turned off, the offset and noise can be compensated for using the above formulae. This should lead to improved accuracy, specially at low light powers. Also the noise can be monitored to see that it remains Gaussian in nature as a check that the instrumentation is working as expected. It is noted that this compensation will work not just for photodiodes but for any type of receiver which has a Gaussian noise characteristic. This method of noise compensation has been used with the data now discussed.

### 6.2. Discussion of Point Statistics

Data obtained using the linescan camera has two aspects of interest; the individual point statistics and the spatial relationships. The individual point statistics from several experiments investigating the apparatus and several types of scattering material will be considered first. A summary of the different experiments performed is given in the table below.

<table>
<thead>
<tr>
<th>experiment / suspension</th>
<th>stirrer speed, rpm</th>
<th>number of data samples</th>
<th>reliability measure</th>
<th>notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>empty cuvette (i)</td>
<td>0</td>
<td>10 000</td>
<td>192</td>
<td></td>
</tr>
<tr>
<td>empty cuvette (ii)</td>
<td>0</td>
<td>8 000</td>
<td>968</td>
<td></td>
</tr>
<tr>
<td>tap water (i)</td>
<td>100</td>
<td>8 000</td>
<td>726</td>
<td></td>
</tr>
<tr>
<td>tap water (ii)</td>
<td>50</td>
<td>20 000</td>
<td>673</td>
<td></td>
</tr>
<tr>
<td>polystyrene spherules (i)</td>
<td>100</td>
<td>20 000</td>
<td>904</td>
<td></td>
</tr>
<tr>
<td>polystyrene spherules (ii)</td>
<td>50</td>
<td>10 000</td>
<td>124</td>
<td></td>
</tr>
<tr>
<td>polystyrene spherules (iii)</td>
<td>50</td>
<td>10 000</td>
<td>145</td>
<td>continuation of previous expt.</td>
</tr>
<tr>
<td>desert dust (i)</td>
<td>50</td>
<td>10 000</td>
<td>847</td>
<td></td>
</tr>
<tr>
<td>desert dust (ii)</td>
<td>50</td>
<td>10 000</td>
<td>222</td>
<td></td>
</tr>
<tr>
<td>desert dust (iii)</td>
<td>50</td>
<td>10 000</td>
<td>288</td>
<td>follow on from tap water (ii)</td>
</tr>
<tr>
<td>soluble oil (i)</td>
<td>50</td>
<td>10 000</td>
<td>556</td>
<td></td>
</tr>
<tr>
<td>soluble oil (ii)</td>
<td>50</td>
<td>5 000</td>
<td>414</td>
<td></td>
</tr>
<tr>
<td>ballotini (i)</td>
<td>200</td>
<td>5 000</td>
<td>1021</td>
<td></td>
</tr>
<tr>
<td>ballotini (ii)</td>
<td>200</td>
<td>5 000</td>
<td>706</td>
<td>continuation of ballotini (i)</td>
</tr>
<tr>
<td>ballotini (iii)</td>
<td>200</td>
<td>10 000</td>
<td>826</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.1. Summary of Experiments on Speckle Statistics.
6.2.1. Empty Cuvette Experiments

The first and most basic results to look at are those from an experiment with nothing in the cuvette. Plots of the dark current moments, moments of scattered light intensity and deconvolved intensity moments are shown in figures 6.4 to 6.6. Points in the plot of the dark current moments line up accurately (correlation coefficient equals 0.9999 to 4 decimal places) with the \( \mu^2 = 3\sigma^2 - 2 \) relationship for a Gaussian distribution. The electronic noise is therefore seen to be typical thermal noise. There being no water or suspended particles in the cuvette it is expected that the detected speckle, arising from scattering off the surfaces of the cuvette, would be stationary leading to normalised moments of value one modulated by the electronic noise. Deconvolved moments should account for the noise and lead to normalised moments that are equal to one. The normalised moments of the measured signal are indeed closely correlated with a Gaussian distribution (a constant convolved with a Gaussian is a Gaussian) and close in value to one. The small deviation of the relationship between the normalised second and third moments from that for a true Gaussian distribution is probably due to mathematical rounding errors since moments lower than those for a Gaussian are not possible.

Some items of note about the deconvolved moments are evident in figure 6.6. Firstly, the deconvolved moments are not all equal to 1 but are spread along a line. The fact that the moments are correlated rather than randomly spread suggests that there is some stochastic variation in the speckle intensity. Various mechanisms for such a stochastic process can be proposed, for example dust in the air, mechanical vibrations or source power variations. The receivers can be ruled out as the cause since the effect would have to be caused by the presence of a constant illumination. Shot noise depends on the mean photocurrent but should have a Poisson distribution. Other mechanisms for increased noise under illumination are hard to conceive of. Possibly heating of the silicon could lead to increased thermal noise although such heating is unlikely with cooled detectors and the low powers of scattered light observed.

An alternative explanation for the residual statistics, and one which the evidence strongly supports, is that they arise from the deconvolution process. If the original measure of the background noise is not precise then the deconvolved moments will not be exact. A brief analysis shows what effects follow. Using the same notation as before and introducing the moment generating functions \( \Phi_m(t) \) for the measured electronic noise and \( \Phi_d(t) \) for the deconvolved mgf, the following relationships can be written;
Figure 6.4. Dark current moments for empty cuvette experiment.

Figure 6.5. Measured light intensity moments for empty cuvette experiment.
\[ \Phi'(l) = \Phi_{\delta}(l)\Phi_{sc}(l) \]  
\[ \Phi_{\delta}(l) = \frac{\Phi(t)}{\Phi_n(t)} = \frac{\Phi_{\delta}(t)}{\Phi_{sc}(t)} \]  

The thermal noise and measured noise are both known to be from a Gaussian distribution and so their moment generating functions can be expanded as

\[ \Phi_{\delta}(t) = \exp \left( \langle i_{\delta} \rangle t + \frac{\langle i_{\delta}^2 \rangle t^2}{2} \right) \]  
\[ \Phi_n(t) = \exp \left( \langle i_n \rangle t + \frac{\langle i_n^2 \rangle t^2}{2} \right) \]

Thus

\[ \frac{\Phi_{\delta}(t)}{\Phi_n(t)} = \exp \left( \langle i_{\delta} \rangle - \langle i_n \rangle \right) t + \frac{\left( \langle i_{\delta}^2 \rangle - \langle i_n^2 \rangle \right) t^2}{2} \]
which still has a Gaussian type of form. Clearly if the measured noise statistics exactly match those of
the electronic noise this combined moment generating function will have the value 1 and the true
scattering statistics will be recovered. If there is some difference between the electronic noise and the
measured noise then there will be a residual Gaussian type noise convolved with the scattering. With a
constant scattered light intensity this means that the deconvolved moments will be that residual
Gaussian type noise. Equation (6.35) is not a moment generating function of a real Gaussian
distribution as can be demonstrated by determining the moments.

\[
\frac{\partial}{\partial \alpha} \left( \Phi_n \right)_{\alpha=0} = \langle i_n \rangle - \langle i_n \rangle
\]

(6.36)

\[
\frac{\partial^2}{\partial \alpha^2} \left( \Phi_n \right)_{\alpha=0} = \langle i_n^2 \rangle - 2 \langle i_n \rangle + \left( \langle i_n \rangle - \langle i_n \rangle \right)^2
\]

(6.37)

If \( \langle i_n^2 \rangle \) is greater than \( \langle i_n \rangle \) as is quite possible, then the normalised second moment will be less than
one as will all higher moments. In this sense the combination mgf of (6.35) is not valid of a true
distribution. The fact that such impossibly low moments appear in figure 6.6 is evidence that the
deconvolution process is the cause of at least some of the residual Gaussian type noise. Inspection and
comparison of figures 6.5 and 6.6 show that the noise deconvolution process has lead to normalised
moments shifting both towards the origin and away from it. The moments of less than one can only be
caused by the deconvolution process but the shift to higher values may or may not be caused by the
deconvolution. Equation (6.36) shows the noise deconvolved mean to be the difference between the
measured noise mean and the actual noise mean. This can approach zero making the normalised
moments large. Thus the shift of normalised moments away from the origin in figure 6.6 could be due
either to the deconvolution process or to the experimental possibilities mentioned earlier.

Having shown that the deconvolution process may increase the spread of normalised moments the
question is raised of what worth is this noise deconvolution. It must be remembered that so far only the
case of constant power has been investigated in any detail. Also in plots of normalised moments it is
the relationship between the different orders of moments that is of as much interest as the actual values.
If (6.35) is considered again it can be seen that in general the combination moment generating function
is that of a Gaussian with smaller mean and smaller variance than that of the original electronic noise.
As such the deconvolved unnormalised moments are equivalent to those of the scattering convolved
with this more delta function like distribution. As a result the deconvolved unnormalised moments will
be more accurate, the precise values of the normalised moments depending on the mean. A constant
scattered light power is a special case in which the mean photocurrent makes no difference to the
normalised moments.
Because all the signal distributions have been identified as Gaussian / normal, described fully by their means and standard deviation, it is possible to calculate the pdfs using the measured means and variances. Probability density functions are shown in figure 6.7 for the electronic noise and speckle signal. The two distributions with the lower means are the narrowest and broadest electronic noise distributions from across the array. The broad distribution is more typical of the whole array than the narrow distribution. The remaining two distributions are the narrowest and broadest speckle signal distributions from across the array. These are not from the same two array pixels as the electronic noise distributions. The broad speckle intensity distribution is broader than the electronic noise indicating that there were variations in the speckle caused by factors other than electronic noise. The narrow speckle distribution is similar in width to the electronic noise indicating that the light intensity in this case was more constant. From these distributions it can be concluded that in at least some cases speckle intensity variations are a contributory factor to points in figure 6.6 not having the co-ordinates (1,1).
6.2.1.1. Summary of Empty Cuvette Experiments

To summarise the result so far, it has been demonstrated that the electronic noise is typical Gaussian noise and that the measured static speckle and noise deconvolved moments are as can be predicted from theory. That the normalised moments of figures 6.4 and 6.5 are so close to 1 can be explained with reference to figures 6.8 and 6.9. These figures show the mean electronic noise (equivalent to offset) and speckle measured in quantisation levels across the CCD array along with the standard deviation. Pixel number one is the furthest from the optical axis. The large mean values relative to the standard deviation leads to the small normalised moments. The deconvolved normalised moments of figure 6.6 give an indication of how accurate the measurements are when a sample set of 10000 measurements is used to estimate the noise statistics. Because it is known that the normalised moments should all be equal to 1 the spread in data indicates the error from all sources including the residual noise and any mechanical vibrations etc. An accuracy of 0.02 in normalised second moments and 0.05 in normalised third moments can be expected. Figures 6.10 to 6.12 show data from another similar experiment of 8000 measurements. Similar results were obtained although the electronic noise data is more closely clustered and there is more spread in the noise deconvolved moments suggesting an accuracy in normalised second moments of 0.04 and 0.12 in normalised third moments. Also visible in figure 6.10 is a single point separate from the other clustered points. This single point corresponds to the first pixel of the array which often exhibits characteristics different to those of the other pixels in the array, commonly high mean intensities.

Figure 6.8. Electronic noise statistics across array for empty cuvette experiment.
Figure 6.9. Speckle statistics across array for empty cuvette experiment.

Figure 6.10. Dark current moments for repeat of empty cuvette experiment.
Figure 6.11. Measured light intensity moments for repeat of empty cuvette experiment.

Figure 6.12. Noise deconvolved intensity moments for repeat of empty cuvette experiment.
6.2.2. Tap Water Experiments

Adding water to the cuvette increases the complexity of the experiment. The static speckle from the cuvette will change due to the different refractive index of the cuvette’s contents. Scattering from the water molecules will be too weak to detect but contamination such as dust and flakes of scale could cause some scattering above that observed with an empty cuvette and motion of the stirred water could also create some effect.

Figure 6.13 indicates that the normalised second and third moments of the background electronic noise are similar to those of the experiments already discussed in this chapter with the data closely clustered near to the co-ordinate (1,1). Data from the first array pixel is again separate from the rest. More interesting are figures 6.14 and 6.15 showing the normalised moments of the signals and noise deconvolved signals. These clearly show a set of points correlated with each other but with a different relationship to that for data from a Gaussian distribution. A least mean squares fit to the data indicates a correlation between normalised second and third moments of $\mu^2 = 4.004^{\mu^3} - 3.083$ and $\mu^3 = 4.498^{\mu^2} - 3.765$ for the noise deconvolved data. This leads to a set of points with normalised moments of value greater than those for a Gaussian distribution but lower than those for a negative exponential distribution or other exotic distributions such as Gamma or K. What type of distribution, if any specific type, these relationships correspond to is not known but the range of measured moments is unexpected. If there were only scattering from the cuvette surfaces then the scattered light intensity would be constant, leading to just the Gaussian distributed noise being measured as in the empty cuvette experiments. The fact that the data is not from a Gaussian distribution suggests that there is some source of random scattering but the reason measured moments are smaller than those of a negative exponential is not clear. The model suggesting a negative exponential was based on an infinite average number of particles, for clean water this is not expected to be realistic and finite number models could be more appropriate.

Another possible explanation for the unusual valued moments is static speckle. It has been shown that electronic offsets can affect the value of normalised moments and static speckle effectively looks like an offset at the detector. Figure 6.16 is a plot of the mean measured intensity across the CCD array showing some mean features, in particular two regions of higher mean intensity. If these mean features are due to a static speckle then they will affect the size of the normalised moments. A second experiment in which the lowest intensities at each pixel were saved shows the effect of the static speckle. Figures 6.17 to 6.19 indicate that the results are generally similar to that of the earlier experiment. Figure 6.20 shows that the mean intensity across the array is similar in form to the lowest measured intensity across the array leading to the conclusion that there is a constant static speckle
Figure 6.13. Dark current moments for clean water experiment, stirrer speed 100rpm.

Figure 6.14. Measured light intensity moments for clean water experiment, stirrer speed 100rpm.
Figure 6.15. Noise deconvolved moments for clean water experiment, stirrer speed 100rpm.

Figure 6.16. Mean speckle intensity across array for clean water experiment, stirrer speed 100rpm.
Figure 6.17. Dark current moments for clean water experiment, stirrer speed 50rpm.

Figure 6.18. Measured light intensity moments for clean water experiment, stirrer speed 50rpm.
Figure 6.19. Noise deconvolved intensity moments for clean water experiment, stirrer speed 50rpm.

Figure 6.20. Mean and minimum speckle intensity across array for clean water experiment, stirrer speed 50rpm.
present. This measured minimum is actually the sum of the smallest (bipolar) electronic noise signal and the (unipolar) speckle intensity. As such it is likely to be smaller than the average electronic offset and static speckle but it is at least a closer estimate to the overall offset than just the electronic offset on its own. Two methods of removing the effect of the static speckle were tried. The first was to treat it as a fixed constant added to the signal such that

\[
\langle i_{ph} \rangle = \langle i \rangle - i_{0s}
\]  

(6.38)

\[
\langle i_{ph}^2 \rangle = \langle (i - i_{0s})^2 \rangle = \langle i^2 \rangle - 2i_{0s}\langle i \rangle + i_{0s}^2
\]  

(6.39)

\[
\langle i_{ph}^4 \rangle = \langle (i - i_{0s})^4 \rangle = \langle i^4 \rangle - 3i_{0s}\langle i^3 \rangle + 3i_{0s}^2\langle i^2 \rangle - i_{0s}^4
\]  

(6.40)

The second method assumes that the static speckle would be modulated with the electronic noise and estimates the resultant distribution as that of the electronic noise with mean equal to that of the measured static speckle. The electronic noise can be shifted to the higher mean value using a similar analysis to (6.38 - 6.40) and then this distribution can be deconvolved from the data using (6.28 - 6.30). The difference in the results of these two methods is so small that they cannot be distinguished apart in figure 6.21. What is obvious from figure 6.21 is that the static speckle is responsible for a significant reduction in the size of normalised moments. The “speckle subtracted” moments are still below those for a negative exponential distribution but they are much closer than those of the raw data. Given that the estimate of the static speckle is probably too small and that the forecast of the best fit line in figure 6.21 passes above the point (2,6), it is tempting to speculate that if the static speckle were known accurately the data would be identifiable as negative exponential distributed.

Such speculation may not be needed to explain the normalised moments being below those of a negative exponential distribution. The linescan camera is an integrating device with a limited read rate leading to a measure that is the integral of light intensity over the period of the read cycle. If the bandwidth of the scattered light fluctuations is greater than the camera can resolve then some integration or averaging of the signal will take place. Integration of the fluctuating signal can be viewed as the summation of a number of random samples and the central limit theorem leads to the conclusion that the result will tend to a Gaussian distribution (the integration time, equivalent to number of summed samples, is constant). The concept of averaging is more applicable to truly analogue detectors where the bandwidth manifests itself as a limited slew rate (CCDs can respond to fast variations but they cannot be sampled quickly). In this case the receiver cannot track the rapid change in signal at the edges of large fluctuations and the result is that large fluctuations become smoothed into smaller broader signals. Whether viewed as integration or averaging the result is the same, the measured moments of a process are reduced to be closer to a Gaussian distribution. How close to a Gaussian the result is depends on the amount of averaging. Figures 6.15 and 6.19 show similar results but the best fit
Figure 6.21. Static speckle subtracted moment for 2nd clean water experiment.

The experimental evidence is therefore that some suspended particles in the tap water cause dynamic light scattering which can be detected. A significant amount of static speckle from the apparatus leads to the measured moments being smaller than they should be. An estimate of the static speckle can be made and a correction to the data attempted. The effect of the static speckle is compounded by a degree of integration of the scattered light by the detector. Because the effect of integration is dependent on the fluid dynamics it is not possible to make any adjustment to compensate for the effect. These factors go some way to explaining why the experimental results do not match the Gaussian statistics (negative exponential distribution) expected. It should also be remembered that with a finite number of particles Gaussian statistics are not necessarily expected and more exotic distributions could result.
6.2.3. Polystyrene Spherule Experiments

Polystyrene spheres were the simplest particles used in experiments because of their narrow size range and uniformity of shape. As such they should be the easiest to explain, but because the polyspheres were supplied as a dense suspension of unknown concentration it is difficult to make quantitative experiments. Three experiments were performed with polyspheres. The first used a drop of the suspension in a cuvette of tap water, stirred at 100 rpm. The second experiment again used a drop of the suspension in a cuvette but was stirred at 50 rpm. The third was an immediate continuation of the second under the same conditions. The concentration of polyspheres was significantly greater than any particulate content of the tap water. Figures 6.22 to 6.26 show the results from these three experiments. The first observation to be made is that the electronic noise is similar in each of the experiments to that obtained in earlier experiments. It is therefore possible to conclude that the instrumentation was working normally. Comparing results from the different experiments indicates that the second and third experiments were indeed similar but that they were slightly different to the first. All three experiments led to statistical distributions more similar to a Gaussian distribution than those from the clean water experiment. This was not expected but some explanation can be made by considering the evidence.

The mean intensity of the scattered light in these experiments (figure 6.25(a)-(c)) is higher than that of the clean tap water experiment. This is consistent with extra scattering from the polystyrene spherules. A higher mean intensity in the second and third experiments suggests that these had a greater concentration of polyspheres than the first experiment. Whereas in the clean water experiment subtraction of the static speckle caused a significant shift of normalised moments towards those expected of a negative exponential distribution, in these experiments the shift is less significant and the speckle subtracted data remains closer to that of a Gaussian distribution. Figure 6.25 indicates that the static speckle measured in these experiments are in the order of 2500 to 3000 quantisation levels compared to 300 to 500 as measured in the tap water experiment. Also there is no large scale structure as is visible in figure 6.20. This shows that the concentration of polystyrene spherules was such that there was always a significant amount of scatter so that a background speckle could not be estimated accurately. The result of this is that the background speckle estimate is too large (leading to over compensation by the deconvolution process) and not necessarily related to the true level of background speckle. Over compensation by the deconvolution process should create normalised moments greater than those of a negative exponential distribution but the results suggest that there is more integration of the scattered light signal than in previous experiments. This is to be expected in a situation where the concentration of particles is higher because a small change in the relative positions of a number of scatterers will have a similarly drastic effect on the speckle pattern as a larger change in position of a few scatterers. For particles moving at similar speeds the speckle will change more rapidly for higher
Figure 6.22. Dark current moments for polysphere experiments.
Figure 6.23. Measured light intensity moments for polysphere experiments.
Figure 6.24. Noise deconvolved intensity moments for polysphere experiments.
Figure 6.25. Mean and minimum speckle intensity across array for polysphere experiments.
Figure 6.26. Static speckle subtracted moments for polysphere experiments.
concentrations and hence the signal is more likely to have a bandwidth wider than that of the receiver leading to greater integration / averaging. This explains why the polysphere experiments produce results fitting Gaussian distributions more closely than the clean water experiments. The lower moments of the first polysphere experiment compared to the second and third experiments indicates that the faster stirring of the suspension had more effect than the increased concentration.

6.2.4. Desert Dust Experiments

The problems of measuring static speckle and particle concentration encountered with polystyrene spherules are also evident with experiments using “desert dust”. Results from experiments using desert dust are shown in figures 6.27 to 6.31. The second experiment was a repeat of the first and as such would be expected to produce the same result. The mean intensities across the array are similar in that there are peaks in mean intensity in the same places, although the relative sizes of each of these features are different. This indicates that either the scattering from these two experiments is similar spatially or that the average features are caused by the experimental apparatus. The measured static speckle in both of these experiments is of the order of 1000 which, as for the polysphere experiments, suggests that there is too much scattering to measure a reliable static speckle. Not having a reliable measure of static speckle means there is little point in trying to subtract the effect of the static speckle which makes a direct comparison with the theoretical model impossible. The results of these two experiments do show broadly similar statistical behaviour, figure 6.29 showing the noise deconvolved normalised moments from both of the first two experiments, shows how the data from the two experiments fit similar trends. The third experiment using desert dust was conducted after the second plain tap water experiment without moving anything other than the stirrer. This allows it to be assumed that the static speckle from the second water experiment is the same as that for the desert dust experiment. Comparing figure 6.30(c) with 6.30(a) and (b) the mean intensity in this third experiment is seen to be generally lower than the previous experiments suggesting a lower concentration of dust particles. Also the noise deconvolved moments are higher which fits in with the idea that with lower concentrations the integration effect is less significant. Using the static speckle measured in the second tap water experiment, the static speckle subtracted moments are as shown in figure 6.31(c). These, as expected, are closer to the co-ordinate (2,6) but the normalised second moments are still too small to fit the Gaussian scattering model. It is therefore concluded that there is some integration of the scattered light signal by the detector array leading to a reduction in the size of the normalised moments. Some of the normalised third moments are however greater than six which would indicate that the true moments, could they be measured, would fit a more exotic distribution than negative exponential, possibly K or Gamma.
Figure 6.27. Dark current moments for desert dust experiments.
(a) 1st experiment, stirrer speed 50rpm.

![Graph](image1)

(b) 2nd experiment, stirrer speed 50rpm.

![Graph](image2)

(c) 3rd experiment, stirrer speed 50rpm, follow on from tap water experiment.

![Graph](image3)

Figure 6.28. Measured light intensity moments for desert dust experiments.
(a) 1st experiment, stirrer speed 50rpm.

(b) 2nd experiment, stirrer speed 50rpm.

(c) 3rd experiment, stirrer speed 50rpm, follow on from tap water experiment.

Figure 6.29. Noise deconvolved intensity moments for desert dust experiments.
Figure 6.30. Mean and minimum intensities across array for desert dust experiments.
Figure 6.31. Static speckle subtracted moments for desert dust experiments.
6.2.5. Soluble Oil Experiments

Quantitative experiments are more easily performed using soluble oil. The volume of oil can easily be measured using microsyringes although the size of the droplets formed is not known precisely. Size analysis using a Coulter counter suggests that the bulk of the droplets are around 1 to 10 microns diameter. The results shown in figures 6.32 to 6.35 are from two experiments using 4μl of soluble oil in 4ml of tap water stirred at 50 rpm. These results show similar behaviour to the earlier experiments with typical electronic noise and no reliable measure of static background speckle. Again the moments are below those of a negative exponential distribution suggesting integration problems. The most obvious difference is the lack of structure across the array indicating an even angular spread of scattered light intensity. This is probably due to the smaller size (or more particles) of the scattering particles compared to the other experiments.

6.2.6. Correlation of Results

Each of the experiments described above has given rise to slightly different normalised moments caused by either the different scattering or the differing static speckle or both. Plotting all the results on the same graph, as in figure 6.36 for noise deconvolved data, shows that the moments from each experiment correlate fairly well (correlation coefficient = 0.9981) suggesting that the data all fit the same type of distribution. It is known that the absolute values of the normalised moments depend on the static speckle in each experiment; that a correlation exists between experiments with different static speckles indicates that the error caused by the presence of static speckle, although affecting the absolute values, does not have a significant effect on the relationship between moments. For many distributions a shift of mean does not affect the relationship between moments at all. It also follows from this that the precise curve fitting the data of figure 6.36 should depend on the amount of integration by the detector and the real unintegrated distribution. There is some evidence in the high valued third moments of the final desert dust experiment that the scattering is from a distribution similar to a K distribution but integration by the CCD array precludes making a firm conclusion. The correlation between data sets points to all the data being from a similar distribution with detector integration affecting all experiments in the same manner. This could be checked by repeating experiments with different integration times. Different types of scattering particles should still lead to statistics that correlate with each other for a particular common integration time but the experiments at different integration times should all fit different curves.
Figure 6.32. Dark current moments for soluble oil experiments.

(a) 1st experiment, stirrer speed 50rpm.

(b) 2nd experiment, stirrer speed 50rpm.
(a) 1st experiment, stirrer speed 50rpm.

![Graph showing measured light intensity moments for soluble oil experiments.](image)

(b) 2nd experiment, stirrer speed 50rpm.

![Graph showing measured light intensity moments for soluble oil experiments.](image)

Figure 6.33. Measured light intensity moments for soluble oil experiments.
Figure 6.34. Noise deconvolved moments for soluble oil experiments.
Figure 6.35. Mean and minimum intensities across array for soluble oil experiments.
Figure 6.36. Noise deconvolved moments for all desert dust, polysphere and soluble oil experiments.

6.2.7. Ballotini Experiments

Some experiments have been made at higher speeds which is equivalent to using a longer integration time. These were experiments with glass ballotini of a wide size range (figure 6.37). These particles need a higher speed of motion to keep them suspended due to their larger mass compared with the other materials used. Even so it is difficult to perform reliable experiments with this material as can be deduced from results shown in figures 6.38 to 6.47. The second set of results comes from a continuation of the first set. The lower mean intensity of the second set of results indicates that the concentration of particles left in suspension was much reduced meaning that the statistical processes being observed are non-stationary. The large reliability factor (c.f. table 6.1) of the first data set also points to a process that is changing as it is observed. Normalised moments in these two experiments are close to those of a Gaussian distribution and are also low in value. These low moments result from using a stirrer speed of 200 rpm in an attempt to keep the ballotini suspended which leads to faster speckle and a greater amount of integration or averaging. Another observation to be made is that the static speckle measured in the first experiment is higher than that in the second indicating that some of the assumed static speckle actually comes from an ambient amount of scattering. Overcompensating for the static speckle produces falsely high normalised moments which explains the marginally higher static speckle subtracted moments of the first experiment (figure 6.42) compared to the second (figure 6.47). If the static speckle measured in the second experiment is used with the first set of data, then the
Figure 6.37. Particle size distribution of ballotini.

Figure 6.38. Dark current moments for ballotini experiment, stirrer speed 200rpm.
Figure 6.39. Measured light intensity moments for 1st ballotini experiment, stirrer speed 200rpm.

Figure 6.40. Noise deconvolved intensity moments for 1st ballotini experiment, stirrer speed 200rpm.
Figure 6.41.  Mean and minimum speckle intensity across array for ballotini experiment, stirrer speed 200rpm.

Figure 6.42.  Static speckle subtracted moments for ballotini experiment, stirrer speed 200rpm.
Figure 6.43. Dark current moments for continuation of ballotini experiment, stirrer speed 200rpm.

Figure 6.44. Measured light intensity moments for continuation of ballotini experiment, stirrer speed 200rpm.
Figure 6.45. Noise deconvolved intensity moments for continuation of ballotini experiment, stirrer speed 200rpm.

Figure 6.46. Mean and minimum intensity across array for continuation of ballotini experiment, stirrer speed 200rpm.
Figure 6.47. Static speckle subtracted moments for continuation of ballotini experiment, stirrer speed 200rpm.

Figure 6.48. Noise deconvolved moments for repeat of ballotini experiment, stirrer speed 200rpm.
Figure 6.49. Noise deconvolved moments from soluble oil, desert dust, polysphere and ballotini experiments.

speckle subtracted moments of the first experiment appear smaller than in the second test. A third experiment with glass ballotini used a lower concentration and lead to larger normalised moments (figure 6.48). Fewer particles in the scattering volume mean a slower speckle and therefore less averaging. Even so the data from this experiment does not fit the same curve as the previous experiments made with a stirrer speed of 50 rpm as shown in figure 6.49.

An important clue as to what effect averaging / integration has on the signals is that the data of figure 6.49 correlates with a curve just above that for a Gamma distribution. The data of the third ballotini experiment fits the Gamma distribution better but the normalised third moments drop below the $ho^{[3]} = 2\left(\rho^{[2]}\right)^2 - \rho^{[3]}$ relationship as the moments become larger. At first sight it may appear that there is some unforeseen cause that makes the incoherent illumination model appropriate. However, it is possible to see that speckle exists, so coherence effects are present and the Gamma statistics produced by incoherent models should not be appropriate. Evidence does however suggest that the results
obtained are sensible, if not to be expected. Firstly a crude estimate of the speed of the suspended particles can be made. The stirrer has an area, $A$, of approximately

$$A = \pi d^2 = 2.4 = 8 \text{mm}^2$$

which when it rotates once moves a volume of water

$$V = A\pi\left(\frac{d}{2}\right)^2 \approx 100 \text{mm}^3.$$  \hfill (6.42)

If the stirrer is rotating at 50 rpm, then on average, in one second this volume will be moved through a distance

$$x = 2\pi \frac{d}{4} \frac{50}{60} \approx 5.2 \text{mm}.$$  \hfill (6.43)

To make a rough estimate of the particle speed $v$, this velocity can be scaled to the proportion of the total volume that the stirrer actually moves. The volume of the cuvette is 4ml and so

$$v = \frac{Vx}{4000} = 0.13 \text{mm/s}.$$  \hfill (6.44)

Using this rough estimate of particle speed, the speed of the speckle can also be estimated. A particle movement of one wavelength would cause a complete cycle of speckle from a minimum to maximum and back to minimum intensity. The HeNe wavelength is 633 nm. This distance would be travelled by a particle in approximately $633 \times 10^{-9} / 0.13 \times 10^{-3} = 4.9 \text{ms}$. Quick measurements using the pin diode detectors suggest that the speckle lasts for 5 to 10 ms which ties in well with this estimate. The minimum linescan camera integration time of 20ms was used in experiments which shows that there will be substantial averaging of the speckle taking place. As the individual speckles are averaged out the measured light intensity will tend to be dominated not by bright points of speckle but by the average amount of light scattered which depends on the number of scattering particles. The light intensity variations measured therefore depend largely on the particle number fluctuations which is what leads to the similarity to Gamma statistics. As the stirrer is speeded up, the distance moved by particles during a CCD exposure time will increase until the particles are traversing a significant proportion of the laser beam. As a result averaging of the particle number fluctuations will start to occur, the measured normalised moments then becoming smaller than those of Gamma distributed data and closer to those of Gaussian distributed data. At a stirrer speed of 200 rpm the suspended particles will travel at about $0.52 \text{mm/s}$ or a distance of $0.01 \text{mm}$ in 20 ms. This, although enough to cause integration of the speckle, will probably only lead to a slight averaging of the number fluctuations since the laser beam diameter is approximately 0.5mm.
Further evidence of the severity of integration is to be found in the amount of correlation between pixels across the CCD array. The correlation data is discussed more completely in the next section but it is worth observing here that the cross-correlation coefficient remains above 0.7 across the whole detector array in all experiments. The fact that the measured intensity is correlated across the array shows that an average is being observed and that the individual speckles are not identified. As the concentration of the suspension is increased or as the stirrer speed is increased, the correlation coefficient also rises in value. Increased correlation indicates a greater amount of integration or averaging.

6.2.8. Reliability of Moments

The final aspect of these experiments dealing with point statistics to be discussed is that of the number of samples taken. In most of the experiments the original criterion for $L > L_c$ was not met because it became impractical to run the experiments for long enough. Referring to figures 5.3 and 5.4 it can be seen that $L_c$ rapidly becomes large as the second normalised moment increases. Gamma distributions with a normalised second moment of 2 would have a $L_c$ of approximately 130000 implying that around 1.3 million samples would be needed to guarantee reliable moments. In the experiments where the third moments are greater than those of a Gamma distribution $L_c$ will be larger still. Despite the large reliability factors, $r$, the results obtained appear to be broadly repeatable. This suggests that the number of samples obtained was sufficient and that the test using $L_c$ is severe.

6.3. Discussion of Cross Correlations

The cross-correlations measured during the experiment are calculated from the light intensity minus the mean background electronic noise level. Some error is introduced by this method because it is achieved using a short term average of the electronic noise. This has to be done since the overall average cannot be calculated until the end of the experiment and storing all the individual data is impractical. The amount of error introduced should be small as the electronic noise has only a small variance compared to its mean level. The standard cross-correlation coefficient is calculated as

$$
\tau(x,y) = \frac{\langle (i(x) - i_{av}(x))(i(y) - i_{av}(y)) \rangle}{\sqrt{\langle (i(x) - i_{av}(x))^2 \rangle \langle (i(y) - i_{av}(y))^2 \rangle}} = \frac{\langle i_{ph}(x)i_{ph}(y) \rangle}{\sqrt{\langle i_{ph}(x)^2 \rangle \langle i_{ph}(y)^2 \rangle}}
$$

(6.45)
where the \( x \) and \( y \) indicate the different angles being correlated or equivalently the detector array pixel numbers.

As in the previous section, the results from each experiment are considered in turn. As the experiments were made different stirrer speeds were tried with the different particles to try and keep them suspended. Details of the different experimental conditions are as listed in table 6.1.

### 6.3.1. Empty Cuvette Experiments

The first and simplest case is with the data from an empty cuvette. Cross correlation data is shown in figure 6.50 (a) to (c). Data shown is for the cross correlation functions \( \tau(212,y), \tau(512,y) \) and \( \tau(812,y) \). The cross correlation values are all greater than 0.984 and typically around 0.998 with no structure. Cross correlation coefficients this close to one are to be expected since the speckle should be unchanging and therefore the light intensity at all detector pixels should be related by a constant amount. Coefficient deviations from the value of one must be due to electronic noise and movement of the apparatus.

### 6.3.2. Tap Water Experiments

Data from the tap water experiments are shown in figure 6.51 and 6.52, again for the functions \( \tau(212,y), \tau(512,y) \) and \( \tau(812,y) \). The difference between these two experiments is that the stirrer speed was 100 rpm in the first and 50 rpm in the second. These figures indicate an amount of decorrelation across the detector array but not complete. The amount of decorrelation is either an indication of the amount of integration by the CCD detectors or evidence that the speckle really is correlated across the array. The width of the peak in cross correlation function about the autocorrelation point gives an idea of the speckle size and also shows that detectable speckle is present. That speckle is present is proof that there was some particulate contamination of the tap water. These two experiments have a similar width of the cross correlation function peak but the baseline level of the function is higher in the first experiment than in the second. This can be explained as the baseline level depending on the amount of detector integration, a complete average would lead to cross correlation coefficients of one. The fluctuations that are not integrated out appear as peaks in the correlation function which are the width of the speckle which in turn is expected to depend on concentration. A faster stirrer speed in the first experiment would lead to the higher correlation function baseline. Some structure is also visible in the correlation function about the baseline. This is clearest in the data from the second experiment but it is
Figure 6.50. Cross correlation data for empty cuvette experiment.
Figure 6.51. Cross correlation data for clean water experiment, stirrer speed 100rpm.

Figure 6.52. Cross correlation data for clean water experiment, stirrer speed 50rpm.
present in both. An interesting point about these ripples in the correlation function is that they appear in the same place on the array independent of the point being cross correlated. These underlying modulations are probably what causes the asymmetric shapes of the correlation function peaks in the first tap water experiment.

6.3.3. Polystyrene Spherule Experiments

Polysphere experiments show similar characteristics (figures 6.53 to 6.55) except that the ripples in the cross correlation function about the baseline are not so obviously in phase. The somewhat periodic nature of the ripples suggests that they could be sidelobes to the main cross correlation function peak, indicating an amount of correlation between speckles. The low amplitude of these sidelobes would result from the integration effect of the detector. The first of the polysphere experiments was made at a stirrer speed of 100 rpm whilst the other two experiments used a speed of 50 rpm. This is reflected in the higher baseline level of the first experiment.

Figure 6.53. Cross correlation data for 1st polysphere experiment, stirrer speed 100rpm.
Figure 6.54. Cross correlation data for 2nd polysphere experiment, stirrer speed 50rpm.

Figure 6.55. Cross correlation data for 3rd polysphere experiment, continuation of 2nd, stirrer speed 50rpm.
6.3.4. Desert Dust Experiments

The experiments with desert dust, figures 6.56 to 6.58, show nothing new in the cross correlations. The modulations in the cross correlation coefficient about the baseline are again in phase with each other. The lowest baseline level occurs in the third experiment which also has the lowest mean intensity of the three experiments (c.f. figure 6.30), evidence that the lower the suspension concentration the slower the speckle changes.

6.3.5. Soluble Oil Experiments

Suspensions of soluble oil also gave similar results, figures 6.59 and 6.60.

Figure 6.56. Cross correlation data for 1st desert dust experiment, stirrer speed 50rpm.
Figure 6.57. Cross correlation data for repeat desert dust experiment, stirrer speed 50rpm.

Figure 6.58. Cross correlation data for 3rd desert dust experiment, stirrer speed 50rpm, follow on from tap water experiment.
Figure 6.59. Cross correlation data for 1st soluble oil experiment, stirrer speed 50rpm.

Figure 6.60. Cross correlation data for repeat soluble oil experiment, stirrer speed 50rpm.
6.3.6. Ballotini Experiments

Experiments using glass ballotini were made with the stirrer running at 200 rpm. This higher speed is reflected in the higher cross correlation coefficients of these experiments as shown in figures 6.61 to 6.63. The higher speed of particulate motion and speckle means that there is a greater amount of integration leading to all the pixels of the array being highly correlated. The first two experiments with the higher concentration of ballotini have a cross correlation function that does not drop below 0.97. The extent of the integration in these two cases is what leads to the normalised moments (c.f. figures 6.40 and 6.45) being close to those of a Gaussian distribution. The third experiment using ballotini at a lower concentration has a lower baseline to the cross correlation function indicating less integration of the speckle. This explains why the moments of this experiment are closer to fitting a Gamma distribution (c.f. figure 6.48). Even so, the cross correlation function is always above 0.94 indicating a still significant amount of integration leading to these moments being smaller than those of a Gamma distribution.

Figure 6.61. Cross correlation data for ballotini experiment, stirrer speed 200rpm.
Figure 6.62. Cross correlation data for continuation of ballotini experiment, stirrer speed 200rpm.

Figure 6.63. Cross correlation data for repeat of ballotini experiment, stirrer speed 200rpm.
6.3.7. Summary of Cross Correlations

In all of the experiments discussed above, there is an amount of speckle detectable as shown by the peak in the cross correlation function. This means that the individual speckle are not completely integrated out and that some of the interference effects should show through in the intensity statistics. Indeed this is the case that was identified in figure 6.36 where the normalised moments generally lie above the Gamma distribution relationship.

The modulations of the cross correlation function could possibly be due to correlation between adjacent speckle, however, if this were the case \( \tau(212,y) \), \( \tau(512,y) \) and \( \tau(812,y) \) would be expected to show similar modulations but out of phase. That the modulations are in nearly all cases clearly in phase implies that there is another cause for the modulations seen. It is unclear exactly what this is but a few unusually bright speckles sweeping across the whole array could lead to such an effect. One of the characteristics of non-gaussian scattering is that large unusually shaped speckles can be formed. Another possibility is that the static speckle is affecting the correlation measurement.

6.3.8. Normalised Intensity Cross Correlation Function

The standard cross correlation function is calculated and saved during the running of the experiments. Because other statistics of the light intensity are also saved, it is possible to recover the normalised intensity cross correlations, \( N^{[2]} \), discussed in chapter 2.

\[
N^{[2]}(x,y) = \frac{\langle f_{ph}(x) f_{ph}(y) \rangle}{\langle f_{ph}(x) \rangle \langle f_{ph}(y) \rangle} = \mathcal{T}(x,y) \frac{\sqrt{\langle (i(x) - i_{ph}(x))^2 \rangle}}{\sqrt{\langle (i(y) - i_{ph}(y))^2 \rangle}}
\]

The \( \langle i - i_{ph} \rangle \) and \( \langle (i - i_{ph})^2 \rangle \) are among the statistics stored by the linescan camera program.

Plots of \( N^{[2]}(212,y) \), \( N^{[2]}(512,y) \) and \( N^{[2]}(812,y) \) are shown in figure 6.64 for data from the experiment using an empty cuvette. From this result it can be seen that the value of \( N^{[2]} \) is always close to one. This is correct since for static speckle the moments should all equal 1 and hence so do their ratios. The spread of data indicates that measurements of \( N^{[2]} \) are accurate to within ±0.01.
Figure 6.64. Normalised intensity cross correlation data for empty cuvette experiment.
(a) $N^{13}(212, y)$, (b) $N^{13}(512, y)$, (c) $N^{13}(812, y)$. 
Normalised intensity cross correlation functions for the other experiments are given in figures 6.65 to 6.74. These all show similar characteristics in that the values of $N^1$ are a maximum near the autocorrelation point and drop to a baseline value which has some modulations on it as was the case for the standard cross correlation function. The amplitude of the peak in the normalised cross correlation function is equal to the normalised second moment:

$$N^1(x,x) = \frac{\langle i(x)i(x) \rangle}{\langle i(x) \rangle^2} = \frac{\langle i^2(x) \rangle}{\langle i(x) \rangle^2}$$ (6.47)

A formula for the normalised cross correlation function with a coherent source was not presented in chapter 2 but the similarity of the integrated signal to the incoherent source case could make comparison with equations (2.122) and (2.124) informative. These two relationships developed in chapter 2 indicate that as the mean number of scattering particles increases, the normalised intensity cross correlation decreases. This appears to be the case generally observed in the experiments. For example, the clean water experiments have a low mean intensity indicating a low number of scatterers and the baseline of the normalised cross correlation function is around 1.1 to 1.3. The desert dust experiments (figures 6.70, 6.71 and 6.72) have a higher mean intensity so more scatterers and a baseline for the normalised cross correlation of about 1.05. The polysphere experiments show a still higher mean intensity of scattered light and normalised cross correlations that drop below one.

The results seem to match the general trend of predictions of the theory presented in chapter 2. However, it must be understood that comparison with the incoherent source theory cannot be expected to be accurate due to a number of factors. Some of these factors will tend to make the cross correlations too large, others will tend to make them too small and other factors have an unknown effect. The first factor to remember is that the obtained data is not from a Gamma distribution but only close to Gamma distributed as illustrated in figure 6.49. What effect this has on the cross correlation values is not known. Another point to note is that the condition of $\theta_2 \gg \theta_1$ used in (2.123) is not necessarily true for the obtained results since the linescan camera array covers only a few degrees of arc. The result of this is expected to be that the cross correlation coefficient baseline appears to be increased since the true baseline is not actually reached. Equations (2.122) and (2.124) were derived assuming that $\alpha$ was the same at $\theta_1$ and $\theta_2$. The results shown in the previous section on point statistics indicate a spread of points meaning that there is also a spread in $\alpha$ across the array. What effect this has on the normalised intensity cross correlations is unclear. The final point of note is that the correlations measured do not take account of the static speckle from the apparatus. This can be compensated for using the following method where $i_s$ is the static speckle intensity.

$$N^2 = \frac{\langle (i_{ph}(x) - i_{st}(x) + i_s(x))(i_{ph}(y) - i_{st}(y) + i_s(y)) \rangle}{\langle (i_{ph}(x) - i_{st}(x) + i_s(x))(i_{ph}(y) - i_{st}(y) + i_s(y)) \rangle}$$ (6.48)
Figure 6.65. Normalised intensity cross correlation data for clean water experiment, stirrer speed 100rpm.

Figure 6.66. Normalised intensity cross correlation data for clean water experiment, stirrer speed 50rpm.
Figure 6.67. Normalised intensity cross correlation data for 1st polysphere experiment, stirrer speed 100rpm.

Figure 6.68. Normalised intensity cross correlation data for 2nd polysphere experiment, stirrer speed 50rpm.
Figure 6.69. Normalised intensity cross correlation data for 3rd polysphere experiment, stirrer speed 50rpm, continuation of 2nd experiment.

Figure 6.70. Normalised intensity cross correlation data for 1st desert dust experiment, stirrer speed 50rpm.
Figure 6.71. Normalised intensity cross correlation data for repeat of desert dust experiment, stirrer speed 50rpm.

Figure 6.72. Normalised intensity cross correlation data for 3rd desert dust experiment, stirrer speed 50rpm, follow on from tap water experiment.
Figure 6.73. Normalised intensity cross correlation data for 1st soluble oil experiment, stirrer speed 50rpm.

Figure 6.74. Normalised intensity cross correlation data for repeat of soluble oil experiment, stirrer speed 50rpm.
\[ N^{[2]} = \frac{\langle (i_{ph}(x) + S(x))(i_{ph}(y) + S(y)) \rangle}{\langle i_{ph}(x) + S(x) \rangle \langle i_{ph}(y) + S(y) \rangle} \]  
(6.49)

\[ = \frac{\langle i_{ph}(x)i_{ph}(y) + S(y)i_{ph}(x) + S(x)i_{ph}(y) + S(x)S(y) \rangle}{\langle (i_{ph}(x) + S(x))(i_{ph}(y) + S(y)) \rangle} \]  
(6.50)

\[ = \frac{\langle i_{ph}(x)i_{ph}(y) \rangle + \langle S(y)i_{ph}(x) \rangle + \langle S(x)i_{ph}(y) \rangle + \langle S(x)S(y) \rangle}{\langle i_{ph}(x) \rangle \langle i_{ph}(y) \rangle + \langle S(y)i_{ph}(x) \rangle + \langle S(x)i_{ph}(y) \rangle + \langle S(x)S(y) \rangle} \]  
(6.51)

\[ \frac{\langle i_{ph}(x)i_{ph}(y) \rangle}{\langle i_{ph}(x) \rangle \langle i_{ph}(y) \rangle} = \frac{N^{[2]}[\langle i_{ph}(x) \rangle \langle i_{ph}(y) \rangle + \langle S(y)i_{ph}(x) \rangle + \langle S(x)i_{ph}(y) \rangle + \langle S(x)S(y) \rangle]}{\langle i_{ph}(x) \rangle \langle i_{ph}(y) \rangle} \]  
(6.52)

This can be calculated since \( i_{os} \) and \( i_s \) are recorded during the experiments and \( \langle i_{ph} \rangle = \langle i_{ph} - i_{os} \rangle - S \).

It can be deduced from (6.52) that compensating for the static speckle will make the normalised intensity cross correlations larger when the uncorrected \( N^{[2]} \) is greater than one and smaller when the uncorrected \( N^{[2]} \) is less than one. Only three experiments have a reliable static speckle measurement (the second water experiment, the third desert dust experiment, and the third ballotini experiment) and the normalised intensity cross correlations corrected for static speckle are shown in figures 6.75 to 6.77 for these experiments. Using these corrected cross correlations it should be possible to use (2.122) and (2.123) to make a rough estimate of the mean number of scatterers. Using the moments calculated for each of these experiments with the static speckle subtracted (figures 6.21, 6.31 and 6.48) and assuming a Gamma distribution of data (moments described by (2.103)), upper and lower estimates for \( \alpha \) can be made. Figures 6.78 to 6.80 show upper and lower estimates for \( \bar{N} \) obtained using \( N^{[2]}(15, y) \) to make \( x-y \) (or equivalently \( \theta_{1}-\theta_{2} \)) as large as possible. Around the autocorrelation point there are discontinuities but it is the other end of the graph where \( x-y \) becomes large that we are interested in. It appears that the mean number of particles in these three experiments were negative, only the ballotini experiment leads to a possible positive estimate for \( \bar{N} \). These estimates of \( \bar{N} \) are obviously in error since negative numbers of particles cannot exist. Accurate estimates cannot be expected for the reasons noted above but there is also the added possibility that the integration effect of the detector changes the value of \( \alpha \) observed. The detector effectively measures the process of births and deaths of the speckle which is not necessarily the same as the birth-death process of the particle number fluctuations. As a result it is possible that a Poisson process of particle number fluctuations (\( \alpha=\infty \)) would lead to speckle fluctuations with a finite value of \( \alpha \). Using too small a value of \( \alpha \) will lead to estimates of \( \bar{N} \) which are too small or even negative.
Figure 6.75. Static speckle corrected, normalised intensity cross correlation data for 2nd clean water experiment, stirrer speed 50rpm.

Figure 6.76. Static speckle corrected, normalised intensity cross correlation data for 3rd desert dust experiment, stirrer speed 50rpm.
Figure 6.77. Static speckle corrected, normalised intensity cross correlation data for 3rd ballotini experiment, stirrer speed 200rpm.

Figure 6.78. Upper and lower estimates of $\bar{N}$ for 2nd clean water experiment, stirrer speed 50rpm.
Figure 6.79. Upper and lower estimates of $\bar{N}$ for 3rd desert dust experiment, stirrer speed 50rpm.

Figure 6.80. Upper and lower estimates of $\bar{N}$ for 3rd ballotini experiment, stirrer speed 200rpm.
With all of these uncertainties it is impossible to make a sensible estimate of $\bar{N}$ but the normalised intensity cross correlations do at least follow a sensible trend in that as $\theta_1-\theta_2$ becomes large, they tend to a baseline value which appears to depend on the concentration of suspended particles.

6.4. Summary of Statistical Results

A noise model for photodiode detectors has been developed which predicts that the receiver noise is dominated by Gaussian thermal noise. Experimental data shows that the electronic noise is indeed distributed as from a Gaussian pdf. With the knowledge that the receiver noise is Gaussian it is possible to compensate for the effect the noise has on the statistics of a detected signal using a deconvolution process. The deconvolution process requires the receiver dark currents to be monitored.

Statistical analysis of point measurements of light intensity scattered from suspensions indicates that the intensity fluctuations are not from a Gaussian distribution. Neither are they from a K distribution as predicted by theory. The moments are too small to be from a negative exponential distribution (so called Gaussian scattering) or a K distribution. The magnitude of the normalised moments decreases with increasing concentration of the suspension and increasing speed of the suspension motion. Both of these will lead to a more rapidly changing speckle and therefore it is believed that the normalised moments are being reduced due to time averaging of the speckle. It would appear to be coincidence that the signal averaging leads to normalised moments approximately those of Gamma distributions.

Background scatter from the apparatus and electronic offsets can also reduce the size of the normalised moments. The electronic offsets can be removed as part of the noise deconvolution process as can the background scatter or "static speckle" if it can be measured.

Cross correlation data confirms a link between the speed of motion of the suspended particles and the amount of speckle time averaging. Normalised intensity cross correlations, $N^{\text{(2)}}(x,y)$, show the general trends predicted by theory for an incoherent source of being a maximum at the self correlation point and tending to a level dependent on the number of scatterers as $x$ and $y$ separate. Had the experiments actually been conducted with incoherent light it may have been possible to estimate the average number of scatterers in the beam of light. Since the experiments were made with coherent light any inference drawn from the similarity between the experimental results and theory for incoherent source must be conjectural.
7. Conclusions

A number of conclusions can be made from the observations made during the course of this study. In this chapter conclusions relating to nephelometry and statistical analysis are presented in turn.

7.1. Nephelometry

The method of using nephelometry with neural network processing has been shown to enable estimates of volume fraction of suspensions to be made.

An advantage of nephelometry over turbidity measurements for estimating particulate concentration is its ability to identify different suspensions. The particulate size of the suspended media appears to have more effect on the scattering than the optical properties and therefore the nephelometer discriminates suspensions predominately by particle size. A nephelometer with neural network processing can therefore be used to build a cheap and simple particle sizer.

Combining the redundancy provided by using a number of sensors at different scattering angles with neural network processing makes a system with some immunity to sensor fouling.

The simplicity of the basic technique makes it a comparatively cheap technology. Combined with its robustness to sensor fouling this makes it a suitable technology for use in-situ or on-line in a process.

In summary, the work on nephelometry with neural network processing has furthered development of optical instrumentation for suspensions by demonstrating an instrument that:

- is not prohibitively expensive
- estimates volume fraction
• discriminates suspensions on the basis of size
• has reduced sensitivity to sensor fouling
• is suitable for in-situ use

7.2. Statistical Analysis

Light scattering from a suspension of particles has been shown to form random speckle in the far field. The speckle is dynamic as the suspended particles move. Point measurements of the speckle have shown the intensity to be from a probability distribution other than Gaussian. To the author’s knowledge this is the first time such statistics have been observed without photon counting detectors and the first time for a range of suspended materials. The actual distribution depends on the suspension and its fluid dynamics. This means it may be possible to use point intensity measurements to infer information about the observed suspension.

Normalised intensity cross correlations as defined in (2.122) give an indication of the mean number of particles in the observation volume. As the angular distance between cross correlated points increases, the normalised intensity cross correlation function tends to a baseline value that is dependent on the concentration of suspended particles.

The statistical analyses investigated are effected by noise and offsets of the detection system. The photodiode receiver systems used have been shown to exhibit Johnson thermal noise as predicted from noise models. Amplifier offset can be treated as a shift in mean of the noise and so electronic noise and offset can be considered as a single statistical signal source convolved with the scattering signal of interest. Deconvolution of this noise statistic is possible by measuring the dark current statistics during the experiments.
8. Discussion of Conclusions and Further Work

8.1. Nephelometry

Using nephelometry and neural networks has been shown to enable estimates of particle size to be made. The accuracy of the estimate depends on the signal to noise ratio of the sensor. The minimum working range of such a system is also to some extent dependent on the signal to noise ratio since once the signal is smaller than the noise no further resolution of concentration is possible. Once the noise floor of the detector has been reached the only way of improving the sensitivity of the sensor is to increase the input power to boost the signal, thus the limit of operation and accuracy depends on both the detector performance and source power. Even with unlimited source power there will come a stage where the concentration is so low that the sensor is detecting the presence or absence of single particles. In this circumstance experimentation becomes difficult since the signal appears to be noisy due to the discrete nature of the scattering. In this scenario the average of a very large number of samples would be needed or the neural network would need to be trained to recognise the particle counting regime and to estimate particle size.

Of the two nephelometers tested in this work the first was more efficient at collecting scattered light and so proved to give the best estimate of oil concentration over the widest range. A mean error of approximately 5% over the range 0 to 100 \( \mu l/l \) was achieved with this sensor. Most of this error is due to poor signal to noise ratios at low concentrations. A lowest detection limit of about 0.3\( \mu l/l \) was achieved with this nephelometer by using the HeNe laser source launched through a fibre. It should be possible to improve upon this by using a more efficient launch mechanism. This sort of performance may be adequate for some applications but it should be possible to improve on this fairly simply by increasing the input power.

The main advantage of nephelometry over turbidity measurements for estimating particulate concentration is its ability to account for different suspensions. The particulate size of the suspended media appears to have more effect on the scattering than the optical properties and therefore the nephelometer acts as a cheap and simple particle sizer. Identification of suspension size enables the nephelometer and neural network to make a more accurate estimate of concentration. There is therefore a double advantage in being able to distinguish suspensions; not only is a knowledge of the suspension size obtained but an improved concentration estimate is made. As for estimating concentration, a
greater signal to noise ratio makes the task of the neural network identifying different suspensions easier.

In applications such as oil pollution monitoring it would probably be necessary to combine nephelometry with another technique such as fluorescence [8.1],[8.2] to gain information on the chemical nature of the suspension, the size being too dependent on (uncontrolled) environmental conditions to be relied on as a method of distinguishing oils. Very different sized suspensions, such as soluble oil and crude oil, are easily distinguished. However, as the suspensions become more similar it becomes harder to distinguish between them. As noted a good signal to noise ratio helps but the optical arrangement is also important. Data provided by the second nephelometer studied suggests that more angular specific receivers would improve the ability of the sensor to sense changes in suspension size distribution.

Further work that could be done in part addresses the practicalities of the current nephelometer designs and would partly be aimed at improving the theoretical design. Practical considerations that are needed to improve the current designs are:

- To maximise the power launched into the sensor.
- Control the volume observed by the receivers.
- Optimise the angular positioning of the receivers.

Direct launch of the source into the test cell is the method that minimises beam attenuation by the optics but it is important to find a method of eradicating condensation on the window. A matching gel placed between the source and window may achieve this or a sealed unit, such as encapsulation in plastic, may be preferable. The combination of size, cost, power and ease of modulation means that the laser diode is probably the best source available. Being a solid state device it may be possible to encapsulate it like an LED without affecting its performance.

A method of controlling the numerical aperture of the receivers but without the attenuation of the probes used in the mkII nephelometer is required. Using fewer lenses is an obvious development to reduce the power lost in reflections, a single aspheric lens probably being the best solution.

To optimise the angular positioning of the receivers, it would be instructive to simulate the scattering expected from a range of suspended particle distributions. This should allow prediction of which
combination of sensing angles will lead to the best performance of the neural network in identifying the different distributions. This should be done with some constraints due to the physical size of the receivers. Some work on the simulation of scattering and network performance has been started within the OEG at Loughborough [8.3] to investigate the number of sensors needed and the best angles at which to place them. This work should be continued and expanded to look at optimising the relationship between the power received by the detectors and the numerical aperture of the receivers. A lower numerical aperture will give better angular resolution but also less light will be collected resulting in a poorer signal to noise ratio. A larger lens could be used to collect more light but this would use up more space limiting the number of sensing angles.

Following the design and manufacture of an optimised nephelometer it will be necessary to test its performance practically. For this it is suggested that experiments be conducted using solids of calibrated distributions. This will be more expensive than the oils but will be a better test of how well the nephelometer distinguishes particle size.

8.2. Statistical Analysis

8.2.1. Practical Considerations

The experiments performed during this research have highlighted some practical considerations in the use of dynamic speckle statistics. The major considerations are;

- Number of samples required and the time taken to obtain them.
- Time integration of the speckle by the detector.
- Effects of scattering from the apparatus.

A problem of using point statistics on their own is the time taken to obtain a reliable statistic. Several thousand uncorrelated samples are required, and even though electronic detectors could sample the scattered intensity at high rates, the need for the data to be uncorrelated means waiting for the fluid dynamics to produce a representative range of speckles. The process of collecting sufficient data quickly can be aided by using a detector array. This could sample an area of the far field larger than individual speckles, but small enough such that the speckle is statistically alike over the whole array. This could speed up the data collection process by a factor equal to the number of pixels in the array.
The observed dynamic speckle is integrated by the linescan camera, reducing the amplitude of the normalised moments. Despite this the point statistics still indicate a non-Gaussian speckle intensity by the third normalised moments being greater than the $I^{[3]} = 3I^{[2]} - 2$ relationship for a Gaussian distribution. The normalised moments acquired from across the array correlate to a line although this is not the $I^{[3]} = \left( I^{[3]} - I^{[2]} \right)$ expected of a K distribution that would arise from a Gaussian scattering process. An indication that the statistics at each pixel are from the same type of distribution is that the statistics measured across the array correlate. Evidence of the integration effect is that the statistics lie between the limits for K and Gaussian distributions. It would appear to be coincidence that the amount of integration is such that the statistics are similar to those of a Gamma distribution.

The cross-correlations between pixels across the array provide further evidence of integration by the fact that the cross-correlation coefficient remains high across the whole array, even though the speckle is only a couple of hundred pixels across. The baseline value of the cross-correlation coefficient is higher at increased stirrer speeds indicating that the integration becomes more serious and confirming the relationship between the speed of particles and the rate of speckle changing. The normalised cross-correlations fit the general trend predicted by the theory presented in chapter 2 in that as the average number of scatterers increases, the normalised cross-correlation coefficient of widely separated points becomes smaller. However, it must be remembered that the theory was based on an incoherent source model. As such, the theory is not directly applicable to the Gamma-like intensity distribution observed arising from coherent scattering. Apart from the model not being based on a coherent source, there were other problems that have prevented experimental data being used to estimate the number of scatterers with this method. The first practicable problem is that of static speckle affecting the mean intensity and thus the normalisation of the cross-correlation. The second is that the integration of the speckle will tend to produce a more constant signal and increase the normalised cross-correlation coefficient. Also the theory assumes the same value of the bunching factor, $\alpha$, at both correlation points. The point statistics indicate that there is a spread in the value of $\alpha$ measured at different angles and so the theory is not directly applicable on a second theoretical count.

Superposed on the dynamic speckle is a background "static" speckle from the apparatus that can be viewed as a type of offset. If this static speckle can be measured accurately then it can be subtracted or deconvolved from the dynamic speckle pattern as the electronic noise and offset can. The problem is complicated somewhat by the fact that the scattering from the suspension could interfere destructively with the static speckle to produce what would look like a negative intensity speckle. There are practicable changes that could help to compensate for or remove the effects of static speckle. Measuring the static speckle from the cuvette filled with water, before adding the suspended particles,
allows the effect of the static speckle to be calculated and subtracted from the measurements. The results discussed in chapter 6 show this to be of some help in improving the absolute accuracy of the results obtained. Another method of removing static speckle is to stop it forming in the first place. A high optical quality test cell would help this but would be expensive to implement in a commercial instrument. As a research tool it is probably worth the expense to obtain unambiguous data. In addition the theory could be developed to incorporate the scattering from the test cell surfaces.

8.2.2 Development of Theory

Further understanding of the practical problems could be gained by developing the theory. Obvious areas for further theoretical work are;

• Scattering from apparatus.
• Improving the receiver noise model.
• Investigating signal integration by the detectors.

Each optical surface of the test cell could be considered as random phase screen which, unlike the suspension, is unvarying. The scattering model would therefore consist of a random walk of $2M + N$ steps where $N$ is a random variable describing the number of scattering particles in the test volume and $M$ is the (constant) number of effective scattering centres on each test cell surface.

The receiver noise model would be made more accurate by including a noise figure for the amplifiers. However, since it is possible to measure the actual statistics of receiver noise during an experiment and compensate, there is little practical reason for extending the theoretical model.

Investigating the theoretical effects of signal integration could prove a useful study. Although using different hardware could remove the integration effect there may be some practicable use of integration. The difference between the moments from non-integrated and integrated data should be different because integration reduces the normalised moments; the amount of difference depending on the integration time compared to the rate of change of speckle. A knowledge of how the normalised moments change with integration time should therefore give an indication of how fast the speckle is changing and hence how fast the suspended particles are moving. It may also prove possible to use integration as a check on the statistical accuracy; if a signal that would lead to K statistics becomes integrated to a signal leading to Gamma statistics, it may give a second measure of the bunching factor.
More than one approach to extending the model to integrated data could be attempted. The theory presented in chapter 2 is based upon the random walk model described by equation (2.8) and rewritten here as (8.1).

\[ E = \sum_{j=1}^{N} A_j e^{\Theta_j} \]  

Detector integration is like the summation of consecutive samples and so the summation of \( T \) consecutive random walks as in equation (8.2) should in some way simulate this effect.

\[ \sum_{j=1}^{T} \left( \sum_{j=1}^{N} A_j e^{\Theta_j} \right) = \int_{0}^{T} \sum_{j=1}^{N} A_j e^{\Theta_j} dt \]  

The difficulty with this method is that the random variables in the summation over \( N \) are not correlated whereas the variable integrated over time should be correlated because the detector is integrating a continuous analogue signal. By ignoring this correlation the case of a long integration period is simulated because all intensities are treated as independent of that measured immediately prior, equivalent to integrating over different speckles. Including a correlation would involve using conditional intensity probabilities in the summation over the integration time, \( T \).

Another way of looking at the problem is to consider why the speckle changes in a continuous manner. The number of scatterers, \( N \), must be discrete and therefore the continuous nature of the summation (8.1) must be derived from the \( A_j \) and \( \Theta_j \) varying in a continuous fashion such that (8.1) can be rewritten as

\[ E(t) = \sum_{j=1}^{N} A_j(t) e^{\Theta_j(t)} \].  

How the \( A_j \) and \( \Theta_j \) vary is unknown but a sensible model would be to assume that the \( \Theta_j \) is proportional to time as might be expected for particles moving at constant speed relative to the detector. The \( A_j \) would be expected to remain fairly constant for a particle that remains in the centre of the illuminating beam of light and only if the integration time was sufficient for the particle to move out of the beam would the \( A_j \) change. The simplest model is therefore to assume that \( A_j \) is constant for \( 0 < t < T_b \) and zero at other times where \( T_b \) is the time taken for the particle to traverse the illuminating beam. More complex models could include the effect of beam profile. Using these simple models the integrated random walk can be written;

\[ \bar{E} = \sum_{j=1}^{N} \left( \int_{0}^{T} A_j e^{\Theta_j(t)} dt \right) \]  

219
This formula could then be used in (2.48) and the intensity pdf calculated using a similar method as in chapter 2. This model could be developed further by letting the integration constant \( T \) be a random variable as well. This would then represent the \( j \) different particles moving in different directions or at different speeds. A series of experiments with different detector integration times should help to identify which model is most appropriate.

In this thesis the statistical analysis has been approached with a view to applications in analysing suspensions of small particles. Other applications should be investigated such as uses in millimetre radar for identifying objects (a single large object could give rise to speckle, like a rough surface, that changes as the object moves). Another modification could be to use multiple sources and receivers, a type of multistatic radar, to detect objects designed to have a small radar echo. The multiple sources and receivers would give more chance of detecting a return and interference effects from the multiple sources could make detection easier. Other potential military applications exist such as identifying the difference between radar returns from a target and chaff used as a countermeasure to mask targets.

### 8.3. General Comment

Statistical analysis of experimental scattering data appears to indicate that the random walk is an appropriate model, and that with improvements to the experiments some knowledge of fluid dynamics and numbers of particles could be obtained. Since the point statistics come "free" with any sampling detector it is sensible to make use of them.

Further work on both techniques is required but whereas the statistical analysis of point data and correlations remains an area for research, nephelometry is at a stage where development towards a particular application could be undertaken. Applications with which the combination of neural networks and nephelometry may be used successfully are water quality testing, oil slick dispersion monitoring, and checking filter effectiveness and ageing in filtration processes.
9. References


[4.9] Green D.A., Naimimohass R., Barnett D.M., Smith P.R., An optimised nephelometer and
non-linear processor for oil-in-water monitoring, *Proc. EUROPTO Series, Air Toxics and

[5.1] *Instructions for InstaSpec II Photodiode Array Detector Model 77112*, Oriel Corporation,
Stratford, CT, 11 Nov. 1993.


[8.1] Hurford N., Buchanan I., Law R.J., Hudson P.M., Comparison of Two Fluorometers for

[8.2] Camagni P., Colombo G., Koechler C., Pedrini A., Omenetto N., Rossi G., Diagnostics of Oil
Pollution by Laser-Induced Fluorescence, *IEEE Trans. Geoscience & Remote Sens.*, Vol GE-

1300.


[A.2] *NB-MIO-16X user manual*, National Instruments, Austin, TX, part No 320157-01, May
1989.
Appendix A

Electrical Hardware

Source Drivers

Each of the source types used needs its own type of drive electronics. The HeNe laser has its own commercial mains power supply and since it is a CW laser it cannot be modulated electrically. The power supply for the HeNe laser is therefore not described further. The current sources for the LEDs and laser diodes were constructed specifically for use in the research work and these are described.

LED Driver

The driver initially used for LEDs consisted of a timer circuit switching a transistor, figure A.1. DC power was obtained from a bench top supply. The timing circuit was based on a dual monostable (74 LS 221), one monostable triggering the other and visa versa. A push button reset was included in the design to make sure the repeating toggle action could be started from the correct initial conditions. In practice the toggle action starts correctly on power up anyway. The lengths of the output pulses from each monostable are controlled by an RC circuit external to the chip. Three different capacitors may be switched into this RC circuit to give three different ranges of operation and a variable resistor allows fine adjustment of the timing within these ranges. An output from one of the monostables is buffered before being used to drive the base of a transistor. This transistor is used as a switch to pulse the LED. Alternatively an external timing source can be used to drive this switch. The LED current is controlled by a variable resistor which limits the voltage drop across the LED. A one ohm resistor in series with the LED is used to monitor the current with an oscilloscope. This driver circuit was used in a number of experiments although it has now been superseded by the laser diode driver which can be used to drive LEDs as well as lasers.
Figure A.1. LED driver circuit.
Laser Diode Driver

The laser diode driver is based on a commercial PCB made by Oxford Optronix. The board performs several functions. Four outputs are provided to drive diode lasers, two of which use pin diode detectors built into the laser package to provide feedback for constant power operation. These constant power drives are intended for CW operation. The other two drives work in constant current mode and may be modulated between two power levels set by potentiometers. Rapid switching of a laser through its threshold current can shorten its life so it is standard practice to modulate between the threshold current and the full drive current rather than from no current to full current. The modulated constant current drive works by starting with a constant current source which supplies the laser. A modulation signal then controls the proportion of that constant current that is sunk down a load parallel to the laser, using this method keeps a constant current load on the power supply. Drive currents can be monitored on an LCD included on the Oxford Optronics LDTC4000. On power up there is a “slow start” to the laser drive currents. This reduces the chance of damaging the lasers with an abrupt switch on from below the lasing threshold.

The laser driver board also has a facility for temperature stabilisation. A 100kΩ thermistor is used as the temperature monitor for the laser mount. An output is provided that is suitable for driving a thermoelectric cooler (TEC). The desired temperature and time constant for the speed of response are set by potentiometers. A second thermistor (10kΩ) can be used to monitor temperature, the output from this measurement is available to the user as a near linear response voltage output.

Electronic interlocks are incorporated into all four laser drives. These work simply by switching off the drive current to the lasers when the interlock signal drops to a logic low. Each laser drive has its own individual interlock.

Full details of the LDTC4000’s features and performance can be found in the user guide [A.1]. Certain modifications to the board detailed below will affect the absolute performance figures given in the user guide to some extent but the changes should be minimal.

The finished laser driver instrument makes all the features of the Oxford Optronics board available to the user from either the front or rear panels of the 1U, 19inch racking case. To do this the potentiometers for controlling the different parameters had to be changed for ones that were suitable for panel mounting. Where possible the same value components were used but in some cases these could
not be obtained so the closest alternative was used. All of these variable resistors are of the same or higher power ratings than the originals they replace. The on-board dual in line LCD selector switch has been replaced with a panel mounting, break before make rotary switch.

Additional circuitry made to supplement the LDTC4000 is shown in figure A.2 and a copy of the PCB track layout in figure A.3. This board provides regulated power supplies at the correct voltage for the laser driver board, electrical isolation of the modulation and interlock inputs plus a means of overriding the interlocks and modulation signals.

The laser driver was designed to be used as part of a 19 inch rack system and so it was assumed that external DC power supplies would always be available (see later). External mains supplies are preferred because it keeps the power supplies away from the more sensitive electronics. The external power supply levels are ±15 Volts, +5 Volts and 0 Volts. The 5 Volt supply is fed straight through to the LDTC4000 as is the ground line. The 15 Volt supplies need to be regulated down to ±12 Volts and this is done using four voltage regulators. 7912 (1 Amp rated) fixed voltage regulators are used for the negative supply and one 78S12 (2 Amp) and one 7812 (1 Amp) are used for the positive supply.

Although the thermoelectric cooler circuit can draw up to 2 Amps on either positive or negative supply, the 19 inch rack power negative supplies are rated lower and so the use of a 1 Amp regulator should not cause problems other than the TEC performance being reduced.

Interlock inputs and modulation inputs are electrically isolated from the laser driver board using 6N137 opto-isolators. This provides overvoltage protection and allows floating earth signals to be used as inputs to the laser driver. Noise immunity is improved because earthing problems between different pieces of equipment are avoided. To make replacement of the opto-isolators easy, should one be destroyed by an overvoltage condition, they have been mounted in DIL sockets. The opto-isolators have a digital output so that they may only switch the lasers on or off, analogue modulation is not possible.

A TTL line driver (74LS241) buffers the opto-isolated modulation signal for connection to the driver board. To allow the constant current drives to be used for CW laser operation without needing an external signal, a toggle switch can connect a logic low to the TTL line driver instead of the modulation signal.
Figure A.2. Laser driver interface electronics.
Figure A.3a. Laser driver interface PCB. Component side.
Figure A.3b. Laser driver interface PCB. Solder side.
Another toggle switch can be used to override the interlocks. This allows lasers to be operated without any interlock, safety procedures are then left to other means. A green LED is illuminated on the front panel when the interlocks are over-ridden. The interlock over-ride works by either enabling or disabling multiplexers (74HCT139) which are otherwise controlled by the interlock inputs, a logic high being needed to enable the lasers. Initially the design included an analogue switch which was to be controlled by the multiplexer outputs. It was later found that the impedance of the analogue switch caused problems and that due to the design of the LDTC4000 board a logic signal to the interlock jumper worked just as well as a physical short. Because of this the analogue switch was removed from the design and the interlock circuitry is driven directly from the multiplexers.

The front and rear panels of the laser driver are shown in figure A.4. On the rear panel are 4mm sockets for the power inputs of -15, 0, +5 and +15 Volts from left to right. The instrument case is not connected to the 0 Volt input and because of this all connections to the unit are insulated. Information about the whole system's earthing is given later. Next to the power inputs are the BNC interlock input connectors in the order constant current and constant power channel A and then constant current and constant power channel B. Right of these is the switch for over-riding the interlocks followed by the modulation input for channel B then channel A. The connector for the temperature monitoring thermistor is next in line. A 3 pin Fischer series 102 socket is used, the top and left pins being the two connected. On the extreme right of the rear panel is a BNC for the output of the temperature monitoring circuit.

Leftmost on the front panel are the two constant current drive outputs, the connectors are 3 pin Fischer series 102. The top socket is for channel A and the lower for channel B. The top pin in each connector should link to the laser anode and the left pin to the cathode (0 Volts). Four knobs immediately to the right of the connectors control the drive and bias currents, the larger knobs the drive current and the smaller ones the bias. The pair to the left control channel A and the pair to the right channel B. When starting to use a modulated diode laser the bias control should be turned fully clockwise (maximum bias) and the drive current control should be turned fully anticlockwise (minimum current). The LCD should be switched to show the appropriate channel. With the laser switched on the drive current should be increased by turning the control clockwise until the desired operating current is obtained as shown on the LCD. The laser should then be turned off by taking the modulation input low and the bias current adjusted using the control knob until the LCD displays the desired bias current, possibly the laser threshold current. The bias control has the dynamic range to take the bias to zero if desired but this can be expected to shorten the laser's life. During modulated operation the LCD will display the mean current unless the modulation rate is slower than the ammeter's response time.
Figure A.4a. Laser driver front panel.
Figure A.4b. Laser driver rear panel.
To the right of the constant current drive controls are the two constant power drive outputs, again using 3 pin Fischer series 102 connectors. These outputs are configured for common cathode operation with the top pin being connected to the laser anode, the left pin common (0 Volts) and the bottom pin connecting to the diode anode. The top output is channel A and the bottom one channel B. To the right of the connectors are the power controls, channel A leftmost, which should be wound fully anticlockwise at power on and then rotated clockwise to increase the power to the lasers’ operating level.

The LCD displays the drive current in mA of one channel at a time, the channel being selected by the rotary switch right of the display. The switch positions are labelled cca, ccb, cpa and cpb to indicate that the channel selected is constant current (cc) or constant power (cp) and channel A or B. To the top and right of the LCD is a green LED. This is lit as a warning when the interlocks are over-ridden.

A toggle switch on the front panel selects whether the constant current drives are being modulated by an external signal (connected on the rear panel) or that they are being used in CW mode with no external connection required.

The remaining two controls and socket on the front panel are for the TEC. One knob sets the temperature and the other sets the time constant, labelled TC, of the TEC response to changes in temperature. The 4 pin Fischer series 102 socket connects to both the temperature monitoring thermistor and the TEC power. The top two pins should connect to the thermistor. The bottom left pin is for the peltier device positive terminal and the bottom right pin for the negative terminal.

**Receiver**

The detector and receiver unit was designed to interface simply with the rest of the system, in particular the data acquisition board that was to be used, a National Instruments NB-MIO-16X. Full details of this board are found in the manual [A.2]. To make the overall system as flexible as possible for future use the receiver has sixteen channels, despite the fact that the nephelometers did not need this number. There are some other design features that arise because of the specific data acquisition board that was to be used, these will be discussed as they are met. The unit is housed in a 2U standard 19inch rack casing which fits into the same rack as the laser driver. Regulated external DC supplies of +15 Volts, -15 Volts and 0 Volts are assumed to be available from the rack’s power supply.
It will be easiest to describe the main features of the receiver unit first and then to describe the more detailed design. Detection is by reversed bias pin diodes (photoconductive mode) giving a linear response with intensity and maximum responsivity at 850nm. Each channel has a variable gain for the preamplifier, set individually using a selector switch within the unit. Selectable gains are 1, 10, 100, 200, 300, 500, 600, 700 and 800. Dynamic range on the output is from -12.5 Volts to +12.5 Volts.

Output connections can be made using a ribbon cable, BNC leads or both. Both types of connector are accessed from the rear panel. The ribbon cable enables easy interfacing to the NB-MIO-16X board in either 16 channel single ended mode or 8 channel differential mode. The ribbon cable is also suitable for connection to other National Instruments data acquisition boards although the order of the wires may need changing at the far connector. The BNC connectors provide single ended signals only.

Optical input connections are by optical fibre with SMA connectors. Coupling can be made on the front panel, or alternatively the fibres may be taken inside the unit via a protective conduit to internally housed diodes.

The only connectors on the front panel are SMA bulkheads for optical coupling. These are arranged in two rows of eight, the top row being channels one to eight from left to right and the bottom row channels nine to sixteen from left to right.

Back panel connections are the ribbon cable for signal output, the BNC signal output sockets, power input and the conduit entry point. The positions of these connections are shown in figure A.5. The ribbon cable is arranged so that pin 1 is to the right as seen from outside the unit, facing the rear panel. The order of the pin connections matches those of the NB-MIO-16X directly. Pins used by the data acquisition board but not by the receiver unit are unconnected.

The BNC sockets are arranged in a single row such that their positions are, as close as possible, directly behind their respective front panel inputs. This means that the channels are interleaved in the order 1,9,2,10,3...8,16 from right to left. The BNC sockets are not insulated from the casing and so the coaxial cable shield is not necessarily at the signals' ground potential.
Figure A.5. Front and rear panel of receiver unit.
Power supplies are connected to the unit via 4mm plugs on the back panel. There are smoothing capacitors inside the unit but the power supplies should be regulated externally. If lower voltage supplies than ±15 Volts are used the unit will still work but with a lower sensitivity and smaller voltage swing of the output. A smaller dark current can also be expected. The 0 Volt power rail is not connected to the instrument case which is the reason the BNC signals are single ended.

A panel cut-out is provided to allow connection of a 25mm conduit with a male end connection. This provides a means of protecting optical fibres from mechanical damage on their way to the detectors. When the front panel detectors are being used it is suggested that this cut-out be closed with a 25mm screw and nut to complete the shielding provided by the casing itself.

Light detection is by pin diodes, reversed biased to 15 Volts (photoconductive mode) with a load resistor of 1MΩ. The pin diode acts as a current source and the voltage dropped across the load is amplified to produce a signal suitable for input to the data acquisition board. A higher bias voltage would give a higher sensitivity as mentioned earlier but would also lead to a larger dark current. Fifteen volts was chosen for the bias voltage because it is the highest regulated supply easily obtained commercially. A 1MΩ load resistor was chosen as a compromise between sensitivity, thermal noise and linearity. The large value of resistance leads to increased thermal noise but this is offset to some extent by the increased sensitivity from a large voltage drop across the resistor. The large resistor value also guarantees a linear voltage response with intensity. The pin diodes are FDR850 IRLCs supplied by Fibre Data in metal SMA housings. Using housed diodes helps to stop ambient light but it has been noted that extraneous light can reach the diodes through the back of housing. For this reason it is advised that the unit is always used with the lid on.

The amplifier chosen was a Burr Brown INA110KP, a high impedance FET input instrumentation amplifier. This device was chosen because of its high input impedance which stops it from sinking current from the 1MΩ load resistor, its low noise, it is highly linear and it also has precision on chip resistors for setting the gain. On chip resistors are useful because they reduce circuit size making the design smaller and less susceptible to radiated noise and they also make it easy to change the gain. The gain is set by linking various pins on the amplifier IC together, a task done in this design using a binary coded rotary switch. Presets for correcting amplifier offset were not used because the accuracy and temperature coefficients of external components were expected to be no better than the on chip components. The detection and amplifier circuit is shown in figure A.6 for one channel. All the channels are of the same design.
The PCB layout was designed such that the components for each channel were as close to each other as possible. This keeps the circuit as small as possible and reduces the amount of radiated noise picked up. The only long signal paths prior to amplification are from the pin diodes on the front panel to the PCB. To provide noise immunity these cables are made from shielded twisted pair. Noise immunity appears to be good but the bandwidth using the on board detectors is greater, suggesting that the capacitance of the longer leads is restricting the bandwidth. For this reason the on board pin diodes are normally used in preference to the ones on the front panel. The bandwidth of the receivers using the on board detectors has been measured as 17.4kHz. Amplifier outputs are connected to the BNC connectors using shielded cable connected as close as possible to the amplifiers. The cable shields are connected to the board's ground but not to the BNC shell and casing. Outputs are also tracked to the ribbon cable using the widest tracks that would fit. A type of bus is formed by adjacent channels which are separated with tracks at the ground potential to reduce crosstalk. These ground tracks also isolate the detector and amplifiers into single channel modules. Normal practice with analogue circuitry is to have a grounded star point to which the ground line of all modules are linked. In the receiver design a ground plane is used, a technique more common in digital electronics. This was because of the difficulty of routing sixteen ground lines around the signal bus and visa versa.
Figure A.7 shows the general component layout of the receiver PCB and indicates the grouping of components according to individual channels. Each channel has its own two switches A and B. Switch A selects either the pin diode detector on the front panel (position 1) or the pin diode detector on the board next to it (position 2). This may be remembered as to use the on-board detector push the switch A towards it, otherwise to use the detector on the front panel push the switch towards the amplifier IC.

Switch B is the binary coded decimal switch that selects the gain according to the table below. Position 9 is not used. This table is reproduced on the top of the PCB for reference during use.

<table>
<thead>
<tr>
<th>position of switch B</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
<th>9</th>
</tr>
</thead>
<tbody>
<tr>
<td>amplifier gain</td>
<td>1</td>
<td>100</td>
<td>200</td>
<td>300</td>
<td>500</td>
<td>600</td>
<td>700</td>
<td>800</td>
<td>10</td>
<td>not used</td>
</tr>
</tbody>
</table>

Table A.1. Amplifier gain settings.

Switches C and D are used to configure the 50 way ribbon cable for single ended or differential mode. In single ended mode the outputs from all sixteen channels are available on pins 3 through 18 with the ground pin being pin 19. In differential mode channels 1 to 8 use pairs of pins to carry the signals and the 0 volt level as in figure A.8. The 100kΩ resistor provides a return path to ground for the bias currents of the amplifier on the NB-MIO-16X board when used in differential mode [A.2]. When used in single ended mode this resistor should be shorted using the jumper beside it. Details of how to configure the data acquisition board for floating or ground referenced / single ended or differential connection are to be found in the user manuals. The receiver board was designed to have one 100kΩ resistor for each channel but was modified because the NB-MIO-16X multiplexes its inputs so that there is only one second stage amplifier and hence only one resistor is required. The board can easily be changed again if future systems do not use a multiplexer.

In configuring the receiver unit for differential mode both C and D should be in position 1 as indicated on the PCB. For single ended mode both switches should be in position 2 and the 100kΩ resistor shorted. It is possible to have a combination of differential and single ended channels as outputs by having C and D in different positions. The possibilities are summarised in table A.2.

If both the ribbon cable and BNCs are used to connect equipment at the same time some consideration may need to be given to loading effects.
Figure A.7. Receiver board component layout.
Figure A.8. Configuration for differential receiver output.

<table>
<thead>
<tr>
<th>position of switch C</th>
<th>position of switch B</th>
<th>1</th>
<th>2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>position of switch B</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>channels 1 to 8 differential mode</td>
<td>channels 1 to 4 &amp; 9 to 12 single ended; channels 5 to 8 differential mode</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>channels 1 to 4 differential mode; channels 5 to 8 &amp; 13 to 16 single ended</td>
<td>channels 1 to 16 single ended mode</td>
</tr>
</tbody>
</table>

Table A.2. Differential and single ended mode combinations.
A regulated power supply with a number of fixed voltage outputs has been built for the racking system. Rather than building a power supply from scratch the unit is based on two commercial power supply modules which are mounted in a 3U 19\text{inch} rack casing. The two modules are a linear supply (Coutant Lambda HTD1) and a switched mode supply (Computer Products NFS110-7604P). This arrangement means that the transmit and receive sides of a sensing system can have separate power supplies, the switched mode module having a high current capacity for driving sources and the linear supply having slightly better line regulation for sensitive receivers. The power supplies are forced air cooled using two fans to allow the power supply modules to be driven to their maximum power ratings.

The DC outputs provided are $+15,-15,+5$ Volts from both the switched mode and linear modules and also a $-5$ Volt output from the switched mode module. The specifications of each output are given in the table below.

<table>
<thead>
<tr>
<th>module</th>
<th>output voltage</th>
<th>maximum current</th>
<th>pk-pk ripple</th>
</tr>
</thead>
<tbody>
<tr>
<td>linear</td>
<td>$+15$ Volts</td>
<td>1.8 Amps</td>
<td>3.0 mV</td>
</tr>
<tr>
<td>linear</td>
<td>$-15$ Volts</td>
<td>1.8 Amps</td>
<td>3.0 mV</td>
</tr>
<tr>
<td>linear</td>
<td>$+5.0$ Volts</td>
<td>6.0 Amps</td>
<td>3.0 mV</td>
</tr>
<tr>
<td>switched mode</td>
<td>$+15$ Volts</td>
<td>5.0 Amps</td>
<td>150 mV</td>
</tr>
<tr>
<td>switched mode</td>
<td>$-15$ Volts</td>
<td>1.0 Amp</td>
<td>150 mV</td>
</tr>
<tr>
<td>switched mode</td>
<td>$+5.1$ Volts</td>
<td>10.0 Amps</td>
<td>50 mV</td>
</tr>
<tr>
<td>switched mode</td>
<td>$-5.0$ Volts</td>
<td>1.0 Amp</td>
<td>50 mV</td>
</tr>
</tbody>
</table>

Table A.3. Power supply performance.

The total output power from the linear supply should not exceed 75 W and from the switched mode supply 110 W. It is recommended that the switched mode module be used with a minimum load, this load is incorporated within the power supply casing itself.

A set of LEDs on the front panel indicate that the power supply outputs are functional. The left column of LEDs are connected to the linear module and the right column are connected to the switched mode module. The top (red) LEDs show when the $+15$ V supplies are working, the second row of (red) LEDs show when the $-15$ V supplies are working. Likewise, the orange LEDs are for the $+5$ V supplies and the green LED is for the $-5$ V supply.
The +15 V and -15 V outputs of the linear supply share the same 0 V level. The +5 V output has a separate 0 V point. Both of these 0 V levels are floating with respect to the building (mains) earth. Because the outputs are fully regulated, they may be tied together without effecting the power supply performance.

There is just a single, shared, 0 V level for all the switched mode module outputs. This is also floating with respect to the building earth and may be tied to the 0 V output(s) of the linear supply if so desired.

The power supply casing is connected to the building earth. Figure A.9 indicates the earthing system for the power supply and figure A.10 for the rack system as a whole. Note that the rack and instrument cases are connected to the building earth and that the ground rails of the instruments are floating with respect to the building's earth.

Figure A.9. Power supply earthing.
There are four fuses used in the power supply, two for the fans and one each for the power supply modules. The switched mode module has a 5A, 250 V fuse on the PCB, the other fuses are accessible from the rear panel. The linear power supply module requires a 1A, 240 V slow blow fuse and the fans each require a 250mA, 240 V fuse. The fans should switch on as soon as the supply is powered on. If this does not happen the fuses should be checked.

Figure A.10. 19" rack system earthing.
Synchronisation

Two different data acquisition systems have been used in this research. One is the National Instruments NB-MIO-16X board installed in a Macintosh computer which did not use any special synchronisation hardware and the other is a National Instruments Lab-PC+ installed in an IBM compatible. The PC system software was a general data acquisition program written in C++ which does need some purpose built hardware to synchronise the data acquisition and light modulation.

For the on-line experiments the Lab-PC+ board is triggered with an external signal which is synchronised with the light modulation so that measurements of background and electronic offsets can be made. This is done by arranging for each channel to be sampled firstly when the source is off and then when the source is on. All four channels are sampled as close together in time as possible and then there is a pause until the source has changed state. Limited bandwidth of the receivers means that there is a finite rise and fall time of the detected signal when the light source is switched. To allow time for the detected signal to rise or fall to its full value there is a delay between the switching of the source and the sampling of the channels.

The circuit of figure A.11 was built using wire wrap to control the source modulation and triggering. The data acquisition is triggered on a rising edge of the trigger signal. To improve noise immunity the trigger signal is normally held in a high logic state and pulsed low to trigger, the sampling occurring when the signal returns high. Even so some filtering is needed on the trigger to attenuate interference picked up on the cables between the synchronisation hardware and the Lab-PC+ board. This filtering is achieved simply by placing a capacitor between the trigger and ground. The timing for the circuit is provided by a 74LS124 which clocks a counter, 74LS590. One of the counter outputs is used to toggle a flip flop, 74LS107, which in turn is used to modulate the light source at 2kHz. A combination of 2 and 3 input AND gates (74LS11 and 74HCT08) generate the required sequence of pulses for the trigger from the counter outputs. The combination of logic results in a trigger sequence such that four trigger pulses are generated in quick succession in the second quarter of a source half period. A reset of the timer is made after the light source has changed its state so that the next sequence of trigger pulses occurs when the light source is in the opposite state. This is illustrated in figure A.12. The trigger is enabled via a two input AND gate only when a control line from the Lab-PC+ board is high. A monostable (4047B) is used to provide a reset pulse to the circuit when the control line is taken high and to provide an extra trigger pulse at the start of data acquisition. A 74HCT139 multiplexer switches between the monostable reset pulses and the timed trigger output depending on the state of the control line. A second monostable (74HCT123) placed between the multiplexer and the counter reset is used to generate a suitable pulse for resetting the counter from a negative edge transition.
Figure A.11. Data acquisition timing circuit.
Figure A.12. Synchronisation between source and trigger.
Stepper Motor Drive Circuit

The circuit of the stepper motor drive used to move a shutter is shown in figure A.13. A high ready signal from the PC triggers the monostable (74HCT123) to send a pulse which resets both the 74HCT4040 counters, forcing the trigger output low. The clocks to the counters are enabled by virtue that the clocks are NANDed with a combination of the counter outputs. As one counter counts up, its least significant bit is used to step the motor via a UCN5804 driver chip. Once the counter has counted far enough to have turned the motor 90° (a count of 100) the NAND gate logic disables the first counter's clock. The second counter continues to count up to 256 before the trigger output is taken high and the second clock is disabled. This delay from the motor finishing its movement to the board sending a trigger signal to the PC/camera is to ensure that a full exposure of the array is obtained before taking a reading. The positive transition of the trigger is also used to toggle a flip flop which controls the direction of the motor drive. In this way the motor's direction of turn is alternated so that even if the rotation is not exactly 90° the error does not build up with consecutive turns. The clock for the motor control is produced by a 555 timer. Power is obtained from a bench top supply with +5 and +15 volt regulated outputs. 100µF capacitors are placed on the motor control board to decouple the +5V supply.
Figure A.13. Stepper motor drive circuit.
# Appendix B

## Summary of Experimental Details

### On-line Experiment Conditions

<table>
<thead>
<tr>
<th>experiment / pollutant</th>
<th>flow speed, l/min</th>
<th>start temp, °C</th>
<th>end temp, °C</th>
<th>notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>mkI / HeNe / soluble oil</td>
<td>≈17.5</td>
<td>not known</td>
<td>not known</td>
<td>gains: 45° = 500</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>90°, 300° = 800</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>135° = 600</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>240° = 700</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>180° = 1</td>
</tr>
<tr>
<td>mkI / white light / soluble oil</td>
<td>≈17.5</td>
<td>not known</td>
<td>not known</td>
<td>low power source</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>gains: 90° = 800</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>45°, 135° = 100</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>240° = 200</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>300° = 500</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>180° = 1</td>
</tr>
<tr>
<td>mkI / white light / soluble oil</td>
<td>≈17.5</td>
<td>not known</td>
<td>not known</td>
<td>high power source</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>gains as previous expt.</td>
</tr>
<tr>
<td>mkI / LED / soluble oil</td>
<td>≈17.5</td>
<td>26.4</td>
<td>33.2</td>
<td>gains: 180° = 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>others = 800</td>
</tr>
<tr>
<td>mkI / HeNe / tap water (i)</td>
<td>≈17.5</td>
<td>18.8</td>
<td>31.5</td>
<td>Varying temp. exp.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>gains: 45° = 200</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>180° = 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>others = 800</td>
</tr>
<tr>
<td>mkI / HeNe / tap water (ii)</td>
<td>≈17.5</td>
<td>19.3</td>
<td>30.0</td>
<td>Varying temp. exp.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>gains: 45° = 200</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>180° = 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>others = 800</td>
</tr>
<tr>
<td>mkI / LED / crude 1</td>
<td>≈17.5</td>
<td>29.4</td>
<td>32.9</td>
<td>gains: 180° = 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>others = 800</td>
</tr>
<tr>
<td>mkI / LED / crude 1</td>
<td>≈17.5</td>
<td>29.0</td>
<td>32.3</td>
<td>Flow speeded up at injection.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>gains as previous expt.</td>
</tr>
<tr>
<td>mkI / LED / soluble oil</td>
<td>≈17.5</td>
<td>31.7</td>
<td>34.8</td>
<td>Flow speeded up at injection.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>gains as previous expt.</td>
</tr>
<tr>
<td>mkI / LED / crude 2</td>
<td>≈17.5</td>
<td>29.0</td>
<td>31.3</td>
<td>Flow speeded up at injection.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>gains as previous expt.</td>
</tr>
<tr>
<td>Material</td>
<td>Variation</td>
<td>Temperature</td>
<td>Flow Speed</td>
<td>Gain</td>
</tr>
<tr>
<td>---------------------------</td>
<td>-----------</td>
<td>-------------</td>
<td>------------</td>
<td>------</td>
</tr>
<tr>
<td>mkl / LED / crude 2</td>
<td>±17.5</td>
<td>28.9</td>
<td>31.4</td>
<td>Flow speeded up at injection. gains as previous expt.</td>
</tr>
<tr>
<td>mkl / LED / crude 2</td>
<td>±17.5</td>
<td>29.3</td>
<td>31.3</td>
<td>Flow speeded up at injection. gains as previous expt.</td>
</tr>
<tr>
<td>mkl / LED / motorcycle oil</td>
<td>±17.5</td>
<td>31.9</td>
<td>34.2</td>
<td>Flow speeded up at injection. gains as previous expt.</td>
</tr>
<tr>
<td>mkl / LED / motorcycle oil</td>
<td>±17.5</td>
<td>27.4</td>
<td>32.7</td>
<td>Flow speeded up at injection. gains as previous expt.</td>
</tr>
<tr>
<td>mkl / LED / Castrol GTX</td>
<td>±17.5</td>
<td>31.4</td>
<td>34.6</td>
<td>Flow speeded up at injection. gains as previous expt.</td>
</tr>
<tr>
<td>mkII / CW LED / tap water</td>
<td>10 - 50</td>
<td>21.1</td>
<td>23.7</td>
<td>Varying flow speed experiment. gains as previous expt.</td>
</tr>
<tr>
<td>mkII / CW LED / tap water</td>
<td>10</td>
<td>10.0</td>
<td>30.0</td>
<td>Varying temperature experiment. gains as previous expt.</td>
</tr>
<tr>
<td>mkII / CW LED / tap water</td>
<td>20</td>
<td>10.0</td>
<td>30.0</td>
<td>Varying temperature experiment. gains as previous expt.</td>
</tr>
<tr>
<td>mkII / LD / Castrol GTX</td>
<td>20</td>
<td>30.3</td>
<td>35.4</td>
<td>Flow speeded up at injection. gains: $180^\circ = \text{attenuated others} = 800$</td>
</tr>
<tr>
<td>mkII / LD / Castrol GTX</td>
<td>20</td>
<td>30.0</td>
<td>33.8</td>
<td>Flow speeded up at injection. gains: $180^\circ = \text{attenuated others} = 800$</td>
</tr>
<tr>
<td>mkII / LD / crude 1</td>
<td>20</td>
<td>30.0</td>
<td>33.9</td>
<td>Flow speeded up at injection. gains: $180^\circ = \text{attenuated others} = 800$</td>
</tr>
<tr>
<td>mkII / LD / crude 1</td>
<td>20</td>
<td>31.0</td>
<td>34.7</td>
<td>Flow speeded up at injection. gains: $180^\circ = \text{attenuated others} = 800$</td>
</tr>
<tr>
<td>mkII / LD / crude 2</td>
<td>20</td>
<td>30.0</td>
<td>34.6</td>
<td>Flow speeded up at injection. gains: $180^\circ = \text{attenuated others} = 800$</td>
</tr>
<tr>
<td>mkII / LD / crude 2</td>
<td>20</td>
<td>30.0</td>
<td>33.9</td>
<td>Flow speeded up at injection. gains: $180^\circ = \text{attenuated others} = 800$</td>
</tr>
</tbody>
</table>
## Off-line Experiment Conditions

<table>
<thead>
<tr>
<th>experiment / suspension</th>
<th>stirrer speed, rpm</th>
<th>number of data samples</th>
<th>reliability measure</th>
<th>notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>empty cuvette (i)</td>
<td>0</td>
<td>10 000</td>
<td>192</td>
<td></td>
</tr>
<tr>
<td>empty cuvette (ii)</td>
<td>0</td>
<td>8 000</td>
<td>968</td>
<td></td>
</tr>
<tr>
<td>tap water (i)</td>
<td>100</td>
<td>8 000</td>
<td>726</td>
<td></td>
</tr>
<tr>
<td>tap water (ii)</td>
<td>50</td>
<td>20 000</td>
<td>673</td>
<td></td>
</tr>
<tr>
<td>polystyrene spherules (i)</td>
<td>100</td>
<td>20 000</td>
<td>904</td>
<td></td>
</tr>
<tr>
<td>polystyrene spherules (ii)</td>
<td>50</td>
<td>10 000</td>
<td>124</td>
<td></td>
</tr>
<tr>
<td>polystyrene spherules (iii)</td>
<td>50</td>
<td>10 000</td>
<td>145</td>
<td>continuation of previous expt.</td>
</tr>
<tr>
<td>desert dust (i)</td>
<td>50</td>
<td>10 000</td>
<td>847</td>
<td></td>
</tr>
<tr>
<td>desert dust (ii)</td>
<td>50</td>
<td>10 000</td>
<td>222</td>
<td></td>
</tr>
<tr>
<td>desert dust (iii)</td>
<td>50</td>
<td>10 000</td>
<td>288</td>
<td>follow on from tap water (ii)</td>
</tr>
<tr>
<td>soluble oil (i)</td>
<td>50</td>
<td>10 000</td>
<td>556</td>
<td></td>
</tr>
<tr>
<td>soluble oil (ii)</td>
<td>50</td>
<td>5 000</td>
<td>414</td>
<td></td>
</tr>
<tr>
<td>ballotini (i)</td>
<td>200</td>
<td>5 000</td>
<td>1021</td>
<td></td>
</tr>
<tr>
<td>ballotini (ii)</td>
<td>200</td>
<td>5 000</td>
<td>706</td>
<td>continuation of ballotini (i)</td>
</tr>
<tr>
<td>ballotini (iii)</td>
<td>200</td>
<td>10 000</td>
<td>826</td>
<td></td>
</tr>
</tbody>
</table>
Appendix C

Linescan Camera Software

The linescan camera programme is split into several different routines, the first of which initialises the values of some variables. A default file extension of .dat is set for the output file. The stepper motor drive is also disabled until the software is ready to start making measurements by setting the auxout 1 port to a logic low. Next the user is asked to input some parameters:

1. the number of data sets or whole experiments to be performed
2. the file name for the output
3. the directory path.

Because the Intraspec Basic does not have the full string handling ability of Basic, the file name should be given without an extension and the directory path should have the correct syntax of backslashes including one at the end as a separator to the filename.

The second subroutine simply zeros the arrays in which all the different measures will be stored. The background store is an exception because that is initialised with the saturation value of the linescan camera. This ensures that the minimum values at each pixel can be identified and stored later.

The third subroutine makes the required measurements. First the stepper motor is triggered by taking the port auxout 1 to a logic high. When the motor drive circuit returns a trigger pulse a single scan is taken from the camera. Readings are taken from the photodiode array with the minimum integration time possible of 20ms. Having taken a set of measurements from the array auxout 1 is again taken to a logic low level. Measurements of the background are taken first to check the offset levels of the array and to check that the noise is Gaussian as expected for thermal noise in the electronics. For this the shutter must start in the open position so that the first rotation causes the source to be blocked. A number of measurements are usually taken in a row, calculating cumulants as the experiment progresses, before a ready signal is output on auxout 1 to start the shutter travel. Once the shutter has opened and the trigger signal has been taken high a series of measurements of the scattered light intensities are made. During these measurements of the scattered light, the array is checked to see if any of the elements have been saturated. If it is found that the array has been saturated a subroutine is called to make a note of the number of times such an event occurs. A later addition to the program was
to store the minimum value of intensity measured at each element across the array. This is done in a subroutine and was to give some idea of what any static speckle was like, it assumes that the total measured intensity is the sum of the static speckle intensity and dynamic speckle intensity, that is there is no interference between the two. The static speckle measure is stored in the background store.

After the measurements have been checked for saturations and minimum values the cumulants are calculated. These are calculated as the measurement progresses for the light intensity samples and the light intensity with background subtracted. Cumulants with the background subtracted can be calculated using the mean already measured for the background.

Spatial correlations are also partly calculated in the third subroutine using the signal with the mean dark current subtracted. The correlations are calculated at a number of positions across the CCD array, the spacing between the chosen points being determined by a variable initialised near the start of the program code. The full normalised correlation coefficients are worked out later but the cumulants of the cross multiplied samples must be calculated here before the individual data are lost.

After the program has looped the required number of times such that a minimum number of samples have been collected, a test is made on the size of the moments in comparison to the number of samples to see how reliable they are. The test is based on the fact that if the moments are reliable then the variance of the moments will be small compared to the square of the moments themselves. It should be pointed out that this is a necessary but not a sufficient condition so it provides a guide to reliability rather than a definitive measure.

The last routine called by the programme stores the data to the named file. First the number of samples, the number of saturated readings and the reliability measure are written to the file. Next the moments are saved in columns, the first three columns being the mean, the second moment and the third moment of the measured signal, the next three columns being those with the background subtracted and the next three being those of the background itself. In following columns the correlations are stored and finally the static speckle measure.

If the operator entered a number greater than one for the number of data sets to be collected at the start of the experiment, the main programme loop will repeat until the required number of tests have been completed.
Program Listing

rem **************************************************************
rem * Dynamic Light Scattering Program
rem * for Oriel line scan camera
rem * D A Green  October 94
rem * last modified 18.7.95
rem **************************************************************

gosub.init
while p > 0
gosub.clear
gosub.measure
gosub.scale
gosub.file
p = p - 1
wend
end

.init
rem ******************************************************
rem * initialise configuration and variables
rem **********************************************

auxout 1 = off
rem Disable trig until ready
a = stores
rem Initialise loop counters
r = 1000
rem reliability parameter
m = 0
rem main loop count
l = 0
rem sub loop count
j = 10
rem No. reads in sub loop
x = 0
rem No. of saturated readings
k = 100
rem Step between correlations
n = 5
rem Nominal No. elements per sample
p = 1
rem No. of passes through main loop
q = 10
rem Quality factor - how much bigger
rem n should be than N Crit
x$ = "dat"
rem Default filename extension
cls
rem Clear the screen
input "Enter number of sets wanted: " p
rem * get file name and directory
a = 0
while a = 0
rem Get output file name
input "Enter name of output file (max 8 chars): " f$
cls: a = 1
if (len(f$) > 8) then
    print "Invalid filename"
a = 0
endif
wend

258
t$ = f$; x$

input "Give directory path : " p$

f$ = p$; t$

cls

return

clear

rem Add extension to file name

rem Get path name for data file

rem Add path to filename

cls

a = stores

while (a-5) > 0

zero #(a-5)

a = a - 1
wend

a = 0

while a < diodes

#bg[a] = 65535

a = a + 1
wend

zero #sig

zero #ref

zero #source

zero #live

return

.measure

rem Zero all numbered stores

rem Saturate static speckle store

rem Zero all other stores

print "ready"

m = 0

while m < n

zero #13

l = 0

rem initialise sub loop count

auxout 1 = on

rem Enable trigger

run(0,1,1,0,0,02,0,0,0,0,0)

rem Take background reading

auxout 1 = off

rem Disable trigger

gotoxy(0,1)

print "bg "; m

while l < j

rem take j samples

#7 = #7 + #sig

rem Sum background mean

#11 = #sig * #sig

rem Square background

#8 = #8 + #11

rem Sum 2nd moment of bg

#11 = #11 * #sig

rem Cube background

#9 = #9 + #11

rem Sum 3rd moment of bg
#13 = #13 + #sig
run(O,1,0,0.0,0.02,0,0,0,0,0)
l = l + 1
wend

#13 = #13 / j
auxout l = on
run(O,1,0,0.0,0.02,0,0,0,0,0)
auxout l = off
gotoxy(0,1)
print "sig ":m
l = 0
while l < j
rem short term mean
rem take another sample
rem scale cumulant
rem Enable trigger
rem Get sample, Min exposure
rem Disable trigger
rem take block of j samples
rem store data
rem check for array saturation
if(max(#sig,0,diodes-l) ~ 0) then
gosub.warn
endif
gosub.static
rem check for static speckle
rem Sum 1st moment
rem Square signal
rem Sum 2nd moment
rem Cube signal
rem Sum 3rd moment
rem store signal
rem measurement - background
a = 0
b = 12
while b < diodes
rem 1st element for correlation
rem Calculate correlations
##(14+a) = #(14+a) + #11[b] * #11
b = b + k
rem Increment angle
rem Increment store
wend
#4 = #4 + #11
#12 = #11 * #11
#5 = #5 + #12
#11 = #12 * #11
#6 = #6 + #11
#12 = #11 * #11
#10 = #10 + #12
run(O,1,0,0,0.02,0,0,0,0,0)
l = l + 1
wend
m = m + 1 :rem Increment loop counter

if(m == n) then :rem Check validity of moments
    print "checking validity of moments"
    #10 = #10 / #6
    #10 = #10 / #6 :rem See Keith's notes on N Crit
    r = max(#10,0,diodes-1) :rem Find max value of var 3rd moment
    :rem show confidence factor

if(r * q > n * j) then :rem if n not >> Ncrit
    print "10 * ";r;" > n"
    input "type I to stop & save now" b

if (b != 1) then :rem escape if want to stop
    n = 2*n :rem Double n
    if (n > 100000) then :rem Escape if n getting too big
        n = 100000
    endif
    cls
    endif
endif
endif
wend
rem * End of loop to build up moments
return

.scale
rem *******************************************************************************
rem * Divide sums by number of elements
rem *******************************************************************************

n = n * j
#1 = #1 / n
#2 = #2 / n
#3 = #3 / n
#4 = #4 / n
#5 = #5 / n
#6 = #6 / n
#7 = #7 / n
#8 = #8 / n
#9 = #9 / n

b = 12
a = 0
while b < diodes :rem Calculate averages
    #11 = #5[b] * #5 :rem mean^2(b) * mean^2(theta)
    #11 = sqrt(#11) :rem Square root
    #11 = #11 * n :rem pre-average #14
    #(14+a) = #(14+a) / #11 :rem normalise correlations
    b = b + k :rem Increment angle
    a = a + 1 :rem Increment store
wend
return
FILE

rem *****************************************************************************************************************
rem * Save data to file  *
rem *****************************************************************************************************************

\texttt{a = 0}
\texttt{print "saving data"}
\texttt{write(f$,"n = ";n,;)} \text{ rem Store No. elements used }
\texttt{write(f$,"sat. = ";x,;)} \text{ rem Stores No. saturated readings }
\texttt{write(f$,"r = ";r)} \text{ rem Store reliability parameter }
\texttt{while a < diodes}
\texttt{\hspace{1em}b = 12}
\texttt{\hspace{1em}c = 0}
\texttt{\hspace{1em}write(f$,#1[a],#2[a],#3[a],;)} \text{ rem Stores 1st 3 moments }
\texttt{\hspace{1em}write(f$,#4[a],#5[a],#6[a],;)} \text{ rem Stores corrected moments }
\texttt{\hspace{1em}write(f$,#7[a],#8[a],#9[a],;)} \text{ rem Store background moments }
\texttt{\hspace{1em}while b < diodes}
\texttt{\hspace{2em}write(f$,#(14+c)[a],;)} \text{ rem Store cross correlations }
\texttt{\hspace{2em}b = b + k}
\texttt{\hspace{2em}c = c + 1}
\texttt{\hspace{2em}wend}
\texttt{write(f$,#bg[a],;)} \text{ rem Store static speckle }
\texttt{write(f$,) \text{ rem Print new line character }
\texttt{a = a + 1}
\texttt{wend}
\texttt{return}

.rem
rem *****************************************************************************************************************
rem * print up number of saturated reads  *
rem *****************************************************************************************************************

\texttt{x = x+1}
\texttt{gotoxy(0,10)}
\texttt{print "array saturated ";x;" times"}
\texttt{return}

 statically
rem *****************************************************************************************************************
rem * check for minimum values which might be static speckle  *
rem *****************************************************************************************************************

\texttt{a = 0}
\texttt{while a < diodes}
\texttt{\hspace{1em}if (#sig[a] < #bg[a]) then}
\texttt{\hspace{2em}#bg[a] = #sig[a]}
\texttt{\hspace{1em}endif}
\texttt{\hspace{1em}a = a + 1}
\texttt{wend}
\texttt{return}

.rem Stop