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Shaping the focal field in three dimensions using polarisation and phase

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Supervisors:
Prof. J. C. Dainty & Dr. D. Lara

A thesis submitted in partial fulfilment of the requirements for the degree of Doctor of Philosophy

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Abstract

Vectorial polarimetry is a novel high-resolution microscopy technique with potential applications in the characterisation of nano-materials, sub-diffraction limit microscopy, and single molecule imaging. Previous work had concentrated on using homogeneous polarisation distributions in the pupil plane of a high numerical aperture objective lens, as well as analysing the scattered and re-collimated light from a nano-scale sample. It has been shown that sub-resolution information can be obtained by measurement of the Stokes parameters of the scattered light. The novel aspect of the vectorial polarimetry system described in this thesis lay in its ability to generate a completely arbitrary polarisation and phase distribution in the pupil of an objective lens. Three passes on two liquid crystal spatial light modulators were used to achieve this; the first pass controlled the absolute phase, while the remaining two were used to tailor the polarisation distribution across the beam.

The system focused a laser beam using a high numerical aperture objective lens; the focal field of such a lens is affected by both the polarisation and phase distributions across the entrance pupil. The field at the focus of the microscope lens was modelled using both the Debye-Wolf integrals and a Fourier transform method. Results from these two methods using similar inputs were found to be sufficiently similar for most applications. This modelling was used to investigate the extent to which the focal field could be shaped using polarisation and phase.

Experiments were carried out by placing a specimen in the focal plane, and collecting and re-collimating the light scattered from the sample. The polarisation state of the field in the exit pupil was then analysed. Sub-resolution displacements of a nano-sphere could be measured, where a change in one of the Stokes parameters increased as the sphere was moved away from the focus. The second sample investigated was a set of gratings with pitches smaller than the diffraction limit. The Mueller matrices of each of the gratings were measured. Decomposition of these matrices showed that polarimetric properties, such as retardance, depended on the pitch of the gratings; the retardance of a 45 nm grating was different than that for a 32 nm grating, suggesting that polarisation can reveal sub-wavelength structural changes.
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<td>BE</td>
<td>Beam expander</td>
</tr>
<tr>
<td>BS</td>
<td>Beam splitter</td>
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<tr>
<td>CCD</td>
<td>Charge coupled device</td>
</tr>
<tr>
<td>DOP</td>
<td>Degree of polarisation</td>
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<tr>
<td>DOAP</td>
<td>Division of amplitude polarimeter</td>
</tr>
<tr>
<td>ECM</td>
<td>Eigenvalue calibration method</td>
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<td>FPALM</td>
<td>Fluorescence photoactivation localisation microscopy</td>
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<tr>
<td>FT</td>
<td>Fourier transform</td>
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<tr>
<td>HWP</td>
<td>Half waveplate</td>
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<tr>
<td>LC</td>
<td>Liquid crystal</td>
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<tr>
<td>LCoS</td>
<td>Liquid crystal on silicon</td>
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<tr>
<td>LCVR</td>
<td>Liquid crystal variable retarder</td>
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<td>Ls</td>
<td>Laser</td>
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<td>LUT</td>
<td>Look-up table</td>
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<td>MM</td>
<td>Mueller matrix</td>
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<tr>
<td>NA</td>
<td>Numerical aperture</td>
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<tr>
<td>OBJ</td>
<td>Objective lens</td>
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<td>NSOM</td>
<td>Near-field scanning optical microscopy</td>
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<tr>
<td>PALM</td>
<td>Fluorescence photoactivation localisation microscopy</td>
</tr>
<tr>
<td>PSA</td>
<td>Polarisation state analyser</td>
</tr>
<tr>
<td>PSG</td>
<td>Polarisation state generator</td>
</tr>
<tr>
<td>SLM</td>
<td>Spatial light modulator</td>
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<tr>
<td>SMP</td>
<td>Sample</td>
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Contributions to the field

Several original results were obtained during the course of the work carried out for this thesis. These related to both the modelling of three-dimensional focal fields and the building of an experimental system capable of generating these fields.

In Chapter 3 original findings comparing results obtained using two of the most common methods for calculating the field at the focus of a high-NA lens are presented. These show that the methods are in agreement for two example input polarisation and phase distributions as the sum of relative errors was found to be on the order of $10^{-4}$, which should be sufficient agreement for most applications. Further results presented in this chapter also show the scope of the use of polarisation and phase in shaping the focal field of a high-NA lens, in particular when the relative phase is defined in terms of Zernike polynomials.

The experimental system described in Chapter 5 was capable of generating any polarisation and phase distribution using three passes on two liquid crystal spatial light modulators. This amounted to two linearly independent rotations of the input polarisation state such that any point on the Poincarè sphere could be reached. The remaining pass modulated the absolute phase of the incident beam, and did not alter the polarisation state. This system was used as the polarisation state generator of an imaging Mueller matrix polarimeter.

Experimental results were obtained using a set of gratings, the pitches of which were smaller than the classically defined resolution of the objective lens. These results are described in Chapter 7. From the measurement of the Mueller matrices of these gratings, it was possible to distinguish between the polarisation effects of a grating of 25 nm pitch and another with 32 nm pitch.
List of publications

Peer-reviewed publications


Conference publications


Chapter 1

Background: polarisation and polarimetry

1.1 Polarised light

The polarisation state of an electromagnetic wave is equivalent to the orientation of vibration of its electric field [Hecht 2002]. This orientation may be constant, which is the case for linearly polarised light. It can also vary periodically with time, for example with circularly polarised light, which carves out a circular pattern as it propagates. Elliptically polarised light can be described as any mixture between circularly and linearly polarised light. If the $x$- and $y$-components of the field are uncorrelated then the light is said to be unpolarised.

Circularly polarised light can always be resolved into two equal orthogonally polarised components. These two components have a phase difference with respect to each other of one quarter of a wave (or $\pi/2$). It is this relative phase that gives rise to the circular pattern made by the electric field as it propagates. The concept of the propagation of circularly polarised is illustrated by Fig. 1.1.

The two types of circular polarisation are termed as left- and right-circular; the difference between them is in the direction of rotation of the electric field as they propagate. Left-circularly polarised light rotates anticlockwise as it propagates towards an observer, while right-circular rotates clockwise when viewed from the same position. These two differ in the sign of their relative phases: $\pi/2$ will give rise to left-circular, while $-\pi/2$ results in right-circular, when the relative phase, $\delta$, is defined as $\delta = \delta_x - \delta_y$.

Ellipticity can be introduced to any linearly polarised light by generating an appropriate relative phase between orthogonally polarised components. An optical element that can generate this phase difference will have a different
1.1. Polarised light

Figure 1.1: Circular polarisation arises from a phase difference of $\pi/2$ between orthogonal field components. Taken from [Hecht 2002]. Peaks or troughs of one orientation of polarisation correspond to points of zero amplitude in the orthogonal component.

refractive index depending on the polarisation of the incident light. Calcite is one type of crystal that has this property, which arises out of its anisotropic lattice structure. This property of a material is known as its birefringence. It can be used to generate a relative phase between orthogonal polarisation states.

Birefringence is defined as the refractive index difference for two orthogonal axes of propagation through the material. The axis with a larger refractive index is known as the slow axis, due to the speed of light being reduced when propagating in this direction through the material. Conversely, the axis with a lesser refractive index is termed the fast axis.

In this thesis, the property of retardance is often referred to instead of the birefringence. This property of a material is defined as the relative phase that will be generated between two orthogonal polarisation states of the light. It can be measured in either radians, fractions of a wave or units of distance. Birefringence is an intensive property of a material, while retardance can be understood as its corresponding extensive property.

A number of materials exist, the birefringence of which is variable and can
be controlled using an applied electric field or voltage. A subset of these is liquid crystals. The molecules of this type of substance are rod-shaped, and will rotate in response to an applied field. Their birefringence depends on the amount of rotation; therefore they can be used to generate an arbitrary relative phase. A number of optical elements that make use of this material were used in the final experimental system of this thesis. Further information will be provided on the use of this substance in an optical element will be provided in Chapter 4, when the characterisation of the polarisation sensitive elements used in the system will be described.

1.2 The Stokes parameters

Polarisation states of beam-like fields can be represented using the four element Stokes vector in this formalism [Shurcliff 1964, Born 1999, Goldstein 2011]. The general Stokes vector, $s$, that can be used to describe any polarisation state can be represented as

$$s = \begin{pmatrix} s_0 \\ s_1 \\ s_2 \\ s_3 \end{pmatrix} = \begin{pmatrix} I_{\text{total}} \\ I_H - I_V \\ I_{+45^\circ} - I_{-45^\circ} \\ I_R - I_L \end{pmatrix},$$ (1.1)

where $I_{\text{total}}$ is the total irradiance of the light being described, $I_H$ and $I_V$ are the irradiances of the light resolved into its horizontal and vertical components, respectively. In a similar way, $I_{+45^\circ}$, $I_{-45^\circ}$, $I_R$ and $I_L$ correspond to light polarised at $+45^\circ$ and $-45^\circ$, and right-circularly and left-circularly respectively.

The degree of polarisation (DOP) of the light under consideration depends on the ratio of polarised light to the total intensity, and is given by

$$DOP = \frac{\sqrt{(s_1)^2 + (s_2)^2 + (s_3)^2}}{s_0}. \tag{1.2}$$

Taking the example of pure horizontally polarised light in the context of Eq. 1.1, $I_H$ will be equal to one, while $I_V$ will be equal to zero as no vertically polarised light is present. Horizontally polarised light can be resolved into two
equal components at $\pm 45^\circ$; therefore $I_{45}$ and $I_{-45}$ will be equal and $s_3$ will be equal to zero. The same can be said for $s_3$, which is also equal to zero. Therefore, the Stokes parameters of horizontally polarised light are

$$
\begin{pmatrix}
1 \\
1 \\
0 \\
0
\end{pmatrix}.
$$

Using Eq. 1.2, the DOP of the above Stokes parameters can calculated to be equal to one.

### 1.3 Mueller and Jones calculus

There are a number of mathematical tools that can be used to model the effects of optical components on an arbitrary given polarisation state. The two most widely-used are Mueller and Jones calculus. Both of these will be used in this thesis, and so a brief description of them will be given. Two good resources for further information on these mathematical techniques are Shurcliff [Shurcliff 1964] and Goldstein [Goldstein 2011].

Both of these formalisms use square matrices to describe an optical element, and vectors to describe polarisation states. Multiplication of the matrix representing an optical element by the vector representation of any polarisation state will result in a vector that describes the polarisation state after the optical element. The vector used to describe polarisation states in Mueller calculus is the previously described Stokes vector.

#### 1.3.1 Mueller matrices

A Mueller matrix is a $4 \times 4$ matrix that describes the polarisation effect of any physically realisable optical element or system, and can be combined with any Stokes vector to calculate the effect of that element on the particular polarisation state. As in the case of the Stokes parameters, Mueller matrices describe the effect of beam-like fields on an optical element.

The simplest Mueller matrix is that of free space or air, which should have
no effect on the polarisation state of light as it propagates, and therefore is equivalent to the $4 \times 4$ identity matrix:

$$M_{\text{air}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}.$$ (1.4)

Another example of one of the simpler Mueller matrices is that of a mirror. A perfect mirror will have no effect on $s_0$ or $s_1$, as the overall irradiance and horizontal and vertically polarised light will remain constant. The effect of the mirror will be to change the sign of the $s_2$ and $s_3$ parameters:

$$M_{\text{mirror}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}.$$ (1.5)

The use of Mueller matrices to represent the polarisation effects of optical elements is best shown by example. Therefore, a simple example of the effect of a polariser will be given in the next section.

### 1.3.2 Mueller calculus

Mueller calculus is a mathematical tool for the description of polarisation states and optical components. Stokes vectors are used to represent polarisation states, and consist of column vectors with four elements. Mueller matrices represent any physically-realisable optical component, and are of dimension $4 \times 4$. Because Mueller algebra incorporates terms related to the total irradiance of the light, information about the depolarisation of the optical element can be computed, along with the degree of polarisation of the light. This is not the case for the Jones formalism where fully polarised light is assumed, and unpolarised light cannot be represented.

A simple example of the use of Mueller algebra is a polarised beam of light incident on a perfect polariser with its transmissive axis perpendicular to the direction of polarisation of the incident beam. In this case we take the incident beam to be vertically polarised and the polariser to be horizontally
transmitting. The Mueller matrix of a horizontal polariser is

\[
M_{\text{pol}}(0^\circ) = \tau \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix},
\] (1.6)

where \( \tau \) is the transmittance of the polariser, and is equal to 0.5 for a perfect polariser. The Stokes vector representing light vertically polarised is \( s_{\text{in}} \):

\[
s_{\text{in}} = \begin{pmatrix} 1 \\ -1 \\ 0 \\ 0 \end{pmatrix}.
\] (1.7)

The resulting Stokes vector, \( s_{\text{out}} \), after experiencing the polarisation effects of the elements of an optical system, is calculated by pre-multiplying the Mueller matrices of the individual elements, in order, by the Stokes vector of the incident beam, \( s_{\text{in}} \):

\[
s_{\text{out}} = M_1 \cdot M_2 \cdots M_N \cdot s_{\text{in}}
\] (1.8)

The Stokes vector of the beam after the horizontal polariser is calculated as follows:

\[
s_{\text{out}} = M_{\text{pol}}(0^\circ) \cdot s_{\text{in}}.
\] (1.9)

Computation of the above equation results in

\[
s_{\text{out}} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix},
\] (1.10)

which implies that, if the polariser is perfect, all of the vertically polarised light will be absorbed by the horizontal polariser. This agrees with the expected observation from such a system in the laboratory. Any optical system can be modelled using Mueller calculus; it is a useful method for prediction of the
polarisation effects of optical elements in any system.

A diattenuating material is one that preferentially absorbs a particular state of polarisation, while a retarding material is one that creates a relative phase between perpendicular polarisation states. The polarisation effects of many optical elements amount to a combination of these two properties, and another useful Mueller matrix is one that describes any linear-diattenuator and/or linear-retarder. The Mueller matrix of this type of optical element is

\[
M_{\text{gen}}(\tau, \Psi, \delta) = \tau \cdot \begin{pmatrix}
1 & -\cos 2\Psi & 0 & 0 \\
-\cos 2\Psi & 1 & 0 & 0 \\
0 & 0 & \sin 2\Psi \cos \delta & \sin 2\Psi \cos \delta \\
0 & 0 & -\sin 2\Psi \cos \delta & \sin 2\Psi \cos \delta
\end{pmatrix},
\]

(1.11)

where \(\tau\) is the total irradiance transmittance, \(\delta\) is the retardance, and \(\Psi\) is related to the relative transmission of parallel (\(\tau_{||}\)) and perpendicular (\(\tau_{\perp}\)) states as follows:

\[
\tan \Psi = \sqrt{\frac{\tau_{||}}{\tau_{\perp}}},
\]

(1.12)

A rotation of an optical component can represented in Mueller algebra by using the Mueller rotation matrix, \(M_{\text{rot}}\):

\[
M_{\text{rot}}(\theta) = \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & \cos 2\theta & -\sin 2\theta & 0 \\
0 & \sin 2\theta & \cos 2\theta & 0 \\
0 & 0 & 0 & 1
\end{pmatrix},
\]

(1.13)

where \(\theta\) is the angle by which the element is rotated. Rotation of an optical element by an azimuthal angle, \(\theta\), is represented by the following product:

\[
M(\theta) = M_{\text{rot}}(\theta) \cdot M(0^\circ) \cdot M_{\text{rot}}(-\theta),
\]

(1.14)

where \(M\) is the Mueller matrix of the optical element, and \(M_{\text{rot}}\) is the Mueller rotation matrix from Eq. 1.13.

The rotation of the Mueller matrix in Eqs. 1.11 and using Eq. 1.14 can be used as a general matrix to describe most optical elements, as the polarisation properties of most elements will be a combination of retardance and
1.3. Mueller and Jones calculus

diattenuation rotated at some angle.

1.3.3 The Poincaré sphere

The Poincaré sphere can be understood as a three-dimensional plot of the Stokes parameters. The six poles of the sphere can be used to compute the Stokes parameters in Eq. 1.1. All pure polarisation states are represented on the surface of the sphere. A plot of the sphere is shown in Fig. 1.2.

![Figure 1.2: The Poincaré sphere illustrated as a three-dimensional plot of the Stokes parameters. Linear polarisation states are positioned on the equator of the sphere, marked by a blue line in this figure, while the two pure circular polarisation states are situated at the poles.](image)

All linear polarisation states are represented along the equator of the sphere, this is region of the sphere where $s_3$ is equal to zero, i.e., no ellipticity is present. This is represented by the blue line in Fig. 1.2. Purely circularly polarised polarisation states correspond to the north and south poles of the
sphere. Any point that lies between the equator and either of the poles represents an elliptically polarised state. The angle of ellipticity depends on its angle along the equator. The azimuthal angle with respect to the equator indicates the degree of ellipticity present.

Purely polarised states are represented on the outer surface of the sphere. Looking at Fig. 1.2, it is clear that Eq. 1.2, the calculation of the DOP, is equivalent to calculating the displacement from the origin of the sphere, which will always be equal to one at the surface, indicating a purely polarised state.

The Poincaré sphere is a useful tool for visualising the effect of an optical element on any polarisation state. For example, the effect of a linear retarder on any polarisation state can be depicted as a rotation on the Poincaré sphere; the axis of this rotation is defined by the orientation of the fast axis of the retarder. Mueller analysis is used below to see this algebraically. The Mueller matrix of a retarder of retardance $\delta$ and with its fast axis at $0^\circ$ is

$$M_{\text{ret}}(\delta, 0^\circ) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & \cos \delta & \sin \delta \\ 0 & 0 & -\sin \delta & \cos \delta \end{pmatrix}. \quad (1.15)$$

In order to convert this into the Mueller matrix of a retarder with fast axis at an angle of $45^\circ$, the following matrix product is computed:

$$M_{\text{ret}}(\delta, 45^\circ) = M_{\text{rot}}(45^\circ) \cdot M_{\text{ret}}(\delta, 0^\circ) \cdot M_{\text{rot}}(-45^\circ), \quad (1.16)$$

where $M_{\text{rot}}(\theta)$ is the Mueller matrix representation for a rotation around the optical axis of an angle $\theta$, previously shown in Eq. 1.13.

We now consider the effect of a retarder, such as the one described by Eq. 1.16, on an incident beam which is linearly polarised in the horizontal direction. Its Stokes vector is

$$s_{\text{in}} = \begin{pmatrix} 1 \\ 1 \\ 0 \\ 0 \end{pmatrix}. \quad (1.17)$$
The Stokes vector of the beam after the retarder at 45° is

\[ s_{out} = M_{ret}(\delta, 45^\circ) \cdot s_{in}. \] (1.18)

Computation of Eq. 1.18, using Eqs. 1.17 and 1.16, reveals that the state of polarisation of the output beam is the result of a rotation of angle \( \delta \) on the surface of the sphere:

\[ s_{out} = \begin{pmatrix} 1 \\ \cos \delta \\ 0 \\ \sin \delta \end{pmatrix}. \] (1.19)

In our example, the starting point of the rotation is linear horizontally polarised light (see point H in Fig. 1.4), and the rotation is left-handed (clockwise) around the 45° axis. The states that are reachable are represented by the blue line in Fig. 1.3. A familiar example of this is when \( \delta = \pi/2 \), which makes the retarder in Eq. 1.18 a quarter waveplate at 45°. Starting from the point on the Poincaré sphere for horizontal polarisation, and rotating by \( \pi/2 \) clockwise around the 45° axis, the right-circular point on the sphere is reached. This is depicted in Fig. 1.3, where the rotation is described using retardance \( \delta \).

A single variable retarder at a fixed orientation can thus introduce a variable rotation on the Poincaré sphere around a fixed axis. In most experimental conditions the initial state of polarisation in the laboratory is fixed; therefore, in order to reach any point on the surface of the Poincaré sphere, two linearly independent rotations – equivalent to two retardances – are needed [Lara 2006]. In other words, two variable retarders are necessary to be able to generate any state of fully polarised light (i.e. to attain full polarisation control).

### 1.3.4 Jones calculus

Another widely used mathematical formalism used to describe polarisation states and the polarisation effect of an optical element is Jones calculus. Description of this calculus can be found in Shurcliff [Shurcliff 1964]. As is the case in Mueller calculus, optical elements are represented by matrices, and
Figure 1.3: A rotation around the sphere can represent the effect of a retardance. In this figure, $\delta$ represents a $\pi/2$ retardance that converts horizontally polarised light to right-circular.

Polarisation states are represented as vectors. However, Jones calculus uses $2 \times 2$ matrices and 2 element vectors.

The two elements of a Jones vector, usually equivalent to $E_x$ and $E_y$ in the appropriate coordinate system, represent the fractions of the light that is polarised in orthogonal directions. For example light polarised in only the $x$ direction is represented by

$$j = \begin{bmatrix} 1 \\ 0 \end{bmatrix}.$$  \hfill (1.20)

Ellipticity is represented by introducing complex terms to the individual elements of the vector, such that the correct phase difference exists between the two. For example, left circular polarisation is represented by

$$j = \frac{1}{\sqrt{2}} \begin{bmatrix} i \\ 1 \end{bmatrix}.$$  \hfill (1.21)
1.4. Mueller matrix polarimetry

An advantage of the use of Jones calculus over Mueller is that Jones can include description of the absolute phase, while Mueller cannot. Jones calculus, however, assumes that the light is fully polarised and coherent. It cannot be used to represent depolarising or scattering devices [Shurcliff 1964].

These two formalisms are useful in differing situations due to the capabilities mentioned above. For example, when measuring a polarisation state it is best to use Mueller calculus, as it is likely that a certain degree of unpolarised light will be present. However, when doing theoretical calculations, full polarisation is often assumed and it can be advantageous to consider the absolute phase of the light.

1.4 Mueller matrix polarimetry

A Mueller matrix polarimeter is an instrument that can measure the Mueller matrix of an optical element, and in order to do so must be able to measure the Stokes vector of any polarisation state using a polarisation state analyser (PSA). It must also be able to generate at least four linearly independent polarisation states, using a polarisation state generator (PSG) [Goldstein 2011].

1.4.1 Mueller matrix and Stokes measurement

In order to explain the measurement of the Stokes parameters, we must look again at its definition:

\[
\mathbf{s} = \begin{pmatrix}
I_{\text{total}} \\
I_H - I_V \\
I_{45^\circ} - I_{+45^\circ} \\
I_R - I_L
\end{pmatrix}
\]  

(1.22)

This equation relates the polarisation state to the irradiances \(I_H\), \(I_V\), \(I_{45^\circ}\), \(I_{-45^\circ}\), \(I_R\), and \(I_L\), which are measurable using combinations of polarisers and waveplates, along with a detector.

Measurement of the Mueller matrix of a specimen involves generating a sequence of polarisation states, which are then incident on a sample, and measuring the Stokes vector of the transmitted or reflected light by analysing at least four of the same set of polarisation states. The chosen states will depend
on the most convenient coordinate system for the setup being used. The irradiance measurements provide information on the response of the sample to the incident polarisation state, i.e., whether light has experienced a retardance and been changed into another polarisation state, whether the light has been either diattenuated, or if it has been depolarised. If the incident polarisation states are linearly independent, the irradiance measurements can be used to construct the measured Mueller matrix. Simultaneous equations for this are given in [Bickel 1985].

Once the Mueller matrix has been measured, a number of physical quantities can be extracted from its elements. For example, the diattenuation is related to elements 2, 3 and 4 of the first column. Also the retardance and angle of the fast and slow axes of the sample are related to the square of coefficients from \( m_{22} \) to \( m_{44} \). A method for this decomposition, which will be used later on some experimental results in this thesis, can be found in [Lu 1996]. This method decomposes a given Mueller matrix into a product of three matrices, each separately representing the depolarisation, diattenuation and retardance of the Mueller matrix.

### 1.4.2 Mueller matrix polarimeter systems

A number of configurations of Mueller matrix polarimeters exist, a few of which will now be discussed. Their common feature is that they must all include both a PSA and PSG, with the sample placed in between the two. In most Mueller matrix polarimeters, the PSG will generate at least four linearly independent polarisation states sequentially using, for example, Pockels cells [Rodríguez-Herrera 2010, Lara 2006, Delplancke 1997], or rotating polarisers and waveplates [Azzam 1978, Hauge 1978]. After transmission or reflection off a sample, the Mueller matrix will be measured using the PSA.

One of the most common is the division-of-amplitude polarimeter (DOAP). The division of amplitude occurs when the light is split into a number of paths, and simultaneous measurements of the necessary irradiances to reconstruct the Mueller matrix are recorded. An advantage of this configuration is that it can have fast acquisition times. However, some disadvantages are that they can be bulky and that errors can arise in the alignment of each separate path. For an imaging polarimeter, it is desirable for each the paths should be the
exact same length in order to have the same magnification and scaling on
each path. Another disadvantage of this configuration of polarimeter is that
the signal-to-noise ratio will be reduced. This arises from the splitting of the
irradiance.

Another type of Mueller matrix polarimeter measures the required irra-
diances sequentially. This type of polarimeter can use voltage controlled
retarders such as liquid crystal variable retarders (LCVRs) [Bueno 2000,
De Martino 2003] or photo-elastic modulators [Compain 1998]. These vari-
able components are used in conjunction with a polariser. Their operation
involves conversion of the desired polarisation state, using retardances, such
that it is isolated and maximally transmitted by the polariser. The state in
question is isolated using the polariser and its irradiance measured afterwards
using a single detector.

Regarding the system built for this thesis, it was decided to use a sequential
type polarimeter. This was because speed of measurement was not anticipated
to be of crucial importance, as well as more straightforward alignment. There
are also advantages to using a single detector, such as ease of calibration and
certainty that path lengths are equal for all irradiance measurements. Six
polarisation states were used in both the PSA and PSG (corresponding to
the six poles of the sphere), resulting in 36 irradiance measurements. An
advantage to using this is that the measured Mueller matrix coefficients are
independent of each other, which reduces the propagation of errors in the
measurements. The two iterations of this system will be described in detail
in Chapter 4.

1.5 Spatial polarisation control

The experimental system described in this thesis generates polarisation and
phase distributions in the entrance pupil of microscope objective lens that
vary spatially across the beam. Tailoring polarisation across a beam has
become a popular field of research due to uses in areas such as optical trapping
[Zhan 2004], laser cutting [Niziev 1999], and improved resolution in imaging
[Sheppard 2004].

A number of methods currently exist for spatial manipulation of
the polarisation distribution. Two of these are spatially varying waveplates [Spilman 2007, Beresna 2010, Beresna 2011], and liquid crystal spatial light modulators (SLMs) [Wang 2007, Boruah 2009, Bashkansky 2010, Beversluis 2006, Moreno 2012], which are devices that can apply a programmable retardance pattern to an incident beam. A detailed description of the use of this type of SLM will be given in Chapter 4.

1.5.1 Complete polarisation control

The PSG used in this system constituted liquid crystal SLMs whose retardance could be controlled using an applied voltage. These elements were specified to have ranges of retardance greater than or equal to $2\pi$. With this flexibility in the retardances that could be applied, the system could generate or analyse any polarisation state. An analysis on the use of such variable retardances will now be given, which proves that with such a system any polarisation state could be generated or analysed using these variable retardances.

We will now consider a pair of devices of retardances, $\delta_1$ and $\delta_2$, with their fast axes oriented at a relative angle of $45^\circ$. The Jones matrices and vectors used in this section can be found in [Goldstein 2011]. We have chosen this formalism in order to later include a term representing the absolute phase in the analysis.

The incident beam used in our analysis was polarised vertically, and thus its normalised Jones vector is:

$$j_{in} = \begin{bmatrix} 0 \\ 1 \end{bmatrix}. \quad (1.23)$$

This vertically polarised beam was then incident on a variable retarder with its fast axis at $45^\circ$, and then a similar variable retarder with its fast axis at $0^\circ$. The general Jones matrix representing a retarder with fast axis at angle $\theta$, and retardance $\delta$ is

$$J_R(\delta, \theta) = \begin{bmatrix} \cos^2 \theta + \exp(-i\delta) \sin^2 \theta & (1 - \exp(-i\delta)) \cos \theta \sin \theta \\ (1 - \exp(-i\delta)) \cos \theta \sin \theta & \exp(-i\delta) \cos^2 \theta + \sin^2 \theta \end{bmatrix}. \quad (1.24)$$

This can be calculated using the Jones matrices for a retarder and the two
corresponding rotation matrices, in a similar way to the Mueller matrix computation in Eq. 1.13.

\[
J_R(\delta, \theta) = J_{rot}(\theta) \cdot J_{Ret}(\delta) \cdot J_{rot}(-\theta),
\]

(1.25)

where

\[
J_{rot}(\theta) = \begin{bmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{bmatrix},
\]

(1.26)

and

\[
J_{Ret}(\delta) = \begin{bmatrix} 1 & 0 \\ 0 & \exp(-i\delta) \end{bmatrix}.
\]

(1.27)

From Eq. (1.24) we calculate the Jones matrix representing the first retar-
1.5. Spatial polarisation control

dation in the analysis, which has retardance $\delta_1$ and its fast axis at $45^\circ$:

$$J_{R}(\delta_1, 45^\circ) = \frac{1}{2} \begin{bmatrix} 1 + \exp(-i\delta_1) & 1 - \exp(-i\delta_1) \\ 1 - \exp(-i\delta_1) & 1 + \exp(-i\delta_1) \end{bmatrix}. \quad (1.28)$$

Similarly we can calculate the Jones matrix for the second retarder at an angle of $0^\circ$ and with retardance $\delta_2$:

$$J_{R}(\delta_2, 0^\circ) = \begin{bmatrix} 1 & 0 \\ 0 & \exp(-i\delta_2) \end{bmatrix}. \quad (1.29)$$

The Jones vector representing the polarisation state of the beam after experiencing the two retardances, $j_{\text{out}}$, is calculated by multiplying $J_{R}(\delta_1, 45^\circ)$ and $J_{R}(\delta_2, 0^\circ)$ by $j_{\text{in}}$ as follows

$$j_{\text{out}} = J_{R}(\delta_2, 0^\circ) \cdot J_{R}(\delta_1, 45^\circ) \cdot j_{\text{in}}. \quad (1.30)$$

$j_{\text{out}}$ is, therefore,

$$j_{\text{out}} = \frac{1}{2} \begin{bmatrix} 1 & 0 \\ 0 & \exp(-i\delta_2) \end{bmatrix} \cdot \begin{bmatrix} 1 + \exp(-i\delta_1) & 1 - \exp(-i\delta_1) \\ 1 - \exp(-i\delta_1) & 1 + \exp(-i\delta_1) \end{bmatrix} \cdot \begin{bmatrix} 0 \\ 1 \end{bmatrix},$$

$$j_{\text{out}} = \frac{1}{2} \begin{bmatrix} 1 - \exp(-i\delta_1) \\ (1 + \exp(-i\delta_1)) \exp(-i\delta_2) \end{bmatrix}. \quad (1.31)$$

Note that the horizontal component of $j_{\text{out}}$ depends only on $\delta_1$, and its magnitude can vary continuously between 0 ($\delta_1 = 0$) and 1 ($\delta_1 = \pm\pi$). Likewise, the magnitude of the vertical component depends solely on $\delta_1$, through factor $(1 + \exp(-i\delta_1))$. The behavior of this magnitude is of opposite trend to the magnitude of the horizontal component. This shows how the first variable retarder defines how much energy is split between the horizontal and vertical polarisation components. Finally, the second variable retardance $\delta_2$ allows us to fine tune the relative phase between the two orthogonal components to define the handedness and final ellipticity of the beam (see Fig. 1.4).

For simplicity, we have presented the analysis of how to generate truly arbitrary states of polarisation using variable retarders on a single pixel. The advantage of using spatial light modulators (e.g. parallel aligned LCoS) is that
δ₁ and δ₂ can both vary spatially over the pupil. Therefore, they can be written as δ₁(x, y) and δ₂(x, y) (in Cartesian coordinates) or δ₁(ρ, φ) and δ₂(ρ, φ) (in polar coordinates). With such generalisation of Eq. (1.31) the polarisation state can be fully and arbitrarily controlled across the initial beam, as will be discussed in later chapters.

The analysis presented in this section shows that two linearly independent retardances can be used to convert a fixed input polarisation state into any arbitrary polarisation state. In order to build a system capable of converting an arbitrary input polarisation state to any output polarisation, three retardances can be used. This was shown by [Zhuang 1999].
Background: optical microscopy

A microscope is defined as any instrument that magnifies the image of small objects in order to view them in more detail. An optical microscope is such an instrument that uses visible light incident on the sample, and a system of lenses to magnify the image.

The experimental system built for this thesis was in essence a high resolution microscope combined with a Mueller matrix polarimeter. The eventual objective of this research is to develop the system presented here further, such that a novel method of high resolution microscope can be attained. This chapter provides some background information on optical microscopy in general, in particular the principles that govern the resolution of an optical microscope. Also, some techniques that have shown themselves to exhibit better resolution than the classical resolution limit of an optical microscope will then be described, as well as a brief overview of the principles behind the experimental system built as a part of this thesis, which also seeks resolution below this limit.

2.1 The diffraction limit

The resolution of an imaging system is usually limited by Rayleigh criterion [Born 1999]. This theory states that the maximum resolvable feature of a microscope using incoherent illumination, $d_{\text{min}}$, is estimated to be

$$d_{\text{min}} = 1.22\frac{\lambda f}{d},$$

(2.1)

where $\lambda$ is the wavelength of the illumination, $f$ is the focal length of the imaging system, and $d$ is the diameter of the limiting aperture. This focusing system is illustrated in Fig. 2.1.
The classical diffraction limit is derived using scalar diffraction theory to calculate the irradiance of a circular aperture focused using a lens. The irradiance pattern at the focus takes the shape of an Airy disk when the aperture is circular in shape [Born 1999]. The Airy disk itself is made up of a central rotationally symmetric peak intensity, surrounded by concentric rings, the brightness of which decreases with increasing radius. The first minimum of the pattern occurs at a radius of $1.22 \frac{\lambda f}{d}$ from the central peak. For a diffraction limited system, two objects can be resolved when the first minimum of one Airy pattern coincides with the maximum of the adjacent Airy pattern [Born 1999]. This separation of the patterns is shown in Fig. 2.1.

The irradiance at the focus of a lens can be accurately predicted using a Fourier transform in place of the scalar diffraction integral. The Airy pattern is therefore equivalent to the Fourier transform of the circular aperture. Calculation of this transform shows that the spacing of the rings in the Airy pattern are represented by a $J_1$ Bessel function of the first kind.

The quantity $f/d$ is related to the numerical aperture (NA) of the system, $n \sin \alpha$. The angle of the semi-aperture, $\alpha$, is limited to 90°; therefore, in most high-resolution microscopes, the NA is usually approximately equal to 1. Because of this, $d_{\text{min}}$ in Eq. 2.1 is often said to be approximately equal to
2.1. The diffraction limit

The diffraction limit is determined by the wavelength of light and the aperture of the lens. The numerical aperture (NA) of a lens is given by the equation: $n \sin \alpha$, where $n$ is the ambient refractive index, and $\alpha$ is the semi-angle of the aperture. This is the angle that the axial marginal ray makes when it crosses the optical axis.

As the NA also depends on the ambient refractive index, $n$, it can be extended up to 1.4 by immersing the lens in a liquid of refractive index greater than air, such as water or oil. This increase in NA results in a smaller focal spot, which gives rise to sharper resolution than can be achieved with focusing with a lens in air.

Rayleigh’s theory describes the resolution of a focusing system best when the NA is low. This is because at higher numerical apertures the polarisation state in the pupil of the lens must be considered [Richards 1959]; Eq. 2.1 contains no terms related to polarisation as it is based on geometrical optics. In 1959 Richards & Wolf calculated that when linearly polarised light is focused...
2.2 High-resolution microscopy

using a high-NA focusing system, the irradiance at the focus will be elongated in the same direction as the incident polarisation. This is solely an effect of the polarisation distribution in the pupil of the lens. Fig. 2.1(a) shows the total irradiance at the focal plane of a 0.95 NA lens, where the incident beam is polarised in the \( x \)-direction. Profiles along the \( x \)- and \( y \)-axes are shown in Fig. 2.1(b) and (c) respectively. These results were calculated using algorithms developed by Oscar Rodríguez as part of his Ph. D work. Fig. 2.3 illustrates that vectorial focusing can result in a loss of circular symmetry in the focal field, depending on the polarisation distribution in the pupil of the lens. This loss of symmetry is solely a high-NA effect, where rotational symmetry depends on the the preservation of the same symmetry in the polarisation distribution in the entrance pupil. After focusing, a proportion of the field will be polarised parallel to the optical axis; this also contributes to the breakdown of circular symmetry in the focal region. Methods for modelling this type of focusing will be discussed further in Chapter 3.

2.2 High-resolution microscopy

Advances in nano-technology and cell biology have increased demand for higher resolution microscopy techniques that can be pushed as far as the diffraction limit and beyond. There are many challenges to high-resolution imaging of biological samples in particular, where not only a 2D image of a sample may be sought, but depth resolution may be required to see the inner workings of a cell, which may have a thickness on the order of microns.

Confocal microscopy is one technique that can provide high three-dimensional resolution [Wilson 1984]. In this technique, the sample is illuminated by point source, and a pinhole is placed at a plane conjugate to this point source that will block any out-of-focus light from other depths in the sample.

Near-field scanning optical microscopy (NSOM) is concerned with harnessing and amplifying the evanescent near-field of a specimen after it has been illuminated. Usually a probe whose tip is much smaller than the wavelength of the light is scanned over the surface of a specimen; the sample can be illuminated by passing light through the probe itself. A review article that provides
further information on this technique is [Hecht 2000]. This technique is useful for the characterisation of surfaces, however, little depth penetration can be attained due to the evanescence of the near field over a very short distance.

Many other high resolution microscopy techniques have been invented in recent years that make use of fluorescent dyes in samples. These dyes can be either excited or depleted by an incident photon. A combination of these can be used to obtain super-resolution images of specimens, usually of biological origin. Many of these are based on the widely used techniques of wide-field and confocal microscopy, in conjunction with fluorescent additives to the specimen. These methods have been successful in obtaining resolution as low as 10 nm in the case of Stimulated Emission and Depletion microscopy (STED).

2.2.1 Stochastic methods

A number of stochastic methods that work on a similar physical principle have been developed in recent years. These differ only in the type of dye used, which depends on the specimen under consideration. Three early methods using this approach are STORM (Stochastic Optical Reconstruction Microscopy) [Rust 2006], PALM (Photoactivated Localisation Microscopy) [Betzig 2006] and FPALM (Fluorescence Photoactivation Localisation Microscopy) [Hess 2006]. The references cited here provide further information than the overview provided in this thesis about these techniques to the interested reader.

These methods are usually similar in configuration to a wide-field microscope. A fluorescent dye is added to a sample. The dye attaches itself to a particular substance or feature wherever it is present in a sample. The dye molecules can then be activated and deactivated using a pulsed laser of the appropriate wavelength. The molecules of the dye will therefore blink, resulting in a transient Gaussian shaped emission region in the sample. The position of the particular feature to be found can be localised to the centroid of the excited area, with small margins of error. As long as the blinking fluorophores are sufficiently well separated, the dye molecule can be located with a high level of accuracy.

Different dyes will correspond to different molecules, and, if the same sample is scanned a number of times using a number of dyes and excitation wave-
2.2. High-resolution microscopy

lengths, a map of various features corresponding to particular molecules of the sample can be built up.

The resolution of these stochastic methods will depend on how accurately the centroids of the detected distributions can be calculated. Localisation accuracy will be degraded by any aberrations in the system; this limits the depth resolution of these techniques. This resolution has recently been reported to be on the order of 10 nm [Juette 2008, Pavani 2009, Huang 2008].

2.2.2 STED

Another fluorescence technique that has become widely used is STED microscopy. This method was first developed by Stefan Hell, and uses a degree of focal field shaping by modulation of the polarisation and phase in the entrance pupil of an objective lens. This technique uses two confocal paths incident on a sample, one of which is diffraction limited while the irradiance of the other takes a doughnut shape. A doughnut-shaped focus can be generated by applying a vortex in absolute phase to the distribution in the entrance pupil, which should also be circularly polarised. Other combinations of polarisation and phase are described by Khonina [Khonina 2012].

STED is a two step process. The first step involves excitation of the sample using the diffraction limited confocal beam, resulting in a Gaussian shaped excitation distribution, the size of which will be limited by the numerical aperture of the focusing objective lens. The second step, which provides the advantage in resolution over a conventional confocal microscope, is to use a doughnut-shaped depletion pulse, which deactivates the edge area of the Gaussian distribution, leaving a much sharper point of light than the original Gaussian excitation area, resulting in improved resolution. The original concept of this technique is described in [Hell 1994]. Lateral resolution using this technique depends on the quality of the zero in the centre of the doughnut-shaped irradiance distribution, as well as the size of the region in the centre of the doughnut.

Many polarisation and phase distributions used in the stimulation and depletion stages of STED, and a comparison of these can be found in [Khonina 2012]. The doughnut shaped beam can be generated, for example, by focusing either an azimuthally polarised cylindrical vector beam,
2.2. High-resolution microscopy

or a circularly polarised beam with a vortex of charge 1 in its absolute phase. Resolution of up to 10 nm has been achieved using this technique [Westphal 2005, Rittweger 2009]. Recently this technique has been used to localise defects in diamond to a resolution of 2.4 nm [Wildanger 2012].

2.2.3 Multiphoton microscopy

The third technique to be described in this section is two-photon/multiphoton microscopy. An advantage of this technique over other fluorescence methods is the use of infrared light instead of visible for excitation of the fluorophore. Scattering is reduced at this wavelength and this means that it is possible to probe depths of over a millimeter [Kobat 2011]. In this method, a pulse of infrared light is focused onto a sample, and a number of the photons from the pulse can combine their energy and cause a fluorescent molecule, which is usually activated by a visible photon, to move from its ground to excited state. This visible light can then be detected and the particular molecule to which the dye was attached can be localised and imaged [Denk 1990].

Due to the depths at which multiphoton microscopy can operate, aberrations can become significant due to the thickness of the sample. Adaptive optics can be employed in order to compensate for this [Débarre 2009]. The resolution of multiphoton microscopy arises from the small probability that a number of photons will combine to excite the fluorophore, and this will only occur inside the tiny focal volume. Therefore there is little stray light from out of focus sources.

Multiphoton, and particularly two photon microscopy has many novels applications, such as analysis of the neurons of animals subject to certain stimuli [Ahrens 2012], and also embryonic development [Jesacher 2009]. Some animals, such as the common fruit fly, can be genetically modified so that their neurons can be activated using a two-photon microscope. This field is known as Optogenetics [Miesenböck 2011]. In this field individual neurons can be activated by using light to generate the action potential required for the neuron to fire, and analysing the induced behaviour.

It must be noted that most fluorescent techniques require a dye of some kind to be added to the sample. Some of these dyes are cytotoxic and kill the cell under consideration. A recent advance, however, uses fluorescence to im-
2.3 Far-field vectorial polarimetry

Vectorial polarimetry is a technique that uses the vectorial nature of light to obtain sub-resolution information about nano-scale samples. The concepts that are used in this technique are illustrated in Fig. 2.3. This figure shows the combination of the techniques of polarimetry with microscopy in the experimental system.

Figure 2.4: Concept of vectorial polarimetry system. High-NA focusing is combined with a Mueller matrix polarimeter. An arbitrary phase and polarisation distribution can be made using the polarisation state generator (PSG), while any polarisation state can be analysed by the PSA. These two parts of the system use spatial light modulators (SLMs) and liquid crystal variable retarders (LCVRs) respectively.

The vectorial polarimeter includes the constituent elements of a Mueller matrix polarimetry (i.e., a PSA and PSG. See Section 1.4.1), and is capable of this type of measurement. With this, it includes a high-NA objective lens whose focal field will depend on the particular polarisation and phase distribution in the pupil of the lens. This concept is based on the previous successful work by Rodríguez [Rodríguez-Herrera 2009], which used Pockels cells to generate homogeneous polarisation states. As part of his work, he achieved detection of sub-resolution displacements of a nanosphere [Rodríguez-Herrera 2010]. The system described in this thesis is an
2.3. Far-field vectorial polarimetry

evolution of Rodríguez’ original system, with the addition of spatial control over both phase and polarisation distributions in the entrance pupil of the objective lens.

The novel aspect of this setup lies in its PSG, which includes two spatial light modulators used in a three pass setup. One of these passes controls the absolute phase distribution, while the other two can generate any polarisation state at any point across the beam [Kenny 2012]. The mathematical explanation of this control will be explained in Section 1.5.1. Using this property, this thesis attempts to use these inhomogeneous polarisation and phase distributions to shape the focal field of a high-NA lens in three dimensions, and to use these fields to obtain further sub-resolution information about nano-scale scatterers.
In focusing, a lens refracts its incident wavefront onto a spherical wavefront, which converges towards the geometrical focus. The field at the focus of a lens with low-NA can be accurately described using scalar diffraction theory, which integrates over the field in the aperture in the lens. The amplitude modulation and aberration function in pupil are considered in calculating this field. It is not necessary to include any terms representing the polarisation of the light in the pupil.

The calculation will be different at higher NAs, where the spherical wavefront after refraction by the lens has a significant curvature. It is this refraction that results in a change of polarisation in the focal field; a component of the field can even become polarised parallel to the optical axis. This was first realised by Richards & Wolf, who described this calculation using the Debye-Wolf diffraction integrals [Richards 1959]. At low-NA the curvature that the wavefront experiences when refracted onto a spherical surface is not significant enough to substantially affect the polarisation.

Fourier transforms were later discovered to be useful for carrying out the calculation at high-NA, with the publication of McCutchen’s method [McCutchen 2002]. An advantage to using a Fourier transform based method is faster computation time than directly integrating over the aperture, and flexibility in defining the field in the entrance pupil of the system.

The experimental system built for this thesis could generate arbitrary polarisation and phase distributions in the entrance pupil of a high-NA lens. Therefore maximal flexibility was required in the modelling of the focal field, and a Fourier transform based method was most suitable. However, before using this method, an investigation was carried out into whether the results obtained from a Fourier transform based method were significantly different from those obtained using Debye-Wolf integration.

In this Chapter we provide a description of the focusing system to be
modelled, further details on each of the methods to be compared, and a comparison of results obtained using similar input fields. Later in this chapter, some examples of focal fields that can be generated using the experimental system are provided, which show some of the capabilities of such a system for three-dimensional focal field shaping.

3.1 High-NA focusing

High-NA focusing requires the use of vectorial diffraction theory, in place of scalar, to accurately predict the focal field. Vectorial diffraction takes the direction of the electric field in the pupil into account when calculating the focal field, which equates to considering the polarisation distribution in the pupil of the lens, and can accurately predict the changes in polarisation that can occur with such tight focusing.

![High-NA focusing system](image)

Figure 3.1: High-NA focusing system. A planar wavefront is mapped onto a spherical cap by the focusing system. As the curvature of this spherical wavefront becomes significant, a larger proportion of the focal field will be polarised parallel to the optical axis, represented by $E_z$ in this diagram.

The focusing system under consideration for this comparison is shown in Figure 3.1. As is shown in the figure, before focusing, the field can be resolved into $x$- and $y$- components, with no component in the $z$-direction, which is parallel to the optical axis and direction of propagation. After focusing, the field converges towards the geometrical focus, and the polarisation state is rotated by an amount that depends on the numerical aperture of the system. The
numerical aperture is, in turn, dependent on the angle of the semi-aperture, \( \alpha \) by the following relation:

\[
NA = n \sin \alpha,
\]

(3.1)

where \( NA \) is the numerical aperture and \( n \) is the refractive index of the ambient medium. It is clear from this figure that as \( \alpha \) increases, the rotation of the polarisation state in the entrance pupil of the focusing system will increase. Figure 3.1 also shows that a field can be refracted substantially enough that a component will be parallel to the optical axis. This axial component was found to be especially prominent when a radial polarisation distribution is present in the entrance pupil [Dorn 2003]. To further clarify this statement the focused field using a radial polarisation distribution as input is shown in Fig. 3.2. This was calculated using a Fourier transform based method which will be described in a following section. As can be seen from this figure, the axial component is larger in magnitude than the \( x \)- and \( y \)-components.

### 3.1.1 Debye-Wolf integration

The integration carried out for the method of Debye-Wolf integration amounts to the coherent superposition of plane waves, each related to a ray originating from every point in the exit pupil of the high-NA system, and directed towards the focal point. Integration of these sources of plane waves calculates the focal field, as is apparent in the following equations for both the electric and magnetic field:

\[
E(p) = \frac{-ik}{2\pi} \iint_{\Omega} \frac{a(s_x, s_y)}{s_z} \exp(ik[\Phi(s_x, s_y) + \hat{s} \cdot \mathbf{r}_p])ds_x ds_y,
\]

\[
H(p) = \frac{-ik}{2\pi} \iint_{\Omega} \frac{b(s_x, s_y)}{s_z} \exp(ik[\Phi(s_x, s_y) + \hat{s} \cdot \mathbf{r}_p])ds_x ds_y.
\]

(3.2)

\( E \) and \( H \) represent the focused electric and magnetic fields, respectively, and these depend on position, \( p \), in the focal region. A weighting is applied to each of the sources of plane waves, which depends on their position in the pupil: \( a \) and \( b \) for the electric and magnetic fields respectively. These are termed the strength factors. \( s \) is a vector defining any of the rays extending from the exit.
3.1. High-NA focusing

Figure 3.2: The magnitude and phase of the focal field components produced by focusing radially polarised pupil distribution. Note that the $z$-component of the field has a larger magnitude compared to the field in $x$ and $y$. Phase distributions are shown in units of $\pi$ radians, while the irradiance distributions have been normalised to the maximum irradiance in the focal plane.

pupil to the focus, while $\Phi$ is the aberration function in the pupil. $\Omega$ is the solid angle subtended by the exit pupil with respect to the geometrical focus, $\mathbf{r}_p$ is a vector extending from the origin to point $p$, as is shown in Figure 3.1. Finally, $k$ is the wavenumber, which is equivalent to $2\pi/\lambda$.

This method can be used to accurately calculate the focal field for incident light of any homogeneous polarisation state. Its use has also been extended to the focusing of beams with inhomogeneous polarisation states such as radial and azimuthal [Youngworth 2000]. It is non-trivial, however, to extend its use to the focusing of any arbitrary polarisation and phase distribution. In this respect, Fourier transform based methods have a distinct advantage.

An algorithm that implements this method of modelling the focal field was written by Rodríguez [Rodríguez-Herrera 2009] as part of his thesis, making use of some solutions from [Török 2006]. This was provided with permission for use in the comparison of methods to be carried out as part of this thesis.
3.1. High-NA focusing

3.1.2 Fourier transform based methods

McCutchen first proposed a three-dimensional Fourier transform based method to describe field at the focus of a lens [McCutchen 1964]. This method takes the field in the entrance pupil, which can include amplitude modulation as well as any aberrations present in the system, and projects them onto the so-called "generalised aperture". This is equivalent to a spherical wavefront, the curvature of which depends on the angle of the semi-aperture and the focal length, which converges towards the geometrical focus. The spherical wavefront is contained within a three-dimensional volume, and a Fourier transform over this volume results in a focal volume that is also in three dimensions:

\[ U(P) = -\frac{i(2\pi)^{\frac{3}{2}}}{\lambda} F(R), \]  

where \( U \) is the complex amplitude of the field distribution in the focal region at point \( P \), \( \lambda \) is the wavelength, and \( F(R) \) is the three-dimensional Fourier transform of the generalised aperture at \( R \), a point in the focal region such that its distance from the geometrical focus is much less than the focal length of the system. A digital implementation of McCutchen’s method was carried out by Iglesias [Iglesias 2007].

McCutchen originally proposed that his method was valid only at low NA; however later revisions extended the validity to high NA [McCutchen 2002], stating that the method could be applied to each component of polarisation in the entrance pupil. Lin et al. later showed this explicitly by showing the equivalence of the generalised aperture with the strength factor used in Debye-Wolf integration (\( a \) in Eq. 3.2) [Lin 2012]. In this paper, the two-dimensional pupil polarisation state was projected onto the three-dimensional generalised aperture. Taking a Fourier transform of this volume gave results that agreed with those obtained using Debye-Wolf integration.

Leutenegger showed that if, instead of representing the spherical cap in a three-dimensional volume, a spherical phase is applied, the generalised aperture can be represented by a two-dimensional matrix [Leutenegger 2006]. The focal field can then be computed, plane-by-plane in the focal region, by taking a two-dimensional Fourier transform of this version of the generalised aper-
3.2 Comparison of methods

As part of the work carried out in [Lin 2012], it was desirable to investigate if digital implementation of the FT method resulted in any discrepancy between the two methods. To this end, a comparison between results obtained using both methods was carried out. Similar input polarisation states were given to both methods and the results from these were compared. Results from this comparison are presented in the following sections.

The comparison has been carried out for a number of example focal fields. One homogeneous pupil polarisation distribution state was used as well as an inhomogeneous polarisation distribution were chosen. These were both readily defined as inputs for both methods. The chosen polarisation distributions were linearly polarised in the $x$-direction and a radial polarisation distribution.

The input parameters for calculation of the focal fields were as follows. The NA used was 0.95, and the wavelength of the light was 532 nm. The pupil was circular with a diameter of 256 pixels. The focal volume was $128 \times 128 \times 128$, with a pixel size of 15.6 nm, equivalent to approximately 0.03$\lambda$. Each calculated focal field was normalised to the maximum total irradiance present in the focal volume.

The first comparison between the results involved plotting the profile along each axis of the three-dimensional focal volume, in both irradiance and phase. Plots of these profiles are shown in Fig. 3.3 (linear) and Fig. 3.4 (radial). In these figures, results from Debye-Wolf integration are shown as a solid black line, while results from the 3D-FT method are plotted as a blue ‘x’. As can be seen qualitatively from these figures, the methods give very similar results in irradiance and phase. Further analysis is needed to provide a more quantitative comparison.

As the difference between the results obtained is a small fraction of the total irradiance, any difference between the results is not obvious in Figs. 3.3 and 3.4. In order quantify if any discrepancy had arisen, the following metric, $\delta$, was calculated. This metric amounts to the magnitude of the difference between the complex amplitudes of the results obtained using each method.
3.2. Comparison of methods

\[
\delta = |E^{DW}(r_i) - E^{FT}(r_i)|, \quad (3.4)
\]

where $E^{DW}(r_i)$ is the complex amplitude in the focal region, calculated at position $r$. $E^{FT}(r_i)$ is a similar quantity calculated using the 3D-FT method.

\( \delta \) was calculated for values in the focal plane ($z = 0$), along both the \( x \)- and \( y \)-directions through the geometrical focus. In clearer terms, for the plot in the \( x \)-direction, both \( y \) and \( z \) were equal to zero. The focal field of a radial polarisation distribution is rotationally symmetric; therefore \( \delta \) had the same symmetry and was plotted along \( r \).

Plots of \( \delta \) are shown in Figs. 3.5 and 3.6. The maximum value for \( \delta \) in these plots is 0.010. The largest discrepancies in the plots of \( \delta \) occur when the total irradiance approaches zero.

A further metric, \( \varepsilon \), can be used to quantify the relative error between the
3.2. Comparison of methods

Due to the rotational symmetry of the focal field in this case, only \( r \) and \( z \) need to be considered. Results from Debye-Wolf integration are shown by the solid black line, while results obtained by a three-dimensional Fourier transform are shown with a blue ‘x’.

two methods. This was used previously in [Török 2006].

\[
\varepsilon = \frac{\sum_{i=1}^{N} |E^{DW}(r_i) - E^{FT}(r_i)|^2}{\sum_{i=1}^{N} |E^{DW}(r_i)|^2}.
\] (3.5)

All quantities are the same as they were in Eq. 1.25. Additionally, \( N \) is the total number of pixels in the focal volume. For the total focal volume, \( \varepsilon \) was calculated to be equal to \( 1.512 \times 10^{-3} \). This shows good overall agreement between the methods. It is also possible to calculate \( \varepsilon \) for each plane along the optical axis in the focal region, in order to investigate whether the relative
Figure 3.5: Plots of $\delta$ along the $y$-axis (left), and $x$-axis (right) for incident polarisation which was linearly polarised in the $x$-direction. $z = 0$ for both plots. Larger values for $\delta$ correspond to minima in the focal field.

error is dependent on defocus. In this case, $N$ is the total number of pixels per plane. Plots of $\varepsilon$ for our two input polarisation distributions are shown in Figs. 3.7 and 3.8. The relative error is lowest at the focus and increases with increasing defocus. The value for $\varepsilon$ on the order of $10^{-3}$ for all of the planes in our focal volume, which would amount to sufficient accuracy for most applications.

It is likely that the small errors that arise between Debye-Wolf integration and the 3D-FT method are due to computational precision in defining the continuous spherical surface on a three-dimensional grid. The direct equivalence between the Debye-Wolf integrals and a three-dimensional Fourier transform was also shown as a part of [Lin 2012]. The main difference in the implementation of these two methods in reality was therefore that errors could arise in the sampling of a spherical surface using a three-dimensional volume of cuboidal pixels. A two-dimensional analog was shown by Rodríguez, and this figure is reproduced in Fig. 3.9. This could be confirmed by re-running the comparison for a range of sampling levels of the three-dimensional volume.

3.3 Focal field shaping

In this section, McCutchen’s method is used to model the field at the focus of a high-NA lens using a number of different input polarisation and phase
3.3. Focal field shaping

Figure 3.6: A plot of $\delta$ along $r$ for incident polarisation radially polarised, and $z = 0$. Larger values for $\delta$ correspond either to minima in the focal field, or points of significant change in the derivative of the profile of the focal field.

distributions. These results were obtained as part of an investigation into shaping the focal field in three dimensions, not only to obtain a sharper focal spot, but to find focal field distributions that could be useful in the technique of vectorial polarimetry. The first distributions to be investigated have a flat phase distribution, and a spatially variant polarisation distribution in the pupil of the lens. After this, in Section 3.3.2, both polarisation and phase modulation are used to shape the focal field.

The calculations in the following sections used an NA of 0.95, and the wavelength of the incident light was 532 nm. The circular pupil was defined over a diameter of 64 pixels. The results obtained were contained in a three-dimensional volume of size $65 \times 65 \times 33$. The pixel size in $x$ and $y$ was approximately 34 nm, while the pixel size in $z$ was 52 nm. The size of the pixels in the $z$-direction was larger due increased sampling of the generalised aperture in the $z$-direction. Increasing sampling along this dimension means that the spherical wavefront could be more accurately represented (see Fig. 3.9). The algorithms used to implement McCutchen’s method were written in MATLAB by Dr. David Lara, and were provided with permission for use in this thesis work.
3.3. Focal field shaping

3.3.1 Polarisation-only control of the focal field

A simple particular case of a focal field that can be generated by controlling only the polarisation state in the pupil of a high-NA lens will now be shown. In this example, the focal field is split into two, spatially separated, perpendicularly polarised spots in the focal plane. This focal field can be produced by generating the polarisation distribution shown in Fig. 3.10 in the pupil of the lens.

To obtain the Stokes vector distribution in Fig. 3.10 a constant $\pi/2$ retardance was applied to the first SLM and five wavelengths of wrapped tilt, also in retardance, were applied to the second SLM. The irradiance of the focal field resulting from this polarisation state distribution is shown in Fig. 3.11, both theoretically calculated and from experimental detection after re-imaging the focus field on the CCD camera.

The two bright spots in Fig. 3.11 have perpendicular polarisations, and the separation between them depends on the magnitude and the direction of the retardance tilt applied to the second SLM. The spots show the typical elongation of the focal spot along the direction of linear polarisation when focusing at high NA [Richards 1959]. The theoretical components of the focused field at the focal plane are shown in in Fig. 3.12.

The experimental results in Fig. 3.11, and later Fig. 3.14, were obtained
3.3. Focal field shaping

Figure 3.8: Plot of the relative error, $\varepsilon$, against defocus or position along the optical axis. $\varepsilon$ was calculated plane-by-plane in the focal volume for incident light radially polarised.

by focusing the polarisation distribution depicted in Fig. 3.10, using a high-NA objective lens (OBJ in Fig. 5.1), onto a microscope slide. The objective used with our system was an Olympus UPLSAPO 100x, NA = 1.4 in $n = 1.518$ immersion oil. The focal field was reflected off the air-glass interface at the focus of the objective, and re-imaged at low-NA (approximately 0.02) onto the plane of detection of the auxiliary camera. This plane was conjugate to the focal plane of OBJ, and a high-magnification image of the focal plane could be obtained there. The separation of the focal spots was detectable at this plane as it is due to differences in the directions of propagation of perpendicular polarisation states, which is not a high-NA effect. The nominal magnification between the field at the tight focus and the image on camera D2 was on the order of 100×, therefore the separation between the measured spots was on the order of 100 $\mu$m, when the wavelength of the focused laser light was 0.532 $\mu$m.

We note that in this example the polarisation state at each position on the pupil is resolved into vertical and horizontal components, therefore they have equal magnitude at all points. The combination of retardances on both SLMs converts the field in the pupil into a coherent superposition of an $x$-polarised incident beam propagating parallel to the optical axis of the objective lens, and a $y$-polarised incident beam with direction of propagation at an angle
3.3. Focal field shaping

Figure 3.9: Sampling of a spherical surface with cubic voxels will lead to errors in the representation of the surface. Here, this is represented by attempting to represent the arc of a circle using rectangular pixels. Taken from [Rodríguez-Herrera 2009].

Figure 3.10: Stokes vector distribution of the polarisation state in the pupil of the high-NA lens which results in two perpendicularly polarised spots in the focal field. A uniform retardance of $\pi/2$ is used on the first SLM, while five wrapped waves of tilt in retardance are applied on the second. The incident beam is uniformly vertically polarised.

with respect to the optical axis. This angle depends on the number of waves of tilt in retardance and the direction of the tilt. This difference in the direction of propagation of the perpendicularly polarised incident fields results in the spatial separation of the perpendicularly polarised focused fields. This is a highly controllable superposition of orthogonally polarised spots with potential application as a dynamic optical probe by adjusting the retardance tilt applied to the SLMs.

The polarisation distribution in this example was constant along the $y$-axis of the pupil, and varied along the $x$-axis of the pupil (see Fig. 3.10). The polarisation modulation along the $x$-axis corresponds to 5 full rotations on the
3.3. Focal field shaping

Figure 3.11: Theoretical (a) and experimental (b) intensities for the split focal field. The right image was acquired using the auxiliary camera (D2 in Fig. 5.1), after re-imaging the field at low-NA. Both irradiance distributions in this figure were normalised to the maximum irradiance value.

Figure 3.12: Magnitude and phase of the three components of the electric field of the intensity distribution in Fig. 3.11(a). Phase distributions are shown in units of $\pi$ radians, while the irradiance distributions have been normalised to the maximum irradiance in the focal plane.
3.3. Focal field shaping

Figure 3.13: Polarisation state in the pupil of the high-NA lens which results in four perpendicularly polarised spots in the focal field. Ten wrapped waves of tilt in retardance are applied by each SLM. These tilts are perpendicular to each other.

Figure 3.14: Irradiance of the focused field, theoretically calculated using McCutchen’s method (a), and experimentally measurement after re-imaging the field at high–magnification (b). Both irradiance distributions in this figure were normalised to the maximum irradiance value.

Poincaré sphere, around the V-H axis, passing through linear at $+45^\circ$, right circular, linear at $-45^\circ$, left circular, and polarisations states in between.

In this first example, the retardance on the first SLM was kept constant, which could be performed with a homogeneous retarder. Strictly speaking only one pass on an SLMs is necessary to generate that distribution. The strong dependence of the field at the focus of a high-NA lens on the pupil polarisation state is evident, and another similar example will now be given, which uses the full capabilities of the spatial polarisation state generator.

In this further example, tilts in retardance are applied to both SLMs in perpendicular directions. The Stokes parameters of the resulting polarisation distribution are shown in Fig. 3.13. Focusing this polarisation distribution
Figure 3.15: Magnitude and phase of the components of the field in the focus using the polarisation state shown in Fig. 3.13. Phase distributions are shown in units of $\pi$ radians, while the irradiance distributions have been normalised to the maximum irradiance in the focal plane.

results in a field at the focus with four bright spots. The theoretical and experimentally measured intensity of the focused field are shown in Fig. 3.14. The magnitude and phase of the components of the electric field at the focus are shown in Fig. 3.15. Theoretical calculations were again obtained using McCutchen’s method with a numerical aperture of 0.95.

We note that the spot at the origin of Figs. 3.11 and 3.14, which corresponds to the geometrical focus, is brighter than the other spots in the field of view. This is due to the pattern on the SLM being wrapped several times across the pupil. At the wrap points, some of the light will remain unmodulated and unaffected by the SLM. This light is focused at the geometrical focus and contributes to the irradiance at the geometrical focus.
3.3. Focal field shaping

3.3.2 Using both polarisation and phase

Further capability for shaping the focal field in three dimensions can be attained when phase modulation, as well as polarisation modulation, is used to control the field in the entrance pupil of a lens. In this thesis, we consider pupil distributions that include a vortex of topological charge 1 in their absolute phase. It has been previously established that a radial polarisation distribution, which includes a vortex in its phase, in the pupil results in a strong axial component in the focal field [Dorn 2003].

![Figure 3.16: Retardance of the pupil polarisation distribution (a), defined using a \( Z_3^3 \) Zernike polynomial with amplitude \( \pi/2 \) (in the Born and Wolf normalisation [Born 1999]). A schematic diagram of the local coordinate systems and polarisation states is shown in (b).](image)

A cylindrical vector beam is essentially a realisation of a phase vortex, which is the result of a localised rotation of the coordinate system that follows the azimuthal angle of the polar cylindrical coordinate system. Azimuthal and radial polarisation distributions are two examples of cylindrical vector beams [Biss 2001]. Both azimuthal and radial polarisation distributions contain only linear polarisation states, the orientation of which changes with azimuthal angle. This means that the relative phase between orthogonal polarisations will be zero at any point across the beam. In this section we investigate the focusing of cylindrical vector beams with spatially varying relative phases; the relative phase, in this case, is defined using a Zernike polynomial.

It is possible to use a polar coordinate system, resolving the field into its radial and azimuthal components instead of \( x \)- and \( y \)-, to describe a cylindrical vector beam. In this case only two parameters, instead of three when using Cartesian coordinates, can be used to define the input. This is because a
3.3. Focal field shaping

Figure 3.17: The magnitude and phase of the focal field components shown in a polar coordinate system. Note the triangular symmetry of the $\vec{E}_p$ and $\vec{E}_s$ components, along with the relatively strong axial component shown in $\vec{E}_z$. Phase distributions are shown in units of $\pi$ radians, while the irradiance distributions have been normalised to the maximum irradiance in the focal plane.

The polar coordinate system intrinsically incorporates a phase vortex of topological charge 1 that is characteristic of cylindrical vector beams.

We now introduce a polarisation distribution that makes use of pure phase control and full polarisation control in terms of cylindrical beams. We use a polar coordinate system, decomposing the field into radial and azimuthal components, $\vec{E}_p$ and $\vec{E}_s$ respectively. We defined the distribution by making the retardance between the radial and azimuthal components a Zernike polynomial [Braat 2002].

In the areas of the pupil where the retardance is zero linearly polarised light is present, however the direction of polarisation changes with azimuthal position such that the radial and azimuthal components have equal magnitude everywhere. Moving towards the edge of the pupil, the retardance tends to increase in some regions (red and blue lobes), hence the ellipticity also increases. Note that this field has a singularity in the middle of the pupil (on
the optical axis of the propagating beam), typical of vortex beams.

The polarisation state in the pupil of the high-NA objective lens is shown in Fig. 3.16(b). The figure shows the field in small local polar coordinate systems, or in other words in radial and azimuthal components, $\vec{E}_p$ and $\vec{E}_s$, respectively. The relative phase difference (retardance) between $\vec{E}_p$ and $\vec{E}_s$, $\delta = \delta_p - \delta_s$, is a distribution given by Zernike polynomial $Z_3^3$. These pupil distributions were first devised by Dr. David Lara, in work related to this thesis, some of which was published as part of [Kenny 2012].

![Figure 3.18: Relative phases of the pupil distributions used to calculate the focal fields in Figs. 3.19 and 3.20, which are defined using Zernike polynomials, $Z_2^4$ (left) and $Z_5^5$ (right), in the Born normalisation. The amplitude of both Zernikes in this figure is $\pi$. These relative phases are shown in units of radians.](image)

The field that can be produced by focusing the polarisation distribution in Fig. 3.16 was calculated and the magnitude and phase of the components of this field are shown in Fig. 3.17. This is a recalculation of previous work done by Dr. David Lara, part of which was published in [Kenny 2012] A triangular shape with 3 lobes is apparent in both the radial and azimuthal components of this focal field. One of the key features of this field is that the state of polarisation varies rapidly through the focal region. For this reason we anticipate that this field could be useful for the technique of vectorial polarimetry to detect asymmetries in nano-scale samples, for example by rotating the triangular lobe-shaped focused field around an asymmetric sample. Possible applications of this technique would be in defect detection and optical storage. Other geometries of focal field can be generated using further Zernike patterns, producing many different symmetries of focal field. It is anticipated that this property could be used to increase to tailoring the focal field to the specimen under consideration, in order to obtain further sub-resolution...
Figure 3.19: The magnitude and phase of the focal field components produced by focusing a cylindrical vector beam with a relative phase defined by a $Z_4^2$. This results in a cross/square-shaped focal field. Phase distributions are shown in units of $\pi$ radians, while the irradiance distributions have been normalised to the maximum irradiance in the focal plane.

Zernike polynomials have a number of advantageous properties which make them useful for defining any properties such as relative or absolute phase across a pupil. Firstly they are defined on a circular pupil; most optical systems use a circular aperture stop and this is especially true of microscopy systems where the aperture is usually determined by the entrance pupil of the microscope objective lens. Zernike polynomials also comprise an ortho-normal set of functions. This property could be useful for further experiments in determining the response of a specimen to a subset of Zernike-defined pupil distributions. Because of this property, cross-talk between the responses could be minimal. Another advantageous property of the Zernikes, when used to define high-NA pupils, is that many of the polynomials have a derivative at the edges of the pupil. In reality, using any of these "lobed" patterns to define a pupil means modulation of the high-NA properties of the focusing system; rays from the edges of the pupil have a larger angle with respect to the optical
3.3. Focal field shaping

Figure 3.20: The magnitude and phase of the focal field components produced by focusing a cylindrical vector beam with a relative phase defined by a \( Z_5^5 \). This results in a pentagon-shaped focal field. Phase distributions are shown in units of \( \pi \) radians, while the irradiance distributions have been normalised to the maximum irradiance in the focal plane.

Other Zernike polynomials can be used to define the relative phase between \( \vec{E}_p \) and \( \vec{E}_s \). Two focused fields of pupil distributions similar to that shown in 3.16 are shown in Figs. 3.19 and 3.20. The pupil distributions for these focal fields are defined in a similar way to the distribution shown in Fig. 3.16. Instead they use the \( Z_4^2 \) and \( Z_5^5 \) to define the relative phase between the azimuthal and radial components. In this case the Zernike polynomials have an amplitude of \( \pi \). These relative phase distributions are shown in Fig. 3.18.

Comparing the pupil relative phase distributions with the focused fields, it is clear that symmetries in the pupil distribution can translate into the same type of symmetry in the focus, with three-, four- and five-fold symmetry being present in these figures. These focal fields could potentially also yield sub-diffraction limit information in the technique of vectorial polarimetry. It is also interesting to note that the axial component remains constant in Figs. 3.17, 3.19, and 3.20 regardless of the relative phase distribution. We
Figure 3.21: Irradiances in the focal plane using a variety of cylindrical vector beams. The top row shows an $x$-$y$ slice of the irradiance in the focal plane. The bottom row shows a line-plot through the focus in $x$. The left most column contains results obtained using a radial polarisation distribution in the pupil of the lens, while the input polarisation distributions for the remaining three columns are defined as described in Fig. 3.16, using Zernike polynomials $Z_3^3$, $Z_4^2$ and $Z_5^5$ to define the relative phase between the radial and azimuthal components of the field.

Note that the example pupil distributions presented here all have a vortex of topological charge one in their absolute phase. Further investigation into these types of focal fields could involve the use of input field distributions with higher topological charges.

As a further aid to the reader in understanding the effect of polarisation and phase control on the focal field, the irradiance of each of the focal fields shown in Figs. 3.17, 3.19 and 3.20 were calculated and are shown in Fig. 3.21. The irradiance at the focus using a radially polarised pupil polarisation distribution is also shown in this figure. Figure 3.21 also shows line-plots through the focus in the focal plane. It is clear from this figure that the irradiances distribution is also profoundly affected by the Zernike polynomial used to define the relative phase between the radial and azimuthal components of the field in the pupil.
The alignment of any optical system can always be challenging, however, when working with polarisation dependent optical elements, another dimension is added to the alignment of the optical system. Any polarisation dependent component must have its axis aligned azimuthally as well as being placed in the correct position in the setup. As well as this azimuthal alignment, the variable properties of voltage controlled elements have to be quantified in order to optimise their use in the system.

Prior to attempting any optical alignment of the components within the setup itself, these components of the system had to be first measured and calibrated independently. Discussion of the characterisation of many of the polarisation dependent optical elements is provided in this chapter. These included the polarisers and waveplates, which had to be rotated at the correct angle such that they operated as required, and the liquid crystal devices that were used, whose variable retardance, phase modulation, and spatial non-uniformity had to be quantified.

## 4.1 Polarisers & waveplates

A polariser can be crossed with respect to another well-calibrated known polariser to find its transmission axis. The axis of the unknown polariser is found by rotating it until a minimum in irradiance is observed; the axis is \( \pm 90^\circ \) away from this minimum point. These two crossed polarisers can then be used to characterise elements that introduce a retardance to the beam of light.

In the alignment of a waveplate the crossed polarisers remain fixed while the waveplate is placed between them and rotated azimuthally. When either
the fast or slow axis is aligned with the transmission axis of one of the polarisers, a minimum in irradiance will be observed after the second polariser. At this position the waveplate is not affecting the polarisation state of the beam after the first polariser, i.e., no relative phase is being generated.

It is useful to know the approximate position of the fast and slow axes, so that they can be easily differentiated; however, it is not necessary as simple experiments can be used to find this out. For example, a half waveplate can be used to rotate an incident linear polarisation; the direction of rotation will depend on the orientation of the fast and slow axes with respect to the incident polarisation state.

### 4.2 Liquid crystal variable retarders

The polarisation state analyser (PSA) used in the system makes use of two liquid crystal variable retarders (LCVRs) and a polariser. These were manufactured by Meadowlark (model no. LVR - 200) and operate in transmission. If the retarders are rotated at an angle of 45° with respect to each other, any polarisation state can be converted to be aligned with the polariser. This operates in a similar way to the complete polarisation control setup described in Section 1.5.1, except in reverse. The calibration of the LCVRs used in this thesis was adapted from the one that Lara [Lara 2005] used for the characterisation of Pockels cells.

#### 4.2.1 Azimuthal axis alignment

A method, similar to that described in Section 4.1, was used to align the fast axis of the variable retarders. Since the voltage applied to the retarder was computer-controlled, a periodically varying signal could be sent to the retarder, which resulted in a periodic response in the effective retardance of the element.

For the alignment, the retarder was placed between parallel polarisers, instead of crossed polarisers, as was the case previously. A periodically varying voltage was then applied to the retarder; at the same time, the retarder was manually rotated. A display of the measured irradiance was obtained using an oscilloscope and a photodetector. When either the fast or slow axes were
4.2 Liquid crystal variable retarders

aligned with the transmission axis of the polariser, a minimum in modulation in the signal on the oscilloscope was observed. Markings provided by the manufacturer on the retarders were helpful in differentiating between the fast and slow axes of the retarders. It was chosen to use parallel polarisers for this measurement so that a more substantial irradiance would reach the detector, and a higher signal to noise level would be attained.

The two retarders were aligned at $45^\circ$ and $0^\circ$ respectively. The $0^\circ$ axis was defined as the transmission axis of the polariser in front of the camera. The setup can convert any polarisation state such that it aligns with the transmission axis of the polariser. The polariser then isolates this component of the light, after which it is quantified using the CCD camera.

4.2.2 Retardance response

The retardance response of liquid crystal depends on the voltage applied to it. This voltage creates an electric field, which rotates the liquid crystal molecules depending on its magnitude. It is this rotation that gives rise to the variable retardance [Kahn 1972].

The active material in the LCVRs was intrinsically non-linear in its response to an applied voltage. This was shown by the calibration curves provided by the manufacturer. Its response was also wavelength dependent due to the dependence of the refractive index of liquid crystal on the wavelength of the incident light [Wu 1984]. It was necessary to find the voltages that corresponded to the appropriate retardances needed to convert the six base polarisation states to vertically polarised light. The same wavelength as was used in the final experimental system (532 nm) was used for this measurement. Characterisation information from the manufacturer was obtained using a wavelength of 633 nm; therefore this information could only be used as a guide to the retardance response to applied voltage. To this end, measurements of the retardance response of each variable retarder, using a wavelength of 532 nm.

A ramp voltage that increased in time was applied to a variable retarder, which was placed between crossed polarisers. The irradiance was measured using a photodetector connected to an oscilloscope. It was assumed during this experiment that no spatial non-uniformity arose in the response of the
Figure 4.1: Retardance response of LCVR 10314. The applied voltage is shown by the black broken line, while the detected irradiance is shown in blue. Points that correspond to retardances of 0, $\pi/2$, $\pi$ and $3\pi/2$ are marked in red.

Figure 4.2: Retardance response of LCVR 10315. The applied voltage is shown by the black broken line, while the detected irradiance is shown in blue. Points that correspond to retardances of 0, $\pi/2$, $\pi$ and $3\pi/2$ are marked in red.

retarder. This assumption was found to be appropriate when the calibration of the overall system was carried out. The method of calibration will be described in Chapter 6.

If the liquid crystal molecules were linear in their response to applied volt-
4.2. Liquid crystal variable retarders

age, the measured irradiance would vary sinusoidally in time. However, as the retarders were non-linear in their response, this was not the observation recorded. The range of the ramp voltages in Figs. 4.1 and 4.2 were chosen to be within the most linear region of response of the retarders—where the response most resembles a sine wave. The chosen range of the voltages also corresponded to approximately a full wave of retardance. The recorded irradiances for the two retarders are shown by the blue curves in Figs. 4.1 and 4.2, while the applied ramp voltages are shown in black.

As the azimuthal position of fast axis of the retarder was half way between the crossed polarisers, the irradiance at the detector could be directly related to the retardance being applied by the LCVR. At the minimum in irradiance, the polarisation of the light incident on the retarder was not changed, indicating that no relative phase was being introduced. Therefore the voltage that gave a minimum in irradiance also corresponds to a zero in retardance. Conversely, a maximum in irradiance corresponded to half a wave of retardance; the polarisation state was rotated by 90° so that it aligned with the transmission axis of the second polariser. At half the maximum irradiance, the retardance was either $\pi/2$ or $-\pi/2$. In the case of the LCVRs used in this work, it was known, from information provided by the manufacturers, that the retardance decreased with increasing voltage. From this it was possible to distinguish the two voltages that resulted in half the maximum irradiance being detected. The higher voltage that results in half the maximum in irradiance being attributed to $\pi/2$ in retardance, while the lower voltage that results in the same irradiance was attributed to $-\pi/2$.

Using this information the corresponding voltage for the retardances for the analysis of the required polarisation state could be found. The voltages were read from the plots shown in Figs. 4.1 and 4.2, and are summarised in Table 4.1. The points from which each voltage was read are marked in Figs 4.1 and 4.2 in red.

Table 4.1 also shows the corresponding polarisation state that can be analysed using pairs of retardances. These retardances were identified by solving the following equation, which models the PSA using Mueller algebra:

$$s_{out} = M_{ret}(0°, \delta_2) \cdot M_{ret}(45°, \delta_1) \cdot s_{PSA}.$$  (4.1)
4.3. Spatial light modulators

<table>
<thead>
<tr>
<th>Pol. state</th>
<th>δ₁ (rad)</th>
<th>Applied voltage (V)</th>
<th>δ₂ (rad)</th>
<th>Applied voltage (V)</th>
</tr>
</thead>
<tbody>
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<td>V</td>
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<td>2.135</td>
<td>0</td>
<td>2.167</td>
</tr>
<tr>
<td>H</td>
<td>0</td>
<td>2.135</td>
<td>π</td>
<td>1.511</td>
</tr>
<tr>
<td>-</td>
<td>3π/2</td>
<td>2.548</td>
<td>π/2</td>
<td>1.842</td>
</tr>
<tr>
<td>+</td>
<td>π/2</td>
<td>1.806</td>
<td>π/2</td>
<td>1.842</td>
</tr>
<tr>
<td>L</td>
<td>π</td>
<td>1.453</td>
<td>π/2</td>
<td>1.842</td>
</tr>
<tr>
<td>R</td>
<td>0</td>
<td>2.135</td>
<td>π/2</td>
<td>1.842</td>
</tr>
</tbody>
</table>

Table 4.1: Retardances that can be used to generate the corresponding polarisation state: δ₁ is the retardance of LCVR 10314. δ₂ is the retardance of LCVR 10315, which is rotated at 45° with respect to LCVR 10314 and the vertical polariser in front of the camera.

In Eq. 4.1, \( \mathbf{M}_{\text{ret}}(\theta, \delta) \) is the Mueller matrix of a retarder rotated at angle \( \theta \), with retardance \( \delta \). \( s_{\text{PSA}} \) is one of the analysis polarisation states that can be found in column 1 of Table 4.1. \( s_{\text{out}} \) is the Stokes vector of the beam transmitted through the system. This equation can be computed for varying values of \( \delta₁ \) and \( \delta₂ \) using each of the analysis polarisation states. When \( s_{\text{out}} \) is equivalent to the Stokes parameters for vertically polarised light (to match the transmission axis of the polariser in front of the detector), the corresponding values for \( \delta₁ \) and \( \delta₂ \) are suitable to be used in the PSA. These are the retardances listed in Table 4.1. This calculation can be carried out for each of the analysis polarisation states.

4.3 Spatial light modulators

A spatial light modulator (SLM) is a device that can control some attribute of a beam non-uniformly across its cross-section, by using a spatially programmable pattern. Spatial light modulators exist that are capable of controlling many attributes of the incident beam; for example, absolute phase can be controlled using a deformable mirror, and amplitude can be controlled using a pupil mask. In this work, we deal solely with electrically addressed nematic liquid crystal SLMs, in which the active property is the refractive index along the slow axis of the liquid crystal. Therefore, depending on the polarisation state of the incident beam and experimental setup, either the absolute phase, the polarisation state, or the amplitude of the beam can be programmed spatially. For modulation of the absolute phase alone, the inci-
4.3. Spatial light modulators

Figure 4.3: Diagram depicting the constituent layers of an SLM. The amount of phase modulation depends on applied voltage to a pixel, which rotates its adjacent liquid crystal molecules.

dent beam must be polarised parallel to the slow (active) axis of the SLM. For amplitude modulation, the SLM must be used in conjunction with a polariser with its transmission axis at ±45° with respect to fast axis of the SLM. Retardance modulation is carried out by polarising the incident beam at an angle between the fast and slow axes. The experimental setup, to be described later in Chapter 4, used both spatial phase and spatial retardance modulation to tailor the entrance pupil of a high-NA objective lens in both its polarisation and phase.

A description of the constituent parts of a nematic liquid crystal SLM is given by Figure 4.3. The SLMs used in this work were manufactured by Boulder Non-Linear Systems (XY Series P512-0532). A layer of liquid crystal molecules, which is protected by a cover glass, lies on top of a reflective dielectric layer, which is coated to be maximally (90–95%) reflective at 532 nm,
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with a 100% fill-factor. Below this is a pixelised $512 \times 512$ array of $15 \, \mu m^2$ electrodes, each of which can be addressed individually with a voltage between zero and five volts using a very-large-scale integration (VLSI) circuit. When a voltage close to 0 V is applied to a pixel on the SLM, the liquid crystal molecules are oriented parallel to the coverglass (see Fig. 4.3). If a higher voltage is applied to the pixel, the molecules rotate such that they are perpendicular to the coverglass. This rotation changes the refractive index of the slow axis of the liquid crystal layer, while the refractive index of the fast axis remains constant.

The variation in the birefringence of this liquid crystal with applied voltage is key to the modulation achieved. This variation is due to the rotation of the orientation of the liquid crystal molecules [Kahn 1972]. A retardation of the absolute phase is experienced by the incident beam after reflection off the device shown in Fig. 4.3. This occurs only for incident light that is polarised parallel to the slow axis of the liquid crystal layer. In order to use the SLMS optimally for retardance modulation, the incident light should be polarised at $\pm 45^\circ$ with respect to the fast axis.

Before the SLMs were used in the experimental setup, it was necessary to characterise their phase and retardance response, and to optimise their operation. With regard to absolute phase modulation, it was necessary to compensate for any surface deviation introduced by reflection off the device. As well as this, an investigation into the influence of a highly biased pixel on adjacent pixels with a lower applied voltage was made. Two SLMs were obtained to be used in the experimental setup. It was found that only one of these had the requisite phase and retardance range of $2\pi$ at a wavelength of 532 nm. Therefore this SLM was chosen to be the phase modulation device, and the phase measurements in the next sections were done using this SLM.

### 4.3.1 Flattening the absolute phase

If a flat wavefront is incident on the type of SLM being used in this work, surface deviations on the reflective backplane will introduce aberrations to the reflected wavefront. The aberrations introduced by these deviations must therefore be measured and accounted for before use in absolute phase control. Surface deviation can be measured using an interferometer, the data from
Figure 4.4: Mean (left) and standard deviation (right) of the flatness SLM 8921, when a uniform voltage pattern was applied to it. This was measured in nanometres using a Twyman–Green interferometer. A total of twenty-four measurements of the flatness were used to calculate the mean and standard deviation. The RMS flatness of this measurement is 0.160 waves. These measurements are plotted in units of nanometres.

which is inverted and converted to a pattern file understood by the spatial light modulator in question. The pattern must be an array of unsigned 8-bit integers.

The interferometer used was manufactured by Fisba (μphase 2), and used a 633 nm 5 mW HeNe laser. The beam was spatially filtered and expanded to a diameter of 10 mm, which was large enough to cover most of the SLM array.

In order to measure the flatness of the SLM, uniform voltages were applied to the SLM, and a number of measurements of the phase after reflection off the device were taken by the interferometer. The uniform voltages applied varied from 0 (0 V) to 255 (5 V) in discrete steps of 32 grey levels. Three measurements of the surface deviation were recorded for each grey level. This amounted to a total of 24 measurements of the flatness of the device. The mean and standard deviation of all the measurements were calculated and are shown in Fig. 4.4. The RMS of this measurement was calculated to be
0.160 waves, which is equivalent to approximately 101 nm as the wavelength of the light used in the measurement was 633 nm.

The mean surface deviation was spatially filtered in MATLAB, removing unwanted high spatial frequencies such as those arising from particles of dust on the optics. It was then inverted and converted into an 8-bit bitmap image that could be applied to the SLM in order to flatten it. This pattern is shown in Fig. 4.5. Six measurements of the flatness with this pattern applied to the SLM were taken using the interferometer, and the mean and standard deviation of this found. The mean and standard deviation of this measurement is shown in Fig. 4.6. The RMS of this measurement was calculated to be 0.002 waves.

The measured SLM was specified range of a full wave at 532 nm, and, since the measurement wavelength used in the interferometer was larger than this (633 nm), it was not possible to wrap the phase beyond one wave. As is shown in Fig. 4.5, in order to flatten the reflected wavefront, wrapping of the
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Figure 4.6: Mean (left) and standard deviation (right) of the flatness of SLM 8921, shown in nanometres. This was measured using a Twyman–Green interferometer. The voltage pattern in Fig. 4.5 was applied to the SLM. The RMS of this measurement was 0.002 waves. Artefacts from some specks of dust on some of the optical surfaces are present in this image. These were not accounted for in the flattening procedure.

phase was required in the corners of the pattern as the peak-to-valley of the surface deviation was larger than 532 nm. These areas show up as erroneously large phase measurements in the measurements after flattening (see Fig. 4.6).

4.3.2 Influence of adjacent pixels

Loktev [Loktev 2007] states that any spreading of the electric field to adjacent pixels is due to two factors. The first of these is the thickness of the dielectric layer lying in between the pixelised electrodes and the liquid crystal layer. The electric field due to the voltages spreads laterally across the array, affecting adjacent pixels of the liquid crystal layer. The second factor is the size of the electrode pixels with respect to the thickness of the liquid crystal layer. The manufacturers specifications do not list these two properties of the SLMs, so we must assume that both contribute to the influence function between pixels. These two properties dictate whether the SLM is analogous to either a piston-
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Figure 4.7: The quartered pattern, which was applied to the SLM, is shown on the right in units of unsigned 8-bit integers corresponding to the voltages applied to the SLM. The measured phase distribution from application of this pattern is shown on the left in nanometres. This measurement was obtained using a Twyman-Green interferometer.

type or modal-type phase modulator. An example of a modal-type modulator is a membrane deformable mirror, where biasing a single actuator will have a pronounced effect on its adjacent actuators. The opposite is the case for an element such as a piston type deformable mirror, where adjacent actuators have no interdependence.

This property was investigated as part of the characterisation of the SLMs, using phase measurements taken using the FISBA interferometer. As is shown in Fig. 4.3, the mirror behind the liquid crystal layer in the SLMs used for this work constitutes a dielectric layer, and spreading of the electric field may occur here.

The SLM pixels were of size 15 \( \mu \text{m}^2 \); thus to bias a single pixel and attempt to measure its phase influence on adjacent pixels would be beyond the resolution of the interferometer. A more straightforward measurement that can be carried out is to measure the edge-spread function. This is a technique used to characterise CCD arrays in order to measure an analogous effect be-
4.3. Spatial light modulators

Figure 4.8: Edge spreading over adjacent pixels for both the horizontal (x) and vertical (y) axes across the face of the SLM. The error bars correspond to one standard deviation. Lines 300 to 450 from the measurement in Fig. 4.7 were used to compute the mean and standard deviation, which are plotted here against the horizontal pixel position.

Between adjacent CCD pixels [Reichenbach 1991]. In this case a pattern that includes a step in phase of $\pi/2$ at 532 nm was applied to the SLM, and the response in phase is measured. An average over lines of pixels can be taken in order to estimate the edge-spread. This assumes that the spreading of the field through adjacent pixels is symmetric. In this case, the quarters pattern shown in Fig. 4.7 was applied to the SLM, which provided the required phase step to measure the edge spread function, and a measurement of the phase was made using the interferometer. This measurement is also shown in Fig. 4.7.

A plot of an estimate of the edge-spread function in both the x- and y-dimensions is shown in Fig. 4.8. This plot was obtained by averaging 150 rows in each direction of the measurement shown in Fig. 4.7. The error bars in Fig. 4.8 correspond to the standard deviation of this set of rows. In this plot, the number of pixels between 10% above the baseline and 10% below the peak of the edge-spread was 7 pixels.

It was also possible to obtain an estimate of the line-spread function of this SLM, by differentiation of the edge-spread shown in Fig. 4.8. Again this was calculated for both dimensions and a plot of the line-spreads is shown in Fig. 4.9. The value of the full width half maximum for this plot was approximately 6 pixels.
4.3. Spatial light modulators

4.3.3 Retardance response

In addition to the non-linearity in the response of the liquid crystal, the retardance typically decreases with applied voltage. The slope of the response increases with applied voltage. It is desirable to create a look up table which artificially linearises the response of the liquid crystal for ease in modelling the polarisation states that are generated from an applied pattern. In order to obtain this, measurements of the retardance response for voltages were taken for a selection of uniform voltages applied to the SLM, which varied from 0 to 255 in steps of 10 grey levels. The experimental setup for this measurement is shown in Fig. 4.10. In this setup the SLM is placed between crossed polarisers in a reflection configuration. The angles of incidence and reflection were kept as small as possible (< 5°) so as to minimise errors. Another difference between this setup and others used to characterise the LCVRs is that a 4f relay are used to image the SLM through the second polariser onto the camera. This was done so that diffraction from the edges of the SLM would not affect the irradiance detected, and also that voltage patterns could be applied to the SLM and their effect on the irradiance distribution viewed on the camera.

This measurement is similar to the retardance characterisation of the LCVRs, in that a sawtooth signal in time was applied to the SLM, which was placed between crossed polarisers. The resulting retardance can be ex-

Figure 4.9: Estimated line-spread function of SLM 8921, obtained by differentiating the edge spread function. The error bars correspond to one standard deviation of the distribution of values used to calculate the line-spread.
4.3. Spatial light modulators

Figure 4.10: Experiment to measure retardance response of SLM8921. The incident beam has previously been expanded, spatially filtered and collimated. The SLM is placed between polarisers, P1 and P2, that are crossed with respect to each other. L1 and L2 image the SLM pixel array onto the CCD camera. An iris was placed at the Fourier plane between L1 and L2 in order to remove any spurious back reflections off the SLM.

Interpolated from the measured irradiance and attributed the voltage at that point in time. In this case, the voltages applied to the SLMs were uniform patterns, varying from 0 to 255 in steps of 5 grey levels. It was assumed, from observing the irradiance distributions, that the retardance response was uniform across the pixel array. It was found from this measurement that SLM 8921 had a larger range of retardance than SLM 8917. This SLM had a full wave of retardance while SLM 8917 did not perform to specification and had half a wave of reliable retardance modulation, decreasing from $\pi$ to 0 with increasing voltage. The voltage corresponding to a zero in retardance was 210 grey levels. It was observed that application of higher voltages than this gave an unstable response, and it was decided that these voltages were too unreliable to be used in final experiments.

Plots of the retardance versus applied voltage for both SLMs are shown in Fig. 4.11. This figure shows the voltages that correspond to a retardance range of $\pi/2$ for SLM 8917 and $\pi$ for SLM 8921. These voltages were then used to generate a LUT for each SLM, which linearised their response to applied voltage.

256 grey levels are available to be applied to each pixel of the SLM and these were used to create the LUT. In creating the LUT, it was decided that 0 in voltage should correspond to $\pi$ in retardance as low voltages corresponded to this retardance for both SLMs. 127 corresponded to 0 in retardance, and 255 was equivalent to $-\pi$. Information from the manufacturer indicated that retardance decreased with applied voltage, as was the case with the LCVRs.
4.3. Spatial light modulators

Figure 4.11: Retardance response of SLM8921. The look-up-table linearised the retardance response of the SLM, resulting in the sinusoidal irradiance pattern when a spatial sawtooth pattern was applied to the device between crossed polarisers. An image of this is shown in Fig. 4.12.

SLM 8917 did not have stable voltages that could be used to obtain retardances less than zero, and so its LUT was left blank for grey levels greater than 127. Other grey levels were defined such that the retardance of the device increased linearly with applied voltage.

In order to test this LUT, a sawtooth pattern was applied to the SLM between crossed polarisers. The irradiance was measured, and if the LUT was successfully linearised and the range of retardance was a full wave, a sinusoidal pattern in the irradiance should be visible. This is shown in Fig. 4.12. As can be seen in this figure, parts of the SLM were observed to fall short of the requisite range of $2\pi$ in retardance, even though the manufacturer specified that a full wave in retardance was achievable. This is most likely due to variations in the thickness of the liquid crystal layer across the device. The mean of the sinusoidal pattern shown in Fig. 4.12 over columns 200 to 240 is shown in Fig. 4.13.

It must also be noted that after taking this image, it was noticed that a seemingly uneven illumination was used; decreasing from left to right across the image. However, the sinusoidal pattern is important in this image, rather than the DC component. An uneven DC component could be accounted for by
4.3. Spatial light modulators

Figure 4.12: Retardance response of SLM8921. The left image is a spatial sawtooth voltage pattern, shown in units of 8-bit integers, applied to the SLM between crossed polarisers. The imaged irradiance distribution after the second polariser is shown on the right.

Figure 4.13: Mean response of the SLM to applied voltage. A sinusoidal pattern in irradiance indicates that the response of the liquid crystal is linear and that the range is a full wave. The mean irradiance is shown by the black markers, while the grey lines show one standard deviation. The mean was calculated from columns 200 to 240 in the right image of Fig. 4.12.

applying a uniform pattern to the SLM and measuring the spatial variation in irradiance. This variation could then be subtracted from the final image. This illumination gradient could also be due to imperfect collimation of the incident
beam onto the SLM, resulting in a variation in the angle of incidence of the incident beam. This could result in a variation in the relative angle between the orientation of the liquid crystals and angle of incidence, amounting to the decrease in irradiance across the SLM.
Two iterations of the experimental systems were built as a part of this research, and a description of both of these systems will be given in this chapter. The main difference between these was that the second iteration included control over the absolute phase, as well as replacement of a component which had been causing unwanted diffraction on the pupil. The first system used two SLM passes in order to attain complete polarization control across the beam. This control was achieved in the same way as the analysis described in Section 1.5.1, using two linearly independent retardances that could vary spatially across the beam. The first system is described in Section 5.1.

The second system uses the same spatial polarization control as the first, with the addition of a third pass on the SLMs for control over the absolute phase. This pass occurs before either of the retardance passes are applied. The second system is described in Section 5.2.

5.1 Polarisation only control

A schematic diagram of the experimental system incorporating polarisation only control is shown in Fig. 5.1. The light source was a linearly polarized 532nm wavelength CW frequency-doubled ND:YAG laser (Melles Griot model 85-GCA-005-100). A calcite polarizer was used to attain high polarization purity and to define the vertical orientation of polarization on the optical bench. The beam was then expanded and spatially filtered. A half waveplate (HWP) with fast axis at 22.5° was used to rotate the polarization of the beam from vertical to 45° before the first SLM (SLM1). After the first retardance distribution was applied to the beam using the SLM1, the polarization state was rotated back −45° by the same waveplate after reflection. The net effect of the half waveplate is to virtually rotate the fast axis of the pixels in SLM1, without having to physically rotate the device. This allowed us to match
5.1. Polarisation only control

Figure 5.1: Schematic diagram of the setup built which can create any polarisation state at any point across a laser beam, limited by the pixel size of the spatial light modulator used.

the square geometry of the pixels of the two SLMs, while still applying two linearly independent retardances to the pupil distribution.

Lenses L1 and L2 and a mirror (M) were then used to optically conjugate the planes of both SLMs; the magnification of this relay was 1, and had to be accurate to within one pixel (15 µm), so that accurate mapping of pixels between the two SLMs was attained. The configuration resulted in a relative orientation of 45° between SLM1 and SLM2, which can be used to reach any point on the Poincaré sphere at any pixel across the width of the beam.

After SLM2, another 4f system (lenses L3 and L4) was used to image the desired polarization distribution onto the pupil of a high-NA lens (OBJ), passing through beam splitter BS. The field at the focus could be tailored dynamically by addressing the retardance distributions on the two SLMs by changing the polarisation distribution as described in Section 1.5.1. This was one of the key features of the system.

A sample (SMP) could be placed in the focal plane, which then interacted
with the field at the focus of OBJ. The light scattered from it was collected, re-collimated and imaged (using L5 and L6) onto a CCD camera (D1). The system included a spatial polarization state analyzer, which consisted of a pair of liquid crystal variable retarders (VR1 and VR2), again at an angle of 45° with respect to each other, a linear polarizer (P) and camera D1. These parts of the system constituted the vectorial polarimeter. The diagram also shows an auxiliary camera (D2) which was accessible by using a flip-in mirror FM. This camera was conjugate to the focal plane of the objective lens, via lens L7, and was useful for the initial positioning of the samples. This could be used in conjunction with an auxiliary laser to obtain a dark-field image of the focal plane of OBJ, which was useful for finding samples. This will be described further in Section 5.3.

Another feature of the experimental system was that the inclusion of the spatial polarization state analyzer (PSA) allowed for the system to be accurately calibrated at every position across the beam. Calibration was performed using the eigenvalue calibration method (ECM) [Compain 1999, De Martino 2004]; systematic errors in the illumination and the detection could therefore be removed separately. For example, beam-splitters such as BS in the system can introduce different polarization effects across the beam [Pezzaniti 1995]. This source, and other similar sources, of error can be accounted for using ECM, as will be demonstrated in Chapter 6

### 5.2 Polarisation and phase control

Recalling Eq. 1.31, which shows that two retardances, $\delta_1$ and $\delta_2$, can be used to change a fixed input polarization state into any other polarization state. These retardances can vary spatially across the waist of the beam, when, for example, spatial light modulators are used to control the retardances, as is the case in our system. A pure phase term $\phi(x, y)$ can also be introduced, which can also vary spatially across the beam. In this case, Eq. (1.31) becomes

$$j_{\text{out}}(x, y) = \frac{\exp(-i\phi(x, y))}{2} \left[ \frac{1 - \exp(-i\delta_1(x, y))}{(1 + \exp(-i\delta_1(x, y))) \exp(-i\delta_2(x, y))} \right]. \tag{5.1}$$
5.2. Polarisation and phase control

Figure 5.2: Modified setup which includes both absolute phase and complete polarization control spatially over the pupil distribution, using three passes on two SLMs.

The experimental setup that could be used to incorporate this absolute phase control, along with the two SLM passes for full polarization control is shown in Fig. 5.2. In this version of the setup, the phase control was performed before the polarization modulation, thus the half wave plate (HWP) was placed in front of the second SLM (SLM2). The incident beam on the first SLM was vertically polarized and aligned with the slow axis of the SLM. Since the slow axis was the active axis, this step modulated only the absolute phase of the beam according to the pattern applied to the part of the SLM on which the beam is incident. This is equivalent to how an SLM is operated in adaptive optics and wavefront control, which is a feature of the system that could be further exploited to compensate for sample or system induced aberrations. The subsequent two SLM passes acted as described in Fig. 5.1 where the half waveplate at 22.5° was placed between the second two passes to give rise to the relative angle of 45° between these two retardances. Mirrors M1 and M2 were used to direct the beam onto a separate part of the SLM, such that it didn’t overlap the section used for control of the absolute phase.
5.3 Dark-field detection of samples

It would be prohibitively time-consuming to use only the focal point at the focal plane of OBJ in the search and positioning of samples (SMP), taking perhaps weeks to locate a single sample. The objective concentrated the light into a tiny area and scanning with such a small spot would be inefficient at discovering the position of the desired sample. A way of increasing the area used to search for samples is to use an auxiliary dark-field microscope path. Therefore, an auxiliary arm was added to the setup, which allowed precise positioning of the nano-scale samples.

A nano-positioning (Piezosystem Jena NV 40/3 CLE) stage with an accuracy of 2 nm and range of 80 µm was used for the finer positioning of the samples. This stage was placed on some Thorlabs linear translation stages, which were used for the initial search and coarse positioning. Both stages controlled the position in three dimensions. The additions to the setup that allowed finding and positioning of the samples are shown in Fig. 5.3.

Figure 5.3: Auxiliary arm added to the setup, highlighted in red, which was used to find and position the samples. This gave a dark-field image of the specimen in the focal plane.
The auxiliary laser (Ls2) had a wavelength of 633 nm. It was incident on the sample, which placed on a microscope slide placed near the focal plane of OBJ, from the side. The green laser (Ls) could be blocked at this point. The focal region of OBJ was conjugate to the auxiliary camera, D2, by the addition of a mirror in a flip-in mount (FM), and another lens (L7). If a sample was present in the focal region, and the light from Ls2 is incident at the correct angle, scattering will take place off the sample. Because the laser is incident from the side, only scattered light will contribute to the image on D2. None of the light due to reflection off or absorption by the samples will be in the image viewed on D2. This is equivalent to the operation of a dark field microscope. The red coloured optical path in Fig. 5.3 shows the components used in the auxiliary dark-field detection arm.

![Edge of 100 nm grating](image)

**Figure 5.4**: Dark field image of the edge of a 100 nm grating. The bar marks in the bottom right of the image are a key to the position and pitch of the grating provided by the manufacturer of this sample.

Bright areas in the image indicated that a sample or nano-sphere may be present. An example of the kind of image that would be seen at this stage is shown in Fig. 5.4, which is the dark field image of the edge of a grating with 100 nm pitch. The next step was to position the sample accurately in
the centre of focal plane, so that it could interact with the focal field in the desired manner. Unblocking the green laser, the focal spot could be visible in the image captured by D2. The focal spot was then overlapped with the sample to be investigated, using both the three linear translation stages and the piezo-electric stage. Once this has been achieved, Ls2 was switched off, and the pupil distribution of the light reflected off the sample was viewed on D1. The nano-positioning stage was then used for fine positioning such that the pupil of the objective was in focus on the camera and the sample was centred in the focal region. An example of such a pupil distribution is shown in Fig. 5.5. In this figure the pupil distribution was obtained using 80 nm gold nano-spheres as a sample.

5.4 Finding the SLM pupils

In order to generate a desired polarisation and phase distribution in the entrance pupil of an objective lens, the correct retardance and phase patterns had to be applied at conjugate planes to the pupil. Lenses L1–L4 in Fig. 5.2 provided accurate 1:1 imaging between conjugate planes in order to ensure accurate mapping between the pixels in each plane. The pixels on each SLM on which each pass was incident for each SLM had to be identified.

The setup described in Fig. 5.2 used two passes on one of the SLMs — one for control over the absolute phase, and the other as the second retardance pass for polarisation control. The SLM array was of dimensions $512 \times 512$ pixels,
5.4. Finding the SLM pupils

with a pixel size of 15 \( \mu m \). The largest pupil that can be accommodated twice on a 512 \( \times \) 512 can be calculated trigonometrically, and has a diameter of 300 pixels. These pupils would be positioned diagonally with respect to each other on the array.

Finding the position of the first pass that controls the absolute phase distribution used a setup similar to Fig. 4.10, where the SLM was placed between crossed polarisers in a reflection configuration, and imaging optics were used to view the spatial irradiance distribution. In order to accomplish this within the final experimental system, the system in Fig. 5.2 was built up until L2, and then a polariser at 45\(^\circ\) was placed before the beam expander (BE). A camera was placed in the position of SLM2 with a polariser at \(-45^\circ\) in front of it. This setup was equivalent to that shown in Fig. 4.10, with the beam incident on the pixels of the SLM used for phase control.

Figure 5.6: Unsigned 8-bit integer pattern used to identify the positions of the pupils on each SLM.

A pattern was then applied to SLM1, which varied spatially across the pixel array. It was chosen to use the picture in Fig. 5.6, which was provided by the manufacturer and was in the required format of a 512 \( \times \) 512 array of unsigned 8-bit integers. The observed irradiance pattern on the camera was then recorded and compared to the original to find the position of this phase pass on the SLM.

The system was completed before the pupils for retardance modulation were found on the SLMs. Stokes measurements were carried out using the
5.4. Finding the SLM pupils

Figure 5.7: Positions of pupils on SLM8921 (left), and the single pupil on SLM8917 (right) with respect to Fig. 5.6

pattern in Fig. 5.6 applied first to SLM1, leaving the part of the SLM used for phase modulation and SLM2 blank. On viewing the Stokes parameters, the part of SLM1 used for retardance measurement could be determined. This was repeated for SLM2; however the retardance pattern on SLM2 had to be inverted so that it would match up the the pattern on SLM1. For these measurements the objective lens was removed, and a mirror placed in the equivalent position of the entrance pupil of the objective.

The size of the entrance pupil of the objective lens was 5.1 mm, and the magnification of lenses L5 and L6 was 1.5. Using this, the required diameter of each pupil on the SLM was calculated to be 3.4 mm, equivalent to 227 pixels on the SLMs. Using the positions of the centre of the pupils on each SLM, the required patterns to generate inhomogeneous phase and polarisation distributions can be generated. The positions of the pupils with respect to the picture in Fig. 5.6 are shown in Fig. 5.7.

An example image of the measurements used to find the SLM pupils is shown in Fig. 5.4. This was one of the images used to find the pupil on SLM 8917 for the first iteration of the experimental system, which used one pass on each of the SLMs. The light incident on the SLM in this case was polarised at 45°, while the analysed light at the camera was −45°. Uniform patterns of zero voltage were applied to the remaining two SLM passes while this measurement was recorded. The pattern visible in the camera could be
5.4. Finding the SLM pupils

Figure 5.8: Experimental image of SLM 8921, used to find the position of the second polarisation modulation pass on this SLM for the first iteration of the experimental system. This image provides an idea of the capability of the SLM in rendering patterns applied to it.

related to the corresponding section of applied image, and the precise pixels used on this pass for polarisation control across the pupil of the beam could be identified. This was repeated for the other passes on the SLM, and these measurements gave an idea of the ability to reproduce applied patterns in reality. This type of characterisation of the SLM response would be important if the system were to be used in fields such as metrology, where it is essential to know the sources of systematic errors that can arise in the system in order to obtain dependable results. The spatial sampling used to obtain this image was slightly below the Nyquist limit that would enable resolution of the individual pixels of the SLM; therefore some subtle Moiré patterns can be seen in this image.
Chapter 6

Calibration and Mueller matrix measurements

6.1 Calibration of setup

Before using any polarimeter for experimental measurements, the system must be calibrated in order to ensure accurate readings and to eliminate any static systematic errors. These errors arise from diattenuation, retardance or depolarisation effects that can be introduced by any of the optical elements in the system. The Fresnel equations for reflection and refraction are polarisation dependent [Born 1999]; therefore any of the optical elements used in the system can cause an effect on the polarisation state, for example non-polarising beamsplitters can introduce a quarter of a wave of retardance [Pezzaniti 1995].

The Eigenvalue Calibration Method (ECM) was chosen to calibrate the experimental system described in this thesis, which was first developed by Compain [Compain 1999]. It had been shown to work reliably for the predecessor to the system built for this thesis [Rodríguez-Herrera 2009]. Some advantages of this method include that it can be used on a pixel-by-pixel basis so that uniformity in the PSA and PSG can be investigated, and also that it requires no modelling of the optical components used. Instead it compiles the errors introduced by individual elements of the setup into two Mueller matrices. One of these matrices represents the polarisation effects introduced by the constituent elements of the PSG arm, while the other represents the polarisation effects introduced by the PSA arm of the system. If the system were aligned and all elements were characterised perfectly, the calibration matrices would be equivalent to two identity matrices. In reality, the calibration matrices reflect the effective operation of the polarisation state generator and analyser.
6.1. Calibration of setup

6.1.1 Eigenvalue Calibration Method

This calibration method centred on obtaining the two experimental matrices that represented the operation of the PSG and PSA arms of the setup. These are termed $W$ and $A$ respectively. A comprehensive explanation of ECM was given by de Martino [De Martino 2004], and a summary of the method will now be given. In the following analysis, the variables are named using the same system as in previous chapters, except that theoretical or ideal matrices will be referred to in upper case italics, instead of using bold type.

The measurement of a Mueller matrix using a polarimeter can be represented as follows:

$$B = A \cdot M \cdot W,$$

(6.1)

where $B$ is the measured Mueller matrix of the sample and $M$ is its theoretical matrix. $W$ and $A$ describe the effects of the PSG and PSA matrices, respectively. When $W$ and $A$ are known, the polarimeter is said to be calibrated, and the Mueller matrix of any element, $M$, can be extracted from a measurement using the inverses of the calibration matrices:

$$M = A^{-1} \cdot B \cdot W^{-1}.$$

(6.2)

Finding $W$ and $A$ involves the measurement of four calibration samples, free space, horizontal and vertical polarisers and a quarter waveplate at 30° ($B_0, B_1, B_2,$ and $B_3$). Since the Mueller matrix of free space is equivalent to the identity matrix, we can say that for $n = 0$:

$$B_0 = A \cdot W$$

(6.3)

and therefore

$$W = A^{-1}B_0.$$  

(6.4)

The next step in the calibration is to calculate the set of products $C_i$, for the remaining calibration samples ($i = 1, 2, 3$):

$$C_i = (B_0)^{-1} \cdot B_i.$$  

(6.5)
6.1. Calibration of setup

Substitution of $B_0$ and $B_i$ using Eqs. 6.3 and 6.2 yields

$$C_i = W^{-1} \cdot A^{-1} \cdot A \cdot M_i \cdot W$$  \hspace{1cm} (6.6)

$$C_i = W^{-1} \cdot M_i \cdot W$$  \hspace{1cm} (6.7)

The products $C_i$ have the same eigenvalues as their corresponding theoretical matrices, $M_i$, since the eigenvalues of a product of matrices do not depend on the order of the product. This means that properties of $M_i$ such as the retardance, angle of rotation and diattenuation can be extracted from the calculation of $C_i$.

Looking at Eq. 6.4, the problem of calibrating the system is reduced to finding either $W$ or $A$, since $B_0$ is a direct measurement. $W$ is the unique solution of the set of simultaneous equations [De Martino 2003]:

$$M_i \cdot W - W \cdot C_i = 0.$$  \hspace{1cm} (6.8)

Lara introduced a modification of ECM that is suitable for a polarimetry system where the sample is in reflective configuration. The Mueller matrix of the samples are each measured in double pass, and this is accounted for in the optimisation of $W$ and $A$ that follows, such that they are equivalent to the measured matrices for single pass ECM. Further description of this can be found in [Lara 2005, Lara 2006]. This algorithm was provided, with permission for use with this system, by Lara, and included the modifications to ECM described in the following section.

For the use of ECM in calibrating the system built for this thesis, it was chosen to calibrate the PSG and PSA matrices individually instead of reconstructing $W$ and $A$ before calibration. The ideal PSG and PSA matrices are constructed from the individual Stokes vectors used in the generation and analysis of polarisation states, in the same sequence as that used experimentally:
6.1. Calibration of setup

\[
PSA = \begin{pmatrix}
1 & -1 & 0 & 0 \\
1 & 1 & 0 & 0 \\
1 & 0 & -1 & 0 \\
1 & 0 & 1 & 0 \\
1 & 0 & 0 & -1 \\
1 & 0 & 0 & 1
\end{pmatrix}.
\]

\[
PSG = \begin{pmatrix}
1 & 1 & 1 & 1 & 1 & 1 \\
-1 & 1 & 0 & 0 & 0 & 0 \\
0 & 0 & -1 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 & -1 & 1
\end{pmatrix}.
\] (6.9)

This modification to the calibration method was made because thirty-six pairs of polarisation states were measured and could be used to reconstruct the Mueller matrix. It was useful to see how well individual polarisation states could be generated and analysed in order to troubleshoot problems more easily.

As the PSA and PSG matrices are of size 6×4 and 4×6 respectively, they are not invertible and their pseudo-inverses must be used in place of \( W^{-1} \) and \( A^{-1} \) when Eq. 6.2. Also, instead of calibrating the measured Mueller matrix, \( B \), a 6×6 matrix constructed from the measured intensities, Eq. 6.2 is therefore a product of matrices of size \((4 \times 6) \cdot (6 \times 6) \cdot (6 \times 4)\), the result of which is the \((4 \times 4)\) Mueller matrix.

The condition numbers of the PSA and PSG matrices used were not optimal for obtaining the best possible measurements. The most optimal polarisation states were found by [Tyo 2002], and were, in fact, a set of elliptical states. For the purposes of this thesis it was advantageous to use the familiar linear and circularly polarised states as outlined in Eqs. 6.9, as these aided the initial characterisation and system set-up. However, the use of the more optimal elliptical states in the PSA and PSG is an improvement that could be made to the calibration procedure for this system. The precise optimal states for a particular system can even be determined experimentally, by finding the PSA and PSG matrices that result in a measured Mueller matrix with maximal condition number.

The calibration of this system was done pixel-by-pixel in the pupil; uni-
formity was not assumed across the pupil. This had to be the case as the aim of the system was to measure spatial distributions of the Stokes parameters. In order to differentiate between valid and invalid pixels on the camera, the determinant of the $6 \times 6$ uncalibrated matrix corresponding to a single pixel was taken. A suitable threshold of the determinant was chosen such that all the pixels in the pupil of the lens would be computed. This amounted to a larger area than the pupil, as no aperture was placed in the position of the pupil for these measurements. If an aperture were introduced, the position of the sample could be less accurate and diffraction from the aperture itself would degrade the calibration measurements.

6.2 Mueller matrix measurements

After obtaining the calibration matrices, $W$ and $A$, the system was ready to make both Mueller matrix and Stokes measurements. The relevant irradiances, as outlined in Section 1.4, could be recorded and used to reconstructed into a Mueller matrix by computation of Eq. 6.2. The Stokes parameters of any polarisation state could also be computed using Eq. 1.1.

In the next sections, we show Mueller matrix measurements of some optical elements with predictable polarisation properties. These measurements were used to test the reliability of the system, and to check whether the impact of some interference fringes due to back reflections off some of the optical surfaces were significant. As is the case in the calibration procedure, these measurements were taken without the objective lens present, unless stated. When the objective was removed, the sample was placed as close as possible to the equivalent position of the entrance pupil of the objective lens.

In the following sections all of the Mueller matrix measurements were shown normalised to their respective $m_{11}$, while $m_{11}$ was shown un-normalised. This means that any intensity fluctuations present in coefficients apart from $m_{11}$ have been removed, and only information related to the polarisation is shown in the remaining coefficients. $m_{11}$ is left un-normalised as it contains information on the overall transmission of the sample. It is plotted on the same scale as the other coefficients so that they can be qualitatively, even though its pixel values never fall below zero. It was possible to carry out this
normalisation due to the single detector used for experimental measurements, which had the same effect on all irradiance measurements.

6.2.1 Calibration samples

The first set of elements to be measured and calibrated were the elements used as the calibration samples. These were a mirror (model no. 10D20DM.11, manufactured by Newport), a horizontal polariser, a vertical polariser (both 10LP-VIS-B, also manufactured by Newport), and a $\lambda/8$ waveplate rotated at $30^\circ$ (custom made by CVI-Melles Griot). Each of the samples was measured in double-pass. A second set of data was taken using the calibration samples, separate to that used to calculate the calibration matrices, $W$ and $A$. These measurements were a useful test of the alignment of the system, as well as the calibration matrices for the PSG and PSA.

The measurements in this section used 36 irradiance measurements to reconstruct the Mueller matrix. In most cases, each of these irradiance measurements was an average of 100 frames. Averaging was done by the camera itself before any files were saved; otherwise there would be too much data and the measurement time would be prohibitively long. The measurements were binned by a factor of 4, from $1024 \times 1328$ to $256 \times 332$, in order to speed up the computation of the calibrated matrices. All of the Mueller matrix measurements shown in this thesis were first normalised by dividing by the measurement for $m_{11}$. After this normalisation, only polarisation information will be shown by the remaining coefficients of the Mueller matrix.

The first sample to be measured was a mirror. As the reflection configuration used in the system was accounted for by Lara's modification to ECM [Lara 2006], the expected Mueller matrix for this sample should be equivalent to that for free space, i.e. the identity matrix. The measurement of this matrix is shown in Fig. 6.1.

As is clear in Fig. 6.1, this matrix is in broad agreement with its theoretical counterpart, which was equivalent to the $4 \times 4$ identity matrix. In $m_{11}$ of Fig. 6.1, it is clear that the irradiance was not entirely uniform across the pupil. This was most likely due to interference from a back reflection off one of the optical elements. As is clear in the other coefficients, it was possible to remove most of this effect by dividing by $m_{11}$. 
6.2. Mueller matrix measurements

Figure 6.1: Calibrated Mueller matrix of free space. As the system operated in reflection configuration, the MM of a mirror was measured instead of free space. This measurement could be equated to a measurement of the Mueller matrix of free space using Lara’s double pass ECM [Lara 2006].

An effect that was not removed by normalising to $m_{11}$ was that of a bubble on one of the optical surfaces. This bubble is not visible in $m_{11}$, and therefore did not introduce a fluctuation in irradiance. It did, however, introduce a polarisation effect, which is visible in $m_{13}$, $m_{22}$ and $m_{43}$ most clearly. This was not removed by the calibration as it was not a static source of error. It is likely that it was affected by vibrations or temperature fluctuations during the course of the measurement.

Figures 6.2 and 6.3 are the calibrated MMs of the two polarisers, a horizontal and vertical polariser respectively. These are the same samples that were used to calculate the calibration matrices, $W$ and $A$. A second set of Mueller matrix measurement data was recorded and calibrated using these
6.2. Mueller matrix measurements

Figure 6.2: Calibrated Mueller matrix of a horizontal polariser. The transmission of the polariser, $\tau_H$ is shown by the $m_{11}$ coefficient. The ideal MM for this polariser is given by Eq. 6.10.

calculated matrices as a test of the calibration procedure. The ideal matrices of a horizontal and vertical polariser differ only in the sign of the two of their coefficients:

$$M_1 = \tau_H \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad (6.10)$$

$$M_2 = \tau_V \begin{pmatrix} 1 & -1 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}. \quad (6.11)$$
Figure 6.3: Calibrated Mueller matrix of a vertical polariser. The ideal MM for this polariser is given by Eq. 6.11.

\( \tau_H \) and \( \tau_V \) are the transmission coefficients for the horizontal and vertical polarisers, respectively. Faint interference fringes are again visible in \( m_{11} \) for both samples. The transmission coefficient for each polariser can also be obtained by averaging \( m_{11} \) across valid pixels in the pupil. \( \tau_H \) was calculated to be while 0.3816, while \( \tau_V \) was found to be 0.4009. It is clear that more errors occur in the measurement of the vertical polariser in Fig. 6.3 than the other Mueller matrix measurements; the worst case being \( m_{13} \) in this measurement.

For this reason, we show, in Fig. 6.4, a calculation of the absolute difference between the measured Mueller matrix of \( B_2 \), and its ideal matrix in Eq. 6.11. In the case of calculating this absolute difference for \( m_{11} \), the distribution in this coefficient was compared to the calculated transmission, \( \tau_V \). The ideal Mueller matrices used in this section to represent the theoretical matrices of the calibration samples were determined using Mueller matrix algebra and the
Figure 6.4: Absolute difference between measured and expected Mueller matrix coefficients for B2, the vertical polariser. The matrix was normalised by dividing all coefficients by $m_{11}$, and then compared to the ideal matrix given in Eq. 6.11. $m_{11}$ was left un-normalised and was compared to the calculated transmission of the polariser.

characteristics of the sample as specified by the manufacturer.

The final calibration sample to be measured was the $\lambda/8$ waveplate rotated at $30^\circ$. This sample, like the others, was measured in double pass in conjunction with a mirror. Its theoretical Mueller matrix was modelled and calculated to be:

$$M_3 = \tau_H \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0.2500 & 0.4330 & -0.8660 \\ 0 & 0.4330 & 0.7500 & 0.5000 \\ 0 & 0.8660 & -0.5000 & 0 \end{pmatrix}. \quad (6.12)$$
6.2. Mueller matrix measurements

This matrix was calculated for a waveplate rotated at 30°. The experimental measurement is shown in Fig. 6.5. Most of the coefficients in the theoretical and experimental matrices are in agreement with each other. The most obvious discrepancy is in $m_{44}$, which should be equal to zero according to the theoretical matrix. It is likely that this discrepancy is due to the retardance of the plate not being exactly equal to $\lambda/8$.

We note that the systematic error, which arose from a bubble on one of the optical elements in the system, was again visible in the Mueller matrix measurements shown in Figs. 6.1 to 6.5. This caused the circular diffraction pattern in some of the Mueller matrix coefficients, such as $m_{32}$.
6.2.2 Objective lens

As the calibration of the system was carried out without the objective lens in place, it was desirable to ascertain whether the lens itself introduced any unwanted polarisation effects. The Mueller matrix of the lens was therefore measured in conjunction with a reference sphere in a reflection configuration. When the reference sphere was positioned correctly, rays diverging from the focal point of the lens were normally incident on the surface of the sphere, and reflected back along the same path. This is illustrated in Fig. 6.6. The sphere itself was custom made with an appropriate curvature using BK7 glass. It was uncoated, which meant that its reflectivity was approximately 4%. A schematic of the setup that was used for this measurement is shown in Fig. 6.6.

![Figure 6.6: Experiment to measure any polarisation effects introduced by the objective lens. Taken from [Rodríguez-Herrera 2009].](image)

The Mueller matrix measurement of the objective lens in double pass with reflection off the reference sphere is shown in Fig. 6.7. If the objective had introduced no polarisation effects, then this matrix would be equivalent to the identity matrix, and resemble Fig. 6.1. The diagonal coefficients are uniform and of approximately equal magnitude, which indicates that little polarisation effects are introduced by the lens. There are, however, some saddle-like distributions visible in coefficients $m_{24}$, $m_{34}$, $m_{42}$ and $m_{43}$. These coefficients show that a retardance is introduced by the lens; this is most likely related to the curved surfaces of some of the elements of the lens, or perhaps stress induced birefringence in some of the elements. It is interesting to note that the reflectivity of the reference sphere (~4%) is borne out in coefficient $m_{41}$. 
A back reflection off one of the curved surfaces of the lens is also visible in the centre of a number of the coefficients, for example $m_{13}$.

To further clarify the scale of the patterns visible in some of the Mueller matrix coefficients in Fig. 6.7, line-plots of two of the coefficients are shown in Fig. 6.8. These are plots in $x$ across the pupil of coefficients $m_{34}$ and $m_{43}$. Other coefficients in this Mueller matrix have patterns of a similar scale.

The diagonal coefficients of the measured matrix in Fig. 6.7 are many times larger than any of the other coefficients in the matrix. It was therefore decided that any polarisation effect introduced by the objective would be insignificant and should not degrade results. This measurement also showed that the polarimeter built was a sensitive Mueller matrix polarimeter, capable of measuring at lower light levels than optimum and sensitive retardance measurement.
6.2. Mueller matrix measurements

Figure 6.8: Line-plots in the $x$-direction of Mueller matrix coefficients $m_{34}$ and $m_{43}$ from Fig. 6.7. These are the values along the $x$-axis for $y = 0$. These particular coefficients are associated with the retardance of the specimen.

6.2.3 Radial polarisation converter

Another sample that could be measured was a radial polarisation converter. This was a birefringent plate, which was specified to have a uniform retardance of one half of a wave at 532 nm. The direction of the fast and slow axes changed with position of the plate, such that incident linear polarisation could be converted into radial or azimuthal polarisation, depending on the handedness of the incident polarisation.

This plate was manufactured from BK7 glass, by generating a nano-grating on the surface of the glass. The direction of the fast and slow axes at any point depended on the orientation of this grating. The grating was generated by scanning the glass with a femto-second laser; the orientation of the axes and the retardance depended on the power and polarisation of the incident beam, respectively [Beresna 2010, Beresna 2011].

The patterns in the coefficients of the measurement are typical of what would be expected for a radial polarisation converter, which should have a uniform retardance and a an azimuthally varying fast/slow axis orientation. An error is apparent, especially in $m_{42}$, due to the previously mentioned bubble on one of the optical surfaces. It is apparent from coefficients $m_{12}$, $m_{13}$,
6.2. Mueller matrix measurements

Figure 6.9: Calibrated Mueller matrix of a radial polarisation converter, measured in a double pass configuration. This element was manufactured by scanning BK7 glass with a femto-second laser, giving rise to a nano-grating and a birefringence pattern across the element.

$m_{21}$ and $m_{31}$ that this element introduces some diattenuation as well as retardance. A Mueller matrix can be decomposed to reveal properties of the optical element such as the retardance and diattenuation [Lu 1996]. Here, we use this decomposition to assess the diattenuation of this specimen. The diattenuation and diattenuation angle are shown in Fig. 6.10.

The Mueller matrix of this specimen was measured in a double pass configuration, with a mirror placed as closely as possible behind the waveplate in order to minimise any diffraction effects. The angle of diattenuation varies with azimuthal angle from $-90^\circ$ to $+90^\circ$, while the diattenuation itself also varies azimuthally. The magnitude of the diattenuation agrees with the irradiance distribution in $m_{11}$ of the Mueller matrix measurement in Fig. 6.9. This diattenuation would result in an irradiance variation in the radial polarisation
6.2. Mueller matrix measurements

6.2.4 Accuracy of measurements

As the calibration samples used were all uniform across the beam, it was expected that the coefficients of Mueller matrix measurements of the calibration samples (Figs. 6.1 to 6.5) would also be uniform across the pupil. In order to quantify this uniformity, a histogram was made of each of the coefficients in one of the measurements. The Mueller matrix of the mirror (Fig. 6.1) was used for this. The number of bins was chosen to be $\sqrt{n}$, where $n$ is the number of valid pixels in the pupil, approximately equal to 10000. Therefore the number of bins used was 100. For the remaining three calibration samples, the mean and standard deviation of the pixel distribution was calculated. The results from these calculations for the samples are shown in Table 6.2.4.

The Mueller matrix of the waveplate ($B_3$) provides some useful information on the accuracy of the calibration routine, particularly in the system’s ability to measure retardances. Comparing $m_{44}$ in the calibrated Mueller matrix of $B_3$, Fig. 6.5 and its theoretical counterpart in Eq.6.12, we see a significant difference between the measured and theoretical values for this coefficient. There are two possible explanations for this discrepancy. The first is that the retardance of the waveplate itself differs from the specified $\lambda/8$, resulting the in an inappropriate theoretical value for $m_{44}$. The second possible reason is that a
Figure 6.11: Distribution of pixel values within the coefficients of the measured and normalised Mueller matrix of free space (Fig. 6.1). The mean and standard deviation of the pixel distribution are annotated to the plot for each individual coefficient. The limits on the $x$-axis have a range of 1 for each plot, and are centred on the mean value.

A larger number of retarding samples should be used in determining the calibration matrices. Introducing a second waveplate of a different retardance and orientation would provide more information on the measurement capabilities of the system. $m_{44}$ is most illustrative in showing the retardance measurement capabilities of the system as its value is independent of the diattenuation and depolarisation properties of the specimen.

The mean and standard deviation of pixel values the pupil was also found for each coefficient. These are annotated to each of the histograms in Fig. 6.11. The Mueller matrix was normalised to $m_{11}$ before doing any calculations, while $m_{11}$ remained un-normalised. The interference pattern visible in $m_{11}$ contributed to the standard deviation of its pixel distribution, making the
Table 6.1: Table showing the mean and standard deviation of the distributions of pixels for the Mueller matrix measurements of each calibration sample.
6.2. Mueller matrix measurements

Figure 6.12: Calibrated Mueller matrix of the objective lens with a mirror placed in its focal plane. This is the mean of six measurements of this matrix.

The standard deviation of its distribution is larger than those of the other coefficients. Standard deviations for the other coefficients are smaller because this intensity variation has been removed by the normalisation.

Another measure of the stability of the system was to take a number of measurements of the same Mueller matrix sequentially, and then to find out whether this measurement changes significantly. This would provide information about the sensitivity of the system to fluctuations in the ambient conditions, such as temperature. This time the measurement of the Mueller matrix of a mirror (manufactured by Newport Corp., part no. 10CM00SB.1) placed at the focal plane of the objective lens was used. This plane mirror was placed in the focal plane of the objective lens, and a Mueller matrix measurement taken. This Mueller matrix is shown in Fig. 6.12. Six measurements of this Mueller matrix were recorded, and the standard deviation over the six measurements found for each coefficient. This is shown in Fig. 6.13, which
illustrates that the measurement did not change significantly between measurements, and that the polarimeter was stable, even though this standard deviation was taken over a relatively small number of measurements. Each of the six Mueller matrices was normalised to its respective $m_{11}$ before finding the standard deviation.

We note that all six MM measurements used to calculate this standard deviation were taken on a single afternoon, while conditions were relatively stable. The optical elements of the system were still susceptible to changes over time, or with varying conditions; therefore the system was calibrated approximately every two months.

![Figure 6.13: Standard deviation of the Mueller matrix measurement of objective lens and mirror. The mean of this measurement is shown in Fig. 6.12. This standard deviation is plotted on the same scale as Fig. 6.12 for ease of comparison. Six measurements of the MM of the mirror were recorded for calculation of the mean and standard deviation.](image)
Chapter 7
Results

7.1 Tests using nano-spheres

A point-like scatterer is often used as the first sample to test a newly built microscope. In this thesis, an 80 nm gold nano-sphere was used for this purpose. This sphere is small enough to be approximately equivalent to a dipole; the sphere will re-radiate incident light in a similar way to a dipole. Rodríguez showed that a vectorial polarimeter should be capable of measuring sub-resolution displacements of such scatterer. Experiments based on this finding were carried out in the testing of the polarimeter.

The nano-spheres rested on top of a glass cover slip, which was kept in place on a microscope slide using a small drop of oil. The spheres themselves were then covered using a drop oil and protected by another cover slip. The spheres were measured using an oil immersion objective lens (UPLSAPO 100XO, NA = 1.4). This was done in order to remove a strong back reflection off the top cover slip that occurred when the dry objective was used.

The objective of this experiment was to detect sub-resolution displacements of a nano-sphere, which was placed in the focal plane. A nano-sphere was located using the auxiliary dark-field setup described in Section 5.3, and carefully aligned using the nano-positioning stage, which had a resolution of 2 nm. The appropriate retardances were applied to the SLMs in order to generate the field in Fig. 7.1. Two Zernike patterns in voltage were applied to the SLMs gave rise to this focal field ($Z_{42}$ and $Z_{55}$ on SLMs 8917 and 8921, respectively).

The Stokes parameters of the scattered light off the nano sphere were then measured using the PSA at four positions of the sphere in the focal plane. The nano-sphere was moved in the $y$-direction away from the focus, equivalent to moving upwards in the image plane on the finder camera, in steps of 50 nm.
7.1. Tests using nano-spheres

Figure 7.1: Calculated focal field used in Stokes measurements of the 80 nm Au nano-sphere. This was calculated using McCutchen’s method. The retardances on the SLMs were defined using Zernikes $Z_{-2}^4$ and $Z_5^5$. Phase distributions are shown in units of $\pi$ radians, while the irradiance distributions have been normalised to the maximum irradiance in the focal plane.

Rodríguez showed that movements of a point-scatterer in the focus generates tilt-like patterns in the $s_3$ parameter, which represents the amount of circularly polarised light in the scattered and re-collimated light, and that an 80 nm nano-sphere can sufficiently approximate such a point scatterer [Rodríguez-Herrera 2010]. We therefore expected similar tilts to be detected in our measured Stokes parameters.

The measurements of the Stokes parameters at the four designated positions in the focal plane are shown in Fig. 7.2 to 7.5. The Stokes parameters were reconstructed from six irradiance measurements, and five images were taken to be averaged for every irradiance. For this measurement the number of irradiances was kept as low as possible in order to avoid drift in the positioning of the samples. Monitoring the piezo-electric stage showed that it varied in position by approximately 2 nm in all three dimensions. Each Stokes measurement was normalised to its respective irradiance distribution,
7.1. Tests using nano-spheres

Figure 7.2: Stokes measurement of the nano-sphere positioned in the focal plane; i.e., $x = y = z = 0$ nm.

Figure 7.3: Stokes measurement of the nano-sphere positioned in the focal plane, moved 50 nm away from the geometrical focus in the $y$-direction; i.e., $x = z = 0$ nm and $y = +50$ nm.

Figure 7.4: Stokes measurement of the nano-sphere positioned in the focal plane, moved 100 nm away from the geometrical focus in the $y$-direction; i.e., $x = z = 0$ nm and $y = +100$ nm.

Figure 7.5: Stokes measurement of the nano-sphere positioned in the focal plane, moved 150 nm away from the geometrical focus in the $y$-direction; i.e., $x = z = 0$ nm and $y = +150$ nm.
7.2. Using a grating

Figure 7.6: Profiles in the $x$-direction ($y = 0$) of the $s_3$ Stokes parameters taken from Figs. 7.2 to 7.2. It is clear that the slope of these profiles increases with increasing distance away from the geometrical focus.

$s_0$, while $s_0$ itself was plotted un-normalised.

It is clear from these figures that the $s_0$, $s_1$ and $s_2$ parameters stay relatively constant in their irradiance distribution, while a tilt in the $s_3$ parameter arises when the sphere is moved away from the focus. This tilt is horizontal, and increases in its magnitude with distance from the focus.

Figures. 7.2 to 7.5 show that the tilt in the $s_3$ distribution increases with increasing position from the geometrical focus. This agrees the results shown in [Rodríguez-Herrera 2010], and indicated that the experimental system was working well as a vectorial polarimeter. We note that these results were measured with the first iteration of the experimental system, described in Section 5.1, which used two retardances to control the polarisation distribution in the pupil of the objective lens, while the phase remained flat.

The increasing slope across $s_3$ with increasing distance away from the focus is further clarified in Fig. 7.6. In this figure the experimental values of $s_3$ along the $x$-axis for $y = 0$ are plotted, along with a linear fit. These plots clearly show the increasing slope in $s_3$ with increasing distance from the focus. While the movement of the nano-sphere was in the $y$-direction, the slope is visible in the $x$-direction.
7.2 Using a grating

A second specimen, which was investigated using the vectorial polarimeter, was a set of nine gratings with pitches varying from 25 nm up to 200 nm. The gratings were manufactured using pure silicon by NTT-AT (part no. NIM-25L/100). The gratings were convex with a height of 100 nm. This meant that the bars of the gratings protrude from the surface of the substrate. Their usual application was to be used as a mould for nano-imprint lithography. 100 line and space pairs were present for each of the nine pitches of grating, which were arranged in a $3 \times 3$ pattern. Larger bar marks were placed at a distance of 50 µm from the gratings and indicated the pitch of its adjacent grating. A schematic of the layout of the gratings on the sample surface is shown in Fig. 7.7. Each individual grating had a corresponding set of bar marks; the number of bars indicated the pitch of the adjacent grating. The largest pitch was labelled with a single bar mark, while the smallest grating was labelled with nine bar marks. A schematic of a single grating is shown in Fig. 7.8.

In order to investigate the polarisation effects of this specimen, the Mueller matrix of each of the gratings was measured. The Mueller matrix measure-
7.2. Using a grating

Figure 7.8: Schematic of one of the set of nine gratings of the NIM-25L/100, manufactured by NTT-AT. Image taken from the corresponding datasheet.

ments of each grating are shown in Appendix A. The 200 nm and the 150 nm gratings were not used as their pitches were close to the diffraction limit of the objective lens. These pitches were not of interest for the experiment as they were within the resolution limit of the optical system. The gratings were positioned in the focal plane of the objective lens by viewing the image of the irradiance distribution in the back aperture of the objective lens. For accurate positioning in the focal plane, the grating was moved along the optical axis using the piezo-electric stage until any circular diffraction patterns in the pupil irradiance distribution were minimised. This distribution was also checked at the end of each Mueller matrix measurement in order to ensure that the specimen had not moved. The Mueller matrix measurements were normalised to $m_{11}$ before being analysed.

The orientation of the gratings was approximately in line with the vertical polariser in front of the camera; it was therefore anticipated that the lines and spaces of the gratings would exhibit some diattenuation in the vertical direction, somewhat similar to the Mueller matrix of $B_2$ in Fig. 6.3. Each of the Mueller matrix measurements could be decomposed to find the diattenuation and other polarisation properties, such as the retardance, using Lu and Chipman’s decomposition [Lu 1996]. An example of the Mueller matrix of one of the gratings, in this case with a pitch of 65 nm, is shown in Fig. 7.9.
Figure 7.9: Mueller matrix measurement of the 65 nm grating. The set of gratings was aligned approximately in line with the vertical analysis polariser in front of the camera.

The rest of the Mueller matrix measurements are shown in Figs. A.1 to A.6 in Appendix A.

Using Lu’s decomposition method, it was found that the diattenuation depended on the pitch of the grating, with the most optimal vertical diattenuation occurring for the 80 nm grating. The diattenuation and angle of this grating is shown in Fig. 7.10. The distribution of the angle of diattenuation of this grating varies between 80° and 100°, which approximately is the same as the orientation of the lines of the grating. The effects of the gratings was not purely diattenuation, as evidenced by the Mueller matrix measurements in Appendix A, where significant retardance can also be qualitatively seen.

The retardance of each grating was also be extracted from the Mueller matrix measurements. It is likely that any retardance exhibited by the gratings will be due to form birefringence arising from the structure of the gratings, or
any innate retardance in the material used in their manufacture. The retardances of the six smallest gratings are shown in Fig. 7.11, while the angle of the fast axis of the calculated retardance is shown in Fig. 7.12. The angle of the fast axes of the gratings is approximately $90^\circ$ for all of the gratings. The apparently noisy points in the angles (especially in the angles of the 80 nm and 65 nm gratings), are due to the decomposition mixing up the fast and slow axes, as these points are approximately at $45^\circ$ to adjacent points.

It is clear from Fig. 7.11 that the magnitude of the retardance decreases with grating pitch, most likely due to a difference in the form birefringence of the varying grating pitches. The measurements show that polarisation dependent properties of a specimen depend on its sub-diffraction limit features. Information about the polarisation effects of the gratings is contained in these measurements, and could aid the tailoring of a focal field that could be used to obtain further information about the samples. For example, an appropriate focal field could be scanned across the lines of the grating and give rise to a signal that varies with position, amounting to super-resolution measurement of the pitch of the grating. Line-plots of the retardance distributions shown in Fig. 7.11 are shown in Fig. 7.13. In this figure, the magnitude of the retardance is plotted along the $x$-axis, for $y = 0$. These plots give a clearer idea of the relative scale of the distributions, as well as the similarity of their numerical values.
7.2. Using a grating

Figure 7.11: Retardance of each of the six gratings with smallest pitches. These were obtained by decomposing the corresponding Mueller matrix measurements in Appendix A.

On first glance, the distributions for the 45 nm and 50 nm grating look indistinguishable in both the retardance distribution (Fig. 7.11) and line-plots of along the $x$-axis (Fig. 7.13), while it appears that the overall magnitude of the retardance decreases for the 32 nm and 25 nm gratings. This is shown most clearly by the line-plots in Fig. 7.13. In order to investigate if any finer metric of distinction could be used to differentiate between of the four smallest gratings, histograms were compiled of the retardance distributions. One hundred bins were used for the histograms, since the number of valid pixels in the pupil was approximately ten thousand. These distributions are shown in Fig. 7.14, and reveal a skewed distribution for the four smallest gratings. This is due to the non-uniformity of the decomposed retardance. A clear peak is visible in each of the histograms and the value of the mode corresponding to this peak is annotated to its respective histogram plot.

The modes of the retardance distributions of the 45 nm and 50 nm gratings were 2.62 and 2.59 radians respectively. Taking the noise level of the retardance distributions into account it would be unwise to draw a conclusion that these values have a sufficiently significant discrepancy to differentiate between
these two pitches of gratings. The modes of the distributions corresponding to the 25 nm and 32 nm gratings, however, have a more significant change in their modal retardance value, decreasing to 2.31 and 2.18 radians respectively. This decrease suggests that the measurements were optimally sensitive to samples with feature sizes in this range. The information gathered from this measurement shows again that the vectorial polarimeter is capable of obtaining sub-resolution information about nano-scale specimens.

These Mueller matrix measurements also provide a clue to the eventual resolution of the method; looking at the histograms of the retardance distributions, there is a significant difference between the 25 nm and 32 nm gratings. This suggests that future work should concentrate on samples of size in the region of 25-50 nm. However, it is possible, when the full capability of tailoring the focal field in three dimensions is used, that this could be improved upon. Before the full potential of the instrument can be realised, an inverse problem must be solved, which involves prediction of the effect of any scatterer on its incident focal field. Solving this problem would provide more information on
7.2. Using a grating

Figure 7.13: Plots of the retardance distributions shown in Fig. 7.11 along the $x$-axis for $y = 0$. These plots provide a clearer view of the similarity of the retardance distributions.

The optimal focal fields to obtain further sub-resolution sensitivity using the technique of vectorial polarimetry.

The experimental results give some further information about the sub-resolution features of the nano-scale gratings. This was accomplished through the consideration of their polarisation effects by the measurement and decomposition of their Mueller matrices. Due to the dependence of properties such as diattenuation and birefringence on the pitch of the gratings, it is conceivable that a similar type of measurement, or a subset of the measurements taken in this experiment could be used in a field such as defect detection, with possible extension into sub-resolution imaging. Potentially this could be useful in fields such as metrology or defect detection in nano-scale structures.
7.2. Using a grating

Figure 7.14: Histograms of the retardance distributions shown in Fig. 7.11. The distributions are skewed Gaussian in shape due to the non-uniformity of the decomposed retardance.
8.1 Summary of this research

The main achievement of the research in this thesis was the building of an experimental system, which was capable of generating arbitrary phase and polarisation distributions across a beam. As the system used a microscope objective with a high numerical aperture, focusing of these beams resulted in a three-dimensional focal field, the shape of which depended on the polarisation and phase distribution present in the entrance pupil of the lens. The two iterations of the system are described in Chapter 5. The first of these controlled only the polarisation across the entrance pupil, while the second could also control the absolute phase.

The system was capable of converting a fixed input polarisation state into any polarisation and phase distribution in the entrance pupil of an objective lens. Analysis that proved this capability was shown in Section 1.5.1 using Jones calculus to represent the optical elements used. The input polarisation state used for our system was vertical. This analysis showed that two variable retardances, which are represented by linearly independent rotations on the Poincaré sphere, can be used to generate any polarisation state. A third optical element can be used to control the absolute phase across the beam. Three passes on two liquid crystal spatial light modulators were used in the experimental realisation of this concept. This analysis also applied to the polarisation state analyser of the system, which acted in reverse of the polarisation state generator.

Thorough characterisation and alignment of the individual optical elements used in the system were carried out as part of thesis. This was necessary due to the number of polarisation dependent elements and devices present. In particular, devices that had variable polarisation and phase properties were accurately calibrated so that they could be used optimally. These included
8.1. Summary of this research

the liquid crystal variable retarders and spatial light modulators. The SLMs were characterised for their use in both phase and retardance modulation, using separate measurements. This was described in Chapter 4.

Some focal fields that could be generated using this system were discussed in Chapter 3, using entrance pupil distributions that depended on polarisation only. Further examples depended on both polarisation and phase. It was found that polarisation only modulation could be used to split the focal field into perpendicularly polarised spots. Also, if a vortex in absolute phase is introduced to the beam, a cylindrical vector beam will be generated. The relative phase of a cylindrical vector beam was found to have a profound effect on the shape of the focal field, with the modelling of triangular, square and pentagonal focal fields. For these examples, the pupil and focal field distributions were represented in a polar coordinate system. These modelled results amounted to a small subset of the focal fields that could be potentially generated using the experimental system.

Debye-Wolf integration is the most common tool used to model the field at the focus of a high-NA objective. As was shown by McCutchen, a three-dimensional Fourier transform can also be used to model the field [McCutchen 1964, McCutchen 2002]. An advantage of using a Fourier transform for the calculation is that arbitrary polarisation and phase distributions are more readily defined as inputs. Faster computation times can also be attained using a Fourier transform based method. In Chapter 3 a comparison was carried out between these two methods, with little errors arising out of using a Fourier transform method. It was concluded that the Fourier transform based method was sufficiently accurate for most applications.

Calibration of the system was carried out using the Eigenvalue Calibration method, which consolidated any errors introduced by the individual optical elements of the system into two calibration matrices, one each for the polarisation state generator and the polarisation state analyser. Mueller matrix measurements of optical elements — including a mirror, polarisers, the high-NA objective itself lens among others — showed that the system had been calibrated successfully. The calibration of the setup is discussed in Chapter 6.

A point-like scatterer was used to test the experimental system in the initial experiments carried out. This sample was a 80 nm gold nanosphere.
8.2. Limitations encountered

It had previously been shown that displacements of a scatterer in the focal plane would give rise to tilts in $s_3$, the Stokes parameter related to circularly polarised light [Rodríguez-Herrera 2010]. Results from this experiment are shown in Chapter 7.

Further experiments used a set of gratings, most of which had pitches below the diffraction limit of the objective lens used in the system. Mueller matrix measurements of each of the gratings were recorded, and could be decomposed to quantify the polarisation dependent properties of the gratings. It was found that some of these properties depended on the pitch of the grating, even when the pitch was much smaller than the diffraction limit. For example, there was a difference between the retardance recorded for a 45 nm grating and that for a 32 nm grating.

The experiments and modelling results shown in this thesis pave the way for a new class of instrument that has potential uses in many fields. The system was shown to be sensitive to position changes in the sample of just 50 nm. This sensitivity could find uses in areas where the position of a component, such as lithography for microelectronics. Solving the inverse problems associated with the focusing and scattering of light off the sample may yet result in a novel method of super-resolution imaging, which requires no dye molecule or additions to the specimen under consideration.

8.2 Limitations encountered

Two inverse problems remain to be solved in order to maximise the information obtained using this method. The first of these involves the high-NA focusing system. It is possible to accurately predict the focal field for any given polarisation and phase distribution; however, for the method of vectorial polarimetry, it would be more useful to be able to define a field in the focal region and from this calculate the field in the entrance pupil of the lens. This is not simply an inverse Fourier transform, as the three-dimensional focal volume cannot be transformed into a two-dimensional plane. It is also probable that the solutions to the problem would not be unique. One method that has been proposed is to use a genetic algorithm, which would stochastically predict the field in the entrance pupil of the lens [Massoumian 2009]. Another
related approach optimised the pupil distribution, such that the focal field was maximally polarised either longitudinally or transversely [De Bruin 2011].

The second inverse problem to be solved regards the interaction of the three-dimensional focal field and the specimen placed in the focal field. Solving this problem means understanding the fundamental physical principles that govern the interaction. FDTD has already been used to model this process with accurate results [Rodríguez-Herrera 2010]; however, it more useful to have a conceptual understanding of the problem as this would likely result in information about the sample that would aid in the tailoring the focal field. It is possible that the interaction could be the result of a plasmonic effect [Prodan 2003], where the large surface to volume ratio results in most of the electrons in the specimen being closer to the surface than is the case in a bulk material, or another effect due to the relatively strong axial component of the focal field.

We note that this limitation of the method, as it stands, was most obvious when the set of gratings was used as a sample (see Section 7.2). It is anticipated that the solution of the second inverse problem would help find a focal field which could be scanned across the lines and spaces of the grating, resulting in a signal that modulated at the same spatial frequency as the pitch of the grating. This would amount to a super-resolution measurement.

8.3 Future work

The future work of this project will firstly involve finding solutions to the two inverse problems described in Section 8.2. The first inverse problem will find the appropriate pupil phase and polarisation distribution such that any desired focal field can be generated. Solving the second inverse problem will provide better understanding of the scattering of the three-dimensional focal field off a scatterer, and will aid in finding the resolution limits of the technique. The solutions to these problems should lead to further information being obtained about the specimen in the focal plane, which may be converted into a super-resolution image of the sample, and will lead to true tailoring of the focal field to the specimen under consideration.

It is also possible that the system could be used to develop a novel
method of polarimetry, which veers away from the use of Mueller matrices and uses Zernike polynomials as basis functions. Considering the results in Section 3.3.2, the field in the focus can be shaped by defining the relative phase in the pupil as a Zernike polynomial. This effect is particularly profound for the results shown in this section, which use Zernikes $Z_3^3$, $Z_4^3$ and $Z_5^5$ to define the relative phase. A new method of polarimetry could investigate the response of a sample to the applied fields as defined by an appropriate subset of the Zernike polynomials. The cylindrical vector beams described in this thesis result in a strong component parallel to the optical axis. It is feasible to suggest that the use of such fields in a polarimeter could result in further information about a specimen that is not usually obtainable with conventional imaging.

Another potential direction for the future research of this project would be to use its polarisation sensitivity in sub-resolution defect detection. An example of the kind of material, for which this could be appropriate, is graphene. Recent research has investigated the light-matter interactions of this substance [Koppens 2011], which showed that it can be used in the field of plasmonics. A further objective of the method described in this thesis would be to attain super-resolution imaging of biological samples. It is possible that the method could be tailored to search for a particular antigen or defect, and aid in diagnosis.
This appendix contains the measurements of the Mueller matrices of six of the gratings in the NI-25L/100L specimen. These results are discussed in Section 7.2. The seven smallest gratings were chosen for this measurement, ranging from a maximum of 100 nm to a minimum of 25 nm. It was anticipated that these pitches could yield sub-resolution information when analysed using the vectorial polarimeter setup. This setup was described in detail in Chapter 7. The grating pitches were 100 nm, 80 nm, 65 nm, 50 nm, 45 nm, 32 nm and 25 nm. The Mueller matrix measurement of the 65 nm grating was shown previously in Fig. 7.9 in Section 7.2, while the remaining measurements are shown in the following figures, in order of decreasing grating pitch. All of the Mueller matrices shown have been normalised to $m_{11}$ before plotting.
Figure A.1: Mueller matrix measurement of 100 nm grating.
Figure A.2: Mueller matrix measurement of 80 nm grating.
Figure A.3: Mueller matrix measurement of 50 nm grating.
Figure A.4: Mueller matrix measurement of 45 nm grating.
Figure A.5: Mueller matrix measurement of 32 nm grating.
Figure A.6: Mueller matrix measurement of 25 nm grating.
Bibliography


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