Spectropolarimetric characterization of light scattering materials

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SPECTROPOLARIMETRIC CHARACTERIZATION OF LIGHT SCATTERING MATERIALS

by

PRASHANT RAMAN

A DISSERTATION

Submitted in partial fulfillment of the requirement for the degree of Doctor of Philosophy in The Optical Science & Engineering Program to The School of Graduate Studies of The University of Alabama in Huntsville

HUNTSVILLE, ALABAMA

2012
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We, the undersigned members of the Graduate Faculty of the University of Alabama in Huntsville, certify that we have advised and/or supervised the candidate on the work described in this dissertation. We further certify that we have reviewed the dissertation manuscript and approve it in partial fulfillment of the requirements of the degree of Doctor of Philosophy in Optical Science and Engineering.

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Committee Chair
Advisor
Date

Program Director
College Dean
Graduate Dean

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ABSTRACT
The School of Graduate Studies
The University of Alabama in Huntsville

Degree       Doctor of Philosophy       Program       Optical Science & Engineering
Name of Candidate                 Prashant Raman
Title     Spectropolarimetric Characterization of Light Scattering Materials

UV-VIS-NIR Mueller matrix polarimeters are virtually non-existent due to the scarce availability of optical components performing optimally in such a broad waveband. A spectropolarimeter that measures the complete Mueller matrix of samples in transmission and reflection (ellipsometric) modes from 300 nm to 1100 nm at a high spectral resolution has been successfully designed, developed, calibrated and tested as part of this dissertation. The complete polarimetric characterization of light scattering materials is thus made possible.

Differentiating polarization properties of light scattered from various atmospheric particulate matter species, including pollen, dust, soot and B. subtilis, have been identified. This demonstrates the feasibility of this technique for the discrimination of bio-aerosol from ambient clutter.

Spectral Mueller matrix measurements of Gallium nanoparticles deposited on Sapphire substrates are presented which enables the developed system to locate the associated Localized Surface Plasmon Resonances (LSPRs). Additionally, the depolarization produced by such a nanoparticle system, not quantified previously, is measured by the developed system and duly presented.
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CHAPTER 1

INTRODUCTION

1.1 Introduction

Polarization is an intrinsic characteristic of electromagnetic radiation much like intensity, frequency and coherence. Three processes that can polarize natural (randomly polarized) light are reflection, refraction and scattering. Polarized light is abundantly found in nature [1]. However, the human eye cannot distinguish between the different states of polarization in the absence of an external aid, with the exception of Haidinger’s brush [2]. Polarimetry is the science of measuring and analyzing the polarization characteristics of light and the matter with which it interacts. Spectral polarization characteristics of light and matter provide information that can be used for several sensing, detection and characterization applications in astronomy, atmospheric remote sensing, biomedical diagnostics, quality control, homeland security, defense, optoelectronics and in the microfabrication industry.

1.2 Background

Different types of polarimeters have been developed in the past ranging from active to passive, from spectral to non-spectral, from monostatic to bistatic and from
imaging to non-imaging, based on intended applications and constraints. The polarimetric techniques and their architectures are well established and are briefly discussed in the chapter on Polarimetry in the Optical Society of America’s Handbook of Optics [3].

The ever-present threat of a chemical or biological attack has focused polarimetry research in the direction of early detection of hazardous materials. The importance of this endeavor, the many techniques employed, and the urgent need for improved technology are highlighted by Hurst [4]. Among the many techniques being explored, Mueller matrix polarimetry is equally promising [5].

The interaction of electromagnetic radiation with metal nanoparticles is of great interest due to the multitude of applications [6–8] and references therein. The polarization property that is exhibited as a result of this interaction especially at Localized Surface Plasmon Resonance (LSPR) is not completely understood. Mueller matrix (generalized) ellipsometry may provide additional and vital information on metal nanoparticle based systems [9].

1.3 Objective and Rationale

The primary objective of this research is to demonstrate the feasibility of adopting the polarimetric technique for non-destructive differentiation between different species of particulate matter such as spores, dust, pollen grains, smoke, etc. that may eventually lead to the development of point or standoff detection of bio-aerosols. The polarization scattering signature is known to depend on the shape, size, orientation and the optical constants of the particles in the scattering medium [10,11]. The size parameter of a
scattering particle is defined as the ratio of its characteristic dimension to the probing wavelength. Spectral variations in the polarimetric properties of scattered light typically increase with size parameter [12]. A qualitative description of the light scattering process is provided in Appendix A.

There have been a few theoretical studies modeling the characteristics of atmospheric aerosols by means of their polarization-dependent scattering [13,14]. Most experimental studies aimed at polarized light scattering from such samples are limited to the IR or visible region and often limited to a few discrete wavelengths [15,16]. Experimental studies over a broad wavelength region are scarce primarily due to lack of available instrumentation.

Another important objective is to measure the complete Mueller matrix and spectral polarization properties of metal nanoparticles in the UV-VIS-NIR region. This is expected to provide valuable insight into the interaction of electromagnetic radiation with nanoparticles, especially at LSPRs. Conventional ellipsometry measures the ratio of the complex reflection coefficients for the two orthogonal states of polarization and estimates the complex refractive index (optical constants) and thickness of a layer. Mueller matrix (generalized) ellipsometry on the other hand, is capable of measuring all 16 elements of the Mueller matrix from which all the polarization properties including depolarization and the optical constants can be calculated.

In order to accomplish the above objectives, a UV-VIS-NIR Mueller matrix polarimeter has been designed, developed, calibrated and tested from the ground up. This system provides a unique platform for the study of spectral polarization properties of samples with high accuracy and precision in the 300 nm to 1100 nm waveband at high
spectral resolution. The system offers the flexibility of performing measurements in different modes such as transmission, reflection, direct backscatter, etc. The system is also versatile enough to enable the study of optical components, electro-optical devices, optically active materials, polymers, etc.

1.4 Organization and Scope

The research work presented in this dissertation can be categorized into three sections, namely, theory, instrument development and experimental results. The first section includes Chapters 2 and 3, which discuss the polarization of light and the theory of polarimetry. The second part, Chapter 4, describes the design, development and testing of the constructed Mueller matrix spectropolarimeter. The final section, comprising Chapters 5 and 6, discusses the spectral Mueller matrix measurements and polarization properties of a variety of samples.

Chapter 2 of this dissertation introduces and describes the basic concepts related to the polarization of light. The emphasis is on the geometric and algebraic representations of polarized light and the calculus which describes the interaction of polarized light with matter. A brief description of the different polarization properties is provided and the Mueller matrices of the corresponding polarization elements are listed.

Chapter 3 briefly describes the different types of polarimeters and the theory of polarimetry specific to the dual rotating retarder polarimeter. This chapter also introduces the basics of ellipsometry since the polarimeter configured in the reflection mode parallels the operation of conventional ellipsometer.
Chapter 4 documents the design goals and considerations; the development, calibration, testing and validation of the Mueller matrix spectropolarimeter in transmission mode and reflection mode. The design and development of each subsystem of the polarimeter is discussed in detail along with the alignment procedure, performance issues and overall system performance. Calibration results are presented along with validation measurements using previously characterized components or theoretical calculations.

Chapter 5 begins with measurement results for PolyStyrene Latex (PSL) microspheres on a glass slide. Then an interpretation of the measurements performed on selected scattering samples is provided. The size and appearance of particulate matter under consideration, which was chosen for its relevance to bio-aerosol discrimination, along with sample preparation methods are discussed. Finally, discriminating polarization properties of different species of particulate matter deposited on microscope slides are presented for transmission and reflection modes.

Chapter 6 deals with the Mueller matrix ellipsometric measurements of Gallium nanoparticles on a sapphire substrate. First, a background is provided on the general theory of plasmonics along with the fabrication and in situ measurement scheme adopted by independent collaborators at Duke University. Then the Mueller matrix ellipsometric measurements from the UAH system is presented and compared to those provided by the same collaborators. Polarization properties, not observed previously to our knowledge, are highlighted.

Chapter 7 highlights the contribution made by this dissertation research and concludes with suggestions for future work.
1.5 Conclusions

This dissertation describes the successful development of a versatile UV-VIS-NIR Mueller matrix spectropolarimeter which provides a unique platform for studying a variety of samples such as optical components, electro-optical devices, polymers, particulate matter, etc. The results from this dissertation strongly suggest that point (or standoff) detection of bio-aerosol using such a polarimeter is entirely viable. In addition, this research paves the way for a comprehensive quantitative study of the polarization characteristics of light scattered from particulate matter and mixtures. Such a study could potentially evolve, for example, into a polarization-based differential LIDAR for standoff detection or a point detector employing an aerosol sampler.

The measurement results of Gallium nanoparticles presented in this dissertation establish the importance in performing complete polarimetric characterization of such nanoparticle systems. The Mueller matrix polarimeter parallels the operation of conventional ellipsometer while providing additional polarization information that has not been quantified previously. This potentially leads to a better and deeper understanding of electromagnetic interaction with nanoparticles.
CHAPTER 2

POLARIZATION OF LIGHT

2.1 Introduction

Based on observation of light propagating through crystals in the seventeenth century, it was deduced that light was not a scalar quantity [17]. The transverse vectorial nature of light is described through polarization. The polarization state of light is the temporal evolution of the electric field vector at a given position in space. The maximum amplitude of (and the phase between) the transverse components of the electric field vector describe the polarization state of light. The classical wave equation leads to the understanding and description of the propagation, diffraction, interference and polarization of light which is in complete agreement with experimental observations.

James Clerk Maxwell was first to formalize the description of electromagnetic waves in general. Maxwell’s equations for free space are given by the following equations [18],

\begin{align}
\nabla \cdot \mathbf{E} &= 0 \quad (2.1) \\
\nabla \cdot \mathbf{B} &= 0 \quad (2.2) \\
\n\nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t} \quad (2.3)
\end{align}
\[ \nabla \times \mathbf{B} = \mu_0 \varepsilon_0 \frac{\partial \mathbf{E}}{\partial t} \]  

(2.4)

where \( \mathbf{E} \) is the electric field, \( \mathbf{B} \) is the magnetic field, \( \mu_0 \) is the permeability of free space and \( \varepsilon_0 \) is the permittivity of free space. From these equations, the following electromagnetic wave equations can be derived [19],

\[ \nabla^2 \mathbf{E} - \mu \varepsilon \frac{\partial^2 \mathbf{E}}{\partial t^2} = 0 \]  

(2.5)

\[ \nabla^2 \mathbf{B} - \mu \varepsilon \frac{\partial^2 \mathbf{B}}{\partial t^2} = 0. \]  

(2.6)

It can be shown using Maxwell’s equations that the electric and magnetic field vectors of a plane wave are transverse to the direction of propagation. The transverse electric fields satisfying the electromagnetic wave equation reveal the nature of polarized light.

Polarized light can be described geometrically and algebraically. Geometrical description includes constructs such as the polarization ellipse and Poincare sphere. Algebraic descriptions utilize the Jones vector, Jones matrix, Stokes vector and Mueller matrix.

### 2.2 Geometrical Description of Polarized Light

Geometrical descriptions afford a visualization of polarized light which aids in the understanding of polarization. In this section the polarization ellipse and the Poincare sphere are briefly discussed.

#### 2.2.1 The Polarization Ellipse

The transverse electric fields \( E_x \) and \( E_y \) of a plane wave traveling along the \( z \) axis, satisfying the wave equation, can be written as,
where \( \tau = \omega t - kz \), the subscripts \( x \) and \( y \) refer to the components in the \( x \) and \( y \) directions, \( E_{0x} \) and \( E_{0y} \) are the maximum amplitudes, and \( \delta_x \) and \( \delta_y \) are the initial phases, respectively.

The amplitudes and phases of the electric field \( \mathbf{E} \) obey the following time independent equation,

\[
\frac{E_x^2}{E_{0x}^2} + \frac{E_y^2}{E_{0y}^2} - \frac{E_x E_y}{E_{0x} E_{0y}} \cos(\delta) = \sin^2(\delta)
\]

where \( \delta = \delta_y - \delta_x \). The above equation can be cast as that of an ellipse implying that the locus of points in the transverse plane described by the electric field vector as it propagates is an ellipse. Hence it is called the polarization ellipse [20] and is illustrated in Figure 2.1. Special cases of the polarization ellipse correspond to linear polarizations at any arbitrary orientation and two circular polarizations having opposite handedness (left and right). The most general case is that of an ellipse with arbitrary orientation, ellipticity and handedness.
Figure 2.1 A left-handed polarization ellipse

The polarization state of light can be described using the elliptical angles $\psi$ and $\chi$, which are defined as

$$\psi = \frac{1}{2} \tan^{-1}\left(\frac{2E_{0x}E_{0y} \cos(\delta)}{E_{0x}^2 - E_{0y}^2}\right), \quad 0 \leq \psi \leq \pi; \quad (2.10)$$

$$\chi = \frac{1}{2} \sin^{-1}\left(\frac{2E_{0x}E_{0y} \sin(\delta)}{E_{0x}^2 + E_{0y}^2}\right), \quad -\frac{\pi}{4} \leq \chi \leq \frac{\pi}{4}. \quad (2.11)$$

The ellipticity ($\varepsilon$) of the polarization ellipse is related to $\chi$ as,

$$\chi = \tan^{-1} \varepsilon, \quad (2.12)$$

where $\varepsilon$ is defined as the ratio of the length of the semi-minor axis to the length of the semi-major axis, viz.,

$$\varepsilon \equiv \pm \frac{b}{a}. \quad (2.13)$$
Positive ellipticity corresponds to right handed states of polarization while negative ellipticity corresponds to left handed states of polarization.

Orthogonal states of polarization have the same ellipticity but have orientations that are orthogonal to each other and have the opposite handedness.

### 2.2.2 The Poincare Sphere

Poincare (1892) discovered that the polarization ellipse can be represented on a complex plane. He then proposed a geometric representation now known as the Poincare sphere where the complex polarization plane is projected onto a unit sphere [20]. This idea was the converse of the more common stereographic projection of a sphere onto a plane. As illustrated in Figure 2.2, a completely polarized state with elliptical angle $\psi$ and orientation angle $\chi$ can be represented by a point on the Poincare sphere with a latitudinal angle of $2\psi$ and a longitudinal angle of $2\chi$. On the Poincare sphere, orthogonal polarization states are represented by two points located diametrically opposite each other. As proposed originally, the Poincare sphere representation only applied to completely polarized light. It has later been generalized to partially polarized light by using the radius to denote the degree of polarization (DoP). As a result, completely polarized light is represented on the sphere, partially polarized states occupy the interior of the sphere and unpolarized light is located at the center of the sphere.

Interestingly, the coordinates of each point on the sphere corresponds to three Stokes parameters (to be discussed in the next section), $S_1$, $S_2$ and $S_3$, for elliptically polarized light and the interaction of the polarized beam with an optical polarizing element corresponds to a rotation of the sphere where the final point describes the new
set of Stokes parameters after the interaction. A good review of the Poincare sphere can be found in [20].

![Figure 2.2 Poincare sphere](image)

2.3 Algebraic Description of Polarized Light

Among the analytical or algebraic approaches for describing polarized light and its interaction with matter, the Jones calculus and Mueller calculus are two popular formalisms. These frameworks are discussed in this section. Appendix B is a compilation of commonly encountered Jones vectors, Stokes vectors, Jones matrices and Mueller matrices.
2.3.1 Mueller Calculus

In 1852, Sir George Gabriel Stokes proposed a method for describing an arbitrary state of polarization. A set of four measurable parameters (Stokes parameters) are defined as

\[
\begin{align*}
S_0 &= \langle E_x E_x^* \rangle + \langle E_y E_y^* \rangle \\
S_1 &= \langle E_x E_y^* \rangle - \langle E_y E_x^* \rangle \\
S_2 &= \langle E_x E_y^* \rangle + \langle E_y E_x^* \rangle \\
S_3 &= i(\langle E_x E_y^* \rangle - \langle E_y E_x^* \rangle),
\end{align*}
\]

where \( E_x \) and \( E_y \) are the transverse components of the electric field vector and the angular brackets denote time average. The time average is performed since instantaneous measurements are impractical at optical frequencies of the order of \( 10^{15} \) Hz.

The Stokes parameters can be written in the form of a column vector in terms of the amplitudes and phases of the transverse components of the electric field as,

\[
\vec{S} = \frac{1}{S_0} \begin{bmatrix} 1 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \begin{bmatrix} E_{0x}^2 + E_{0y}^2 \\ E_{0x}^2 - E_{0y}^2 \\ 2E_{0x}E_{0y}\cos\delta \\ 2E_{0x}E_{0y}\sin\delta \end{bmatrix} \equiv \begin{bmatrix} I_{\text{tot}} \\ I_{\text{H}} - I_{\text{V}} \\ I_{45^\circ} - I_{135^\circ} \\ I_{R} - I_{L} \end{bmatrix}.
\]

The first element of the Stokes vector (\( S_0 \)) corresponds to the total irradiance of the light beam. The other three elements correspond to the differences between three orthogonal states of polarization, namely, horizontal and vertical polarization, 45° linear and 135° linear polarization, and right circular and left circular polarization.

Stokes parameters describe both completely polarized light as well as random and partially polarized light. The Stokes vector of randomly polarized light is,
The Stokes vector of two or more incoherent beams can simply be added to give
the Stokes vector of the combined beams. As a result, randomly polarized light can be
written as the sum of two (or more) orthogonal polarization states such as,

$$\begin{bmatrix}
1 \\
0 \\
0 \\
0
\end{bmatrix} = \frac{1}{2} \begin{bmatrix}
1 \\
s_2 \\
s_3 \\
s_4
\end{bmatrix} + \frac{1}{2} \begin{bmatrix}
1 \\
-s_2 \\
-s_3 \\
-s_4
\end{bmatrix}. \quad (2.20)$$

This form of representation reveals that randomly polarized light can be seen as a
composition (or incoherent sum) of an infinite pair of orthogonal polarization states.

From the Stokes parameters, the Degree of Polarization (DoP) can be defined as
the ratio of the irradiance of polarized components to the total irradiance of light.

$$\text{DoP} = \frac{I_{pol}}{I_{tot}} = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} \quad (2.21)$$

where $0 \leq \text{DoP} \leq 1$. DoP = 1 represents completely polarized states and DoP = 0
represents randomly polarized light, while partial polarization states assume values in
between. In the most general case,

$$S_0 \geq \sqrt{S_1^2 + S_2^2 + S_3^2}. \quad (2.22)$$

The Degree of Linear Polarization (DoLP) and the Degree of Circular
Polarization (DoCP), as the names suggest, quantify the amount of linear and circular
polarizations respectively. They are defined as,

$$\text{DoLP} = \frac{\sqrt{S_1^2 + S_2^2}}{S_0} \quad (2.23)$$
The interaction of light with a medium invariably causes a change in the polarization of light. This interaction can be modeled as a linear transformation that maps an incident Stokes vector ($\vec{S}_{in}$) onto an exiting Stokes vector ($\vec{S}_{out}$), written as,

$$\vec{S}_{out} = \mathbf{M} \cdot \vec{S}_{in} = \begin{bmatrix} m_{00} & m_{01} & m_{02} & m_{03} \\ m_{10} & m_{11} & m_{12} & m_{13} \\ m_{20} & m_{21} & m_{22} & m_{23} \\ m_{30} & m_{31} & m_{32} & m_{33} \end{bmatrix} \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix}. \tag{2.25}$$

The 4x4 matrix is called the Mueller matrix and represents the polarization characteristics of the medium. The first element of the Mueller matrix ($m_{00}$) corresponds to the absolute throughput of the medium for incident unpolarized light. The other elements of the matrix relate to the different polarization properties of the medium such as its retardance, diattenuation and depolarization in a complex manner. However, Mueller matrices do not contain any absolute phase information and hence cannot be applied to interferometry or holography. Also, not all 4x4 real matrices are physically realizable Mueller matrices just as not all 4x1 real column vectors are valid Stokes vectors [21,22].

Mueller matrices can be viewed as a linear transfer function in that they transform an input Stokes vector into an output Stokes vector. Thus a train of optical polarization elements can be represented as a single Mueller matrix equal to the product of the constituent Mueller matrices, such as,

$$\mathbf{M}_{sys} = \mathbf{M}_n \cdot \mathbf{M}_{n-1} \ldots \mathbf{M}_2 \cdot \mathbf{M}_1 \tag{2.26}$$
2.3.2 Jones Calculus

Based on the amplitudes and the phases of the transverse components of the electric field, Jones in 1941 proposed a complex vector representation for completely polarized light. The Jones vector is a 2-element column vector given by,

\[
\vec{J} = \begin{bmatrix} E_x \\ E_y \end{bmatrix} = \begin{bmatrix} E_{0x} e^{i\delta_x} \\ E_{0y} e^{i\delta_y} \end{bmatrix},
\]

(2.27)

where \(E_{0x}\) and \(E_{0y}\) are the amplitudes, and \(\delta_x\) and \(\delta_y\) are the phases of the electric field in the \(x\) and \(y\) direction respectively.

Jones also proposed a 2x2 complex matrix representation, now called a Jones matrix, to characterize a polarizing optical component. Polarized light interacting with such an optical component is mathematically described by the optical component’s Jones matrix linearly transforming the Jones vector of incident light. The Jones matrix can be written out as,

\[
J = \begin{bmatrix} J_{11} & J_{12} \\ J_{21} & J_{22} \end{bmatrix}.
\]

(2.28)

Any complex 2x2 matrix is a valid Jones matrix just as any 2x1 complex vector is a valid Jones vector. If \(\vec{J}_{\text{in}}\) and \(\vec{J}_{\text{out}}\) are the incident and exiting Jones vectors respectively, the transformation is then written as,

\[
\vec{J}_{\text{out}} = J \cdot \vec{J}_{\text{in}}
\]

(2.29)

Unlike Mueller calculus, Jones calculus can only deal with completely polarized light. However, the Jones approach includes phase information. The Jones matrix, too, can be seen as a linear transfer function between the incident and exiting Jones vectors. As a result, a train of optical polarization elements can be represented as a product of individual Jones matrices as,
\[ J_{\text{sys}} = J_n \cdot J_{n-1} \cdots J_2 \cdot J_1 \] (2.30)

### 2.4 Polarization Properties of Polarizing Elements

The polarization state of the incident light invariably undergoes a change when it interacts with matter. An optical element that is capable of deterministically changing the polarization state of light is called a polarization element. There are essentially four different mechanisms by which a polarization element can manipulate incoming light, namely, diattenuation, retardation, rotation and depolarization. The elements that manipulate incident light in one of the above four ways are called diattenuators (or polarizers), retarders, rotators and depolarizers, respectively.

Diattenuation refers to the different amounts of attenuation experienced by the orthogonal states of polarization. The attenuation is maximum for one state and minimum for its corresponding orthogonal state. Linear diattenuation and circular diattenuation refers to the diattenuation experienced by a pair of orthogonal linear and circular states, respectively. Polarization elements exhibiting this property are called diattenuators. The diattenuation (D) is quantitatively defined as,

\[ D = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}}, \] (2.31)

where \( I_{\text{max}} \) and \( I_{\text{min}} \) refer to the irradiances of the orthogonal polarization states. In the strictest sense, only ideal diattenuators with \( D = 1 \) qualify to be called pure polarizers. However commercially available diattenuators are commonly referred to as polarizers since they have \( D \to 1 \). A neutral density filter would have \( D = 0 \).

Another quantity popular in commercial circles is the Extinction Ratio (ER) which specifies the strength of the diattenuation. ER is defined as,
ER = \frac{1 + D}{1 - D} \quad (2.32)

ER = \infty \text{ for an ideal polarizer and } ER = 1 \text{ for a non-polarizer. A typical Glan-Thompson prism has an extinction ratio of about } 10^6. \text{ Polarization elements are based on polarization by absorption (dichroism), refraction (birefringence) and reflection (total internal reflection or Brewster angle reflection).}

Retardation refers to polarization-dependent phase change. The orthogonal states of polarization experience a differential phase change and the phase difference between the two orthogonal states of polarization (\(\delta\)) is called retardance. The polarization elements exhibiting this property are called retarders, waveplates, or compensators. Retardance can be achieved using different mechanisms like birefringence and total internal reflection. For a retarder based on birefringence, retardance is given by,

\[ \delta = \frac{2\pi}{\lambda} \Delta n \ d, \quad (2.33) \]

where \(\Delta n = |n_1 - n_2|\) is the birefringence or the difference in the refractive indices for the pair of orthogonal states and \(d\) is the thickness of the birefringent material.

Rotation of the plane of polarization can occur through optical activity, the Faraday Effect and by the action of certain liquid crystals. Optical activity can be explained in terms of left and right circularly polarized states and the refractive index that these states experience. The rotatory power of an optically active medium is

\[ \rho = \frac{\pi (n_L - n_R)}{\lambda} \quad (2.34) \]

in degrees per centimeter, where \(n_L\) and \(n_R\) are the refractive indices for left and right circularly polarized states, respectively. If \(d\) is the thickness of the optically active medium, then the rotation angle is
Depolarization refers to the process of coupling polarized light into unpolarized light. An ideal depolarizer transforms any incident state of polarization into randomly polarized light. The degree of polarization (DoP) of light exiting an ideal depolarizer equals zero. The depolarization index [3] is quantified as,

\[
\text{Dep}(M) = 1 - \sqrt{\frac{\sum_{i,j} m^2_{i,j} - m^2_{00}}{\sqrt{3} m_{00}}}.
\]

(2.36)

The depolarization index can take values between 0 and 1. Dep(M) = 0 corresponds to a non-depolarizing sample while Dep(M) = 1 indicates a complete or ideal depolarizing sample.

Polarizance refers to the property of the sample whereby unpolarized light is transformed or coupled into polarized light. The polarizance can be linear or circular based on the transformed state of polarization being linear or circular, respectively.

### 2.5 Mueller Matrix of Polarizing Elements

Polarimetry involves the measurement and analysis of the polarization characteristics of light and the matter with which it interacts. The Stokes vector – Mueller matrix framework deals with measurable irradiances in the Stokes vector while the Jones vector – Jones matrix framework deals with electric field amplitudes and phases, both of which are difficult or impossible measure. Hence, the Stokes vector – Mueller matrix framework is preferred for this application. Goldstein [20] discusses in detail the Mueller matrices of various polarization elements. A brief review of the Mueller matrices of polarizing elements is provided here.
2.5.1 Linear Polarizer

Suppose \( p_x \) and \( p_y \) are the amplitude attenuation coefficients along orthogonal transmission axes of the polarizer. The electric field components of the emerging beam \( E'_x \) and \( E'_y \) in terms of the incident electric field components \( E_x \) and \( E_y \) are written as,

\[
E'_x = p_x E_x; 0 \leq p_x \leq 1 \tag{2.37}
\]
\[
E'_y = p_y E_y; 0 \leq p_y \leq 1. \tag{2.38}
\]

The Mueller matrix describing the polarizer can be derived to be,

\[
M_p = \frac{1}{2} \begin{bmatrix}
  p_x^2 + p_y^2 & p_e^2 - p_y^2 & 0 & 0 \\
  p_x^2 - p_y^2 & p_x^2 + p_y^2 & 0 & 0 \\
  0 & 0 & 2p_x p_y & 0 \\
  0 & 0 & 0 & 2p_x p_y
\end{bmatrix} \tag{2.39}
\]

For an ideal linear polarizer along the x-axis, \( p_x = 1 \) and \( p_y = 0 \). Using parametric substitution \( p_x = p \cos \gamma \), \( p_y = p \sin \gamma \) and \( p_x^2 + p_y^2 = p^2 \), the Mueller matrix for a polarizer may be written as

\[
M_p = \frac{p^2}{2} \begin{bmatrix}
  1 & \cos 2\gamma & 0 & 0 \\
  \cos 2\gamma & 1 & 0 & 0 \\
  0 & 0 & \sin 2\gamma & 0 \\
  0 & 0 & 0 & \sin 2\gamma
\end{bmatrix} \tag{2.40}
\]

where \( 0 \leq \gamma \leq 90^\circ \). For an ideal linear polarizer \( p = 1 \). For a linear horizontal polarizer \( \gamma = 0^\circ \) and for a linear vertical polarizer \( \gamma = 90^\circ \).

2.5.2 Linear Retarder

A retarder introducing a phase shift of \( \varphi \) between two orthogonal linear states of polarized light can be viewed as imparting a phase of \( + \varphi/2 \) to one polarization state (fast axis) and a phase of \( - \varphi/2 \) to its corresponding orthogonal polarization state (slow axis). The electric field components of the emerging beam are related to the electric field components of the incident beam by,
$$E_x' = e^{+i\varphi/2}E_x$$
$$E_y' = e^{-i\varphi/2}E_y.$$  \hspace{1cm} (2.41)

The Mueller matrix of such a retarder can be written as,

$$M_R = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & \cos \varphi & \sin \varphi \\ 0 & 0 & -\sin \varphi & \cos \varphi \end{bmatrix}.$$ \hspace{1cm} (2.43)

### 2.5.3 Rotator

A rotator rotates the orthogonal electric field components through an angle ($\theta$).

The Mueller matrix for rotation is found to be,

$$M(\theta) = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2\theta & \sin 2\theta & 0 \\ 0 & -\sin 2\theta & \cos 2\theta & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}.$$  \hspace{1cm} (2.44)

Note that the above matrix resembles a traditional 3-D rotation matrix.

### 2.5.4 Rotated Polarizing Elements

If $M_X$ is the Mueller matrix of a polarization element then the Mueller matrix of the polarization element when rotated by an angle $\theta$ is given by [20],

$$M_X(\theta) = M(-\theta) \cdot M_X \cdot M(\theta).$$  \hspace{1cm} (2.45)

The Mueller matrix of a polarizer rotated by an angle $\theta$ is given by,

$$M_P(\theta) = \begin{bmatrix} 1 & \cos 2\gamma & \cos 2\gamma \cos 2\theta & \cos 2\gamma \sin 2\theta & 0 \\ \cos 2\gamma \cos 2\theta & \cos^2 2\theta + \sin 2\gamma \sin^2 2\theta & (1 - \sin 2\gamma) \sin 2\theta \cos 2\theta & 0 \\ \cos 2\gamma \sin 2\theta & (1 - \sin 2\gamma) \sin 2\theta \cos 2\theta & \sin^2 2\theta + \sin 2\gamma \cos^2 2\theta & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}.$$ \hspace{1cm} (2.46)

The Mueller matrix of a retarder with phase shift $\varphi$ rotated by an angle $\theta$ is given by,

$$M_R(\theta) = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos^2 2\theta + \cos \varphi \sin^2 2\theta & (1 - \cos \varphi) \sin 2\theta \cos 2\theta & -\sin \varphi \sin 2\theta \\ 0 & (1 - \cos \varphi) \sin 2\theta \cos 2\theta & \sin^2 2\theta + \cos \varphi \cos^2 2\theta & \sin \varphi \cos 2\theta \\ 0 & \sin \varphi \sin 2\theta & -\sin \varphi \cos 2\theta & \cos \varphi \end{bmatrix}.$$ \hspace{1cm} (2.47)
2.5.5 Metal Mirror

A metal mirror imparts diattenuation and retardance to the light incident on it. The magnitude of diattenuation and phase shift is dependent on the complex refractive index of the metal (n+ik) and the incident angle (θ). The Mueller matrix of such a metal mirror has been derived by Deibler and Smith [23]. The Mueller matrix has the form,

\[
M_{\text{mir}} = \frac{1}{D} \begin{bmatrix}
m_{00} & m_{01} & 0 & 0 
m_{10} & m_{11} & 0 & 0 
0 & 0 & m_{22} & m_{23} 
0 & 0 & m_{32} & m_{33}
\end{bmatrix},
\]

(2.48)

where,

\[
D = (\cos^2\theta + n^2 + k^2 + 2n \cos \theta)(1 + n^2 \cos^2 \theta + k^2 \cos^2 \theta + 2n \cos \theta),
\]

(2.49)

\[
m_{00} = n^2 + k^2 + [1 - 4n^2 + (n^2 + k^2)^2 + (n^2 + k^2) \cos^2 \theta] \cos^2 \theta,
\]

(2.50)

\[
m_{01} = 2n(n^2 + k^2 - 1) \cos \theta \sin^2 \theta,
\]

(2.51)

\[
m_{22} = n^2 + k^2 - [1 - 4k^2 + (n^2 + k^2)^2 - (n^2 + k^2) \cos^2 \theta] \cos^2 \theta,
\]

(2.52)

\[
m_{23} = -2k(n^2 + k^2 - 1) \cos \theta \sin^2 \theta.
\]

(2.53)

2.5.6 Dielectric Plate

A dielectric plate, on the other hand, imparts diattenuation to the incident light. The diattenuation is dependent on the incident angle and refractive indices of the incident medium and dielectric plate. Another important factor to take into account is the multiple internal reflections that occur within the plate which greatly influences the polarization properties of the dielectric plate for high incident angles. The Mueller matrix for reflection from and transmission through a dielectric plate based on Fresnel coefficients have been derived by Collett [24]. The Fresnel coefficients for reflection and transmission for the s and p polarization states are given by,
\[ R_s = \left( \frac{\sin(\theta_i - \theta_t)}{\sin(\theta_i + \theta_t)} \right)^2, \]  \hspace{1cm} (2.54)

\[ R_p = \left( \frac{\tan(\theta_i - \theta_t)}{\tan(\theta_i + \theta_t)} \right)^2, \]  \hspace{1cm} (2.55)

\[ T_s = \left( \frac{2 \sin \theta t \cos \theta t}{\sin(\theta_i + \theta_t)} \right)^2, \]  \hspace{1cm} (2.56)

\[ T_p = \left( \frac{2 \sin \theta t \cos \theta t}{\sin(\theta_i + \theta_t) \cos(\theta_i - \theta_t)} \right)^2. \]  \hspace{1cm} (2.57)

where \( \theta_i \) is the incident angle and \( \theta_t \) is the transmitted angle.

The Mueller matrix of the dielectric plate in transmission is given by,

\[
\begin{bmatrix}
T_s^2 + T_p^2 & T_s^2 - T_p^2 & 0 & 0 \\
T_s^2 - T_p^2 & T_s^2 + T_p^2 & 0 & 0 \\
0 & 0 & 2T_sT_p & 0 \\
0 & 0 & 0 & 2T_sT_p
\end{bmatrix}
\]

(2.58)

The Mueller matrix of the dielectric plate in reflection is given by,

\[
\begin{bmatrix}
R_s + R_p & R_s - R_p & 0 & 0 \\
R_s - R_p & R_s + R_p & 0 & 0 \\
0 & 0 & 2(R_sR_p)^{1/2} & 0 \\
0 & 0 & 0 & 2(R_sR_p)^{1/2}
\end{bmatrix}
\]

(2.59)

2.6 Conclusions

This chapter has introduced the basic concepts related to polarization of light and provides a necessary background for understanding polarimetry and the experimental results in later chapters.
CHAPTER 3

POLARIMETRY

3.1 Introduction

Polarimetry is the science of measuring and analyzing the polarization characteristics of light and the matter it interacts with. It can be thought of as radiometry with polarization elements. There are different types of polarimetric instrumentation, based on the application, to facilitate the measurement being made. There are also several analysis techniques available, based on the chosen instrumentation and application, to extract relevant polarization information from polarimetric data. The process of extracting the Mueller matrix from a set of polarization modulated irradiance measurements is referred to as polarimetric data reduction. The accuracy of the data reduction process depends on the errors in the radiometric measurement and the inherent sensitivity of the system to these errors.

There are two general types of measurement errors: systematic and random. Random errors by definition are not deterministic and hence cannot be analytically modeled. Random errors are minimized by cutting off stray light, using lock-in detection schemes and statistical techniques such as oversampling. Systematic errors are inherent instrumental errors that can be estimated and compensated for in the data processing.
Performing accurate polarization measurements is difficult on account of the systematic errors in the non-ideal polarization elements. The process of estimating and compensating the systematic errors in a polarimeter (or any piece of measurement equipment) is referred to as calibration. The calibration and subsequently the data reduction process following the polarimetric measurements must correct for the systematic errors in the polarimeter.

This chapter gives a brief background of polarimetric instrumentation and then proceeds to discuss the theory of the dual rotating retarder Mueller matrix polarimeter. Analytical and numerical calibration schemes accounting for the systematic errors in the polarimeter are dealt with. A data reduction framework based on linear algebra for computing the sample’s Mueller matrix is also discussed, along with post-processing techniques dealing with how the closest physically realizable Mueller matrix is computed and decomposed to determine the polarization properties of the sample. Finally, the basics of ellipsometry are introduced since this dissertation also involves polarimetric measurements in reflection (similar to ellipsometry) and calculation of the optical constants of an important type of sample.

3.2 Overview of Polarimetric Instrumentation

Different types of polarimeters have been developed in the past ranging from complete to incomplete, from active to passive, from spectral to single channel, from monostatic to bistatic and from imaging to non-imaging, purely based on intended applications and constraints. These polarimetric techniques and their architectures are
well established and are discussed in the chapter on Polarimetry in the Handbook of Optics [3]. Ellipsometry is discussed by Azzam in the Handbook of Optics [25].

Passive polarimeters have a set of polarization elements to analyze the incoming light, and measure the Stokes vector corresponding to the polarization state of light. Such polarimeters determine the direction of oscillation of the electric field vector for linearly polarized light, the helicity of circularly polarized light or the parameters of elliptically polarized light. Sample-measuring polarimeters or active polarimeters determine the relation between the polarization states of the incident and exitant light beam, thus characterizing the polarization properties of the sample. The exiting beam includes light that is transmitted, reflected, diffracted or scattered. Active polarimeters include a Polarization State Generator (PSG) between the source and the sample and a Polarization State Analyzer (PSA) between the sample and the detector. The PSG and PSA are a separate set of polarization elements.

Complete polarimeters are those that determine the entire Stokes vector or all the elements of the Mueller matrix, while incomplete polarimeters are those that do not. Monostatic polarimeters are those in which the PSG and PSA are collocated and bistatic polarimeters are those in which the PSG and PSA are not collocated.

Multiple measurements must be made from which the Stokes vector or the Mueller matrix can be determined. Three modulation techniques are available in order to generate and analyze polarization states and hence achieve the necessary measurements: temporal, spatial and spectral modulation. In a polarimeter employing the temporal modulation technique, the measurements are taken sequentially in time and the polarization analyzer and/or polarization generator is changed (for example rotating
polarization elements or electro-optical modulators) between consecutive measurements. Polarimeters based on spatial modulation employ multiple polarization analyzers operating side-by-side in a subdivided aperture like a microgrid array [26]. Spectral modulation is achieved in polarimeters using techniques like ‘channeled spectropolarimetry’ for passively determining the spectrally resolved Stokes vector of incoming light [27] and ‘snapshot polarimetry’ for actively determining the spectrally resolved Mueller matrix elements of the sample [28]. Each of the above modulation techniques have their own advantages and disadvantages, and are chosen based on requirements and constraints specific to the intended application. The dual rotating retarder Mueller matrix polarimeter discussed here and developed as part of the dissertation employs the temporal modulation scheme.

### 3.3 Dual Rotating Retarder Polarimeter

The dual rotating retarder polarimeter proposed by Azzam [29] is the most popular and widely adopted architecture for complete Mueller matrix polarimetry. The polarimeter is composed of a combination of a polarizer and a linear retarder in reverse order in the PSG and PSA respectively, as can be seen in Figure 3.1. The polarimetric measurement sequence begins with the transmission axes of the polarizers co-aligned with the fast axes of the linear retarders. Radiometric measurements are performed at specific rotational positions of the two linear retarders, maintaining a ratio of 1:5 between the rotations of the retarder in the PSG to that of the PSA. The 1:5 ratio allows for the most accurate inversion of the DRM to yield the Mueller matrix. If N is the highest harmonic present in the modulation, sampling theory dictates that a minimum of
(2*N) + 1 measurements be performed. Hence, a higher ratio will require an even larger number of measurements to be performed. A different ratio may well be used if numerical techniques are employed instead of the analytical Fourier decomposition technique.

The model equation describing the polarimeter operation is given by,

\[ \vec{S}_{\text{out}}^{(q)} = P_2 R_2^{(q)} (5\theta) M R_1^{(q)} (\theta) P_1 \vec{S}_{\text{in}} \]  

where \( P_1 \) and \( P_2 \) are the Mueller matrices of the polarizers and \( R_1^{(q)} \) and \( R_2^{(q)} \) are the Mueller matrices of the retarders in the PSG and PSA respectively. The index \( q \) denotes different rotational positions of the retarders and \( \theta \) is the rotation angle of the first retarder corresponding to the \( q^{\text{th}} \) measurement. \( M \) is the Mueller matrix of the sample while \( \vec{S}_{\text{in}} \) and \( \vec{S}_{\text{out}} \) are the incident and exitant Stokes vectors respectively.
3.4 Data Reduction Framework

In order to measure all 16 elements of the Mueller matrix of a sample, a dual rotating retarder polarimeter should make \( Q \geq 16 \) irradiance measurements at unique orientations of the PSG and PSA retarders. This provides a system of equations with 16 or more equations from which all the 16 unknowns (Mueller matrix elements) can be determined. This is achieved by rotating retarders \( R_1 \) and \( R_2 \) in discrete angular steps, maintaining a ratio of 1:5 between them to encompass all the non-degenerate measurement states. The indexing runs from \( q = 0, 1, 2, \ldots, Q-1 \). For the \( q^{th} \) measurement, the irradiance measured \( (I_q) \) is given by the first element of the Stokes vector \( \tilde{S}_A^{(q)} \) in the model equation above. The irradiance \( I_q \) can be written in terms of the Stokes vector of the beam produced by the PSG \( (\tilde{S}_q) \), the Mueller matrix of the sample \( (M) \) and the analyzer vector \( (A_q) \), as follows,

\[
I_q = A_q^T M \tilde{S}_q. 
\]  

(3.2)

The analyzer vector \( (A_q^T) \) is a row vector composed of the first row of the Mueller matrix of the PSA as opposed to a regular Stokes vector which is a column vector. Figure 3.2 shows the normalized irradiance modulation plot when the Mueller matrix is an identity matrix (corresponding to the Mueller matrix of air). The rotation of the retarders in the PSG and PSA, as discussed earlier, follows the 1:5 ratio while each retarder ideally produces a retardance of a quarter wave. The polarizers in the PSG and PSA are assumed to be ideal and the polarization axes of the polarizers and retarders are co-aligned at the beginning.
Equation 3.2 can be written as a vector-vector dot product as,

$$I_q = \overline{W}_q \cdot \overline{M}, \quad (3.3)$$

where $\overline{W}_q$ is a 16 x 1 polarimetric vector defined as,

$$\overline{W}_q = (a_{q,0}s_{q,0} \ a_{q,0}s_{q,1} \ a_{q,0}s_{q,2} \ a_{q,0}s_{q,3} \ a_{q,1}s_{q,0} \ \cdots \ a_{q,3}s_{q,3})^T \quad (3.4)$$

and $\overline{M}$ is a 16 x 1 column vector constructed using the Mueller matrix elements,

$$\overline{M} = (m_{00} \ m_{01} \ m_{02} \ m_{03} \ m_{10} \ \cdots \ m_{33})^T. \quad (3.5)$$

Equation 3.3 represents a single irradiance measurement at state $q$. Generalizing the above into a matrix equation describing all $Q$ measurements gives,

$$\bar{I} = W\overline{M} = \begin{bmatrix} W_0^T \\ W_1^T \\ W_2^T \\ W_3^T \\ \vdots \\ W_{Q-1}^T \end{bmatrix} \begin{bmatrix} m_{00} \\ m_{01} \\ m_{02} \\ m_{03} \\ \vdots \\ m_{33} \end{bmatrix}, \quad (3.6)$$
where \( \mathbf{I} = (I_1 \ I_2 \ I_3 \ \ldots \ I_Q)^T \) is a column vector representing \( Q \) irradiance measurements. The matrix \( \mathbf{W} \) is called the polarimetric measurement matrix or calibration matrix. The data reduction equation which calculates the Mueller matrix from irradiance measurements in the most general case is given by,

\[
\mathbf{M} = \mathbf{W}^{-1} \mathbf{I}
\]  

(3.7)

where \( \mathbf{W}^{-1} \) is the pseudoinverse of the matrix \( \mathbf{W} \). For \( Q = 16 \) linearly independent measurements, \( \mathbf{W} \) is a 16x16 square matrix and hence \( \mathbf{W}^{-1} \) simplifies to \( \mathbf{W}^{-1} \), provided \( \mathbf{W} \) is nonsingular. The inverse of \( \mathbf{W} \) can now be uniquely determined and hence all the elements of the Mueller matrix can be uniquely determined. However, random errors propagate to the determined Mueller matrix when merely 16 measurements are performed. For \( Q > 16 \) measurements, \( \mathbf{W} \) is a rectangular matrix and the inverse of \( \mathbf{W} \) is not unique and the pseudoinverse provides the optimal least squares solution. The least squares approach minimizes the effect random error has on the determined Mueller matrix. The matrix \( \mathbf{W}^{-1} \) is termed the Data Reduction Matrix (DRM) and serves as a system transfer function for the polarimeter.

In an ideal case, the DRM can be constructed from the knowledge of the Mueller matrices of the polarizers and retarders in the PSG and PSA, and the rotation states of the retarders at which \( Q \) measurements are made. However in practice, the polarizers and retarders in the PSG and PSA are not ideal. Certain non-ideal parameters can be modeled and these systematic errors can be estimated from a calibration. These estimated parameters are used to construct the optimal DRM for the polarimeter. A well-constructed DRM produces a well-conditioned Mueller matrix leading to a good estimation of the polarization properties.
3.5 Systematic Errors and Calibration

Non-ideal polarization elements and errors in the alignment of the polarization axes of these elements are the major causes of systematic errors in polarimetry. Some of the most common defects found in real polarization elements have been listed by Chipman [3]. It may not be possible to model some of these defects in the Stokes-Mueller formalism. These systematic errors, not taken into account, lead to the measured Mueller matrix of the sample differing from its actual Mueller matrix. The aim of the calibration process is to identify and estimate the systematic errors in a polarimetric setup so that an optimal DRM for the polarimeter can be constructed. One approach for achieving this is to have the polarization elements used in the polarimeter characterized using another well calibrated polarimeter. This method is direct but not very practical without an instrument such as the one developed here.

Another technique is to characterize the polarization elements using the same polarimeter conducting measurements on a polarization standard with a known Mueller matrix (for example, the 4x4 identity matrix of air for a polarimeter designed for transmission mode measurements). This technique allows one to encode and retrieve the systematic errors in the system using mathematical models, and is aptly called “self calibration.” The idea is to iteratively reduce alignment errors using error estimations from repeated calibration trials. When the systematic errors (specifically alignment errors) are optimized (or minimized), the calibrated values for the PSG and PSA elements \[ \tilde{S}_q = R^{(q)}_1(\theta) P_1 \tilde{S}_{\text{in}} \text{ and } \tilde{A}_q = P_2 R^{(q)}_2(5\theta) \] can be used to construct the optimal DRM, as described in the previous section. This DRM can be subsequently used to convert the measured irradiance modulation of a sample to the Mueller matrix of the sample.
Figure 3.3 Dual rotating retarder polarimeter depicting systematic errors

The following sections describe analytic and numeric techniques employed in the calibration of broadband spectropolarimeters.

### 3.5.1 Analytical Calibration Technique

Goldstein [30,31] and Chenault [32] identified five systematic errors that are significant in the case of broadband dual rotating retarder spectropolarimeters. Figure 3.3 depicts the 5 systematic errors, namely, the actual magnitude of retardances ($\delta_1$ and $\delta_2$) and errors in the orientation of the fast axes ($\varepsilon_3$ and $\varepsilon_4$) of the achromatic linear retarders $R_1$ and $R_2$, respectively, along with error in the orientation of the transmission axis ($\varepsilon_5$) of the polarizer $P_2$. Note that the error in orientations of $R_1$, $R_2$ and $P_2$ is defined with respect to the orientation of the transmission axis of $P_1$.

Here, the polarizers in the PSG and PSA are assumed to be ideal, having a diattenuation equal to one. Allowing the diattenuation of the polarizers to vary from 0.98 to 1.0, the percentage RMS error in the estimated Mueller matrix is calculated. This simulation is shown plotted in Figure 3.4. Typical prism polarizers have an extinction ratio of $1:10^5$. This relates to a diattenuation of 0.99998 and the corresponding RMS
error is less than 0.1%. For comparison, a 2° error in the orientation of the PSA polarizer produces a RMS error of 3% while a 2° error in the orientation of the PSG and PSA retarder produces a RMS error of 5% and a 10° difference in the magnitude of PSG and PSA retardance produces a RMS error of 14%. Hence the polarizers may be assumed to be relatively ideal.

The model equation of such a polarimeter with the systematic errors enumerated above is given by,

$$\tilde{\mathbf{S}}_{\text{out}}^{(q)} = P_2(\varepsilon_5)R_2^{(q)}(\delta_2, 5\theta + \varepsilon_4) \mathbf{M} R_1^{(q)}(\delta_1, \theta + \varepsilon_3)P_1 \tilde{\mathbf{S}}_{\text{in}}.$$  \hfill (3.8)

Air (or vacuum) does not have any significant polarization (diattenuation, retardance and depolarization) characteristics over the entire electromagnetic spectrum and hence the corresponding Mueller matrix at each wavelength is a 4x4 identity matrix. Air can be considered a standard sample in Equation 3.8 for $Q \geq 16$ irradiance.

Figure 3.4 RMS error due to non-ideal PSG and PSA polarizer
measurements made by rotating the PSG and PSA retarders in a 1:5 ratio, as discussed earlier. The first retarder is rotated in steps of 6° while the second retarder is rotated in steps of 30°. Performing 61 such measurements causes the first retarder to rotate from 0° to 360°. The irradiance modulation measured by the detector is periodic and has the five systematic errors encoded in its frequency spectrum. The irradiance modulation can be written down in terms of the Fourier coefficients as,

$$I(\theta) = A_0 + \sum_{n=1}^{12} (A_n \cos(2n\theta) + B_n \sin(2n\theta))$$

The 1:5 ratio rotation of retarders causes the systematic errors to be encoded into the first 12 harmonics of the Fourier decomposition. Since the off-diagonal elements of the Mueller matrix during calibration are equal to zero, all odd Fourier coefficients ($A_n$ and $B_n$ where $n = 1, 3, 5, ...$) become zero. Also, since the diagonal elements in this case equal one, the coefficients of the twelfth harmonic ($A_{12}$ and $B_{12}$) vanish [33]. Once the Fourier decomposition is performed on the irradiance modulation, the five systematic errors ($\delta_1, \delta_2, \epsilon_3, \epsilon_4$ and $\epsilon_5$) can be calculated based on the Fourier coefficients ($A_n$ and $B_n$) using analytical expressions derived by Chenault, et al. [34].

Figure 3.5 depicts the normalized irradiance modulation for the ideal case of zero systematic errors and the case where the systematic errors are chosen to be $\delta_1=75^\circ$, $\delta_2=120^\circ$, $\epsilon_3=5^\circ$, $\epsilon_4=-3^\circ$ and $\epsilon_5=4^\circ$. $I_{\text{mod}}$ corresponds to the ideal error-free case and $I_{\text{mod}}$ (error) corresponds to the modulation in the presence of systematic errors. Figure 3.6 is the Fourier analysis performed on the irradiance modulations shown in Figure 3.5. $A_n$ and $B_n$ are the Fourier coefficients of $I_{\text{mod}}$ while $A_n$ (error) and $B_n$ (error) are the Fourier coefficients of $I_{\text{mod}}$ (error). The analysis stated above regarding the Fourier coefficients can be easily observed in Figure 3.6.
Figure 3.5 Normalized irradiance modulations with and without systematic errors

Figure 3.6 Fourier analysis of irradiance modulations with and without errors
3.5.2 Numerical Calibration Technique

Mathematically, 16 independent measurements are sufficient to estimate the 16 unknowns (elements of the sample’s Mueller matrix). However, oversampling is done to statistically minimize random errors in the measurement process. As can be seen in Figure 3.7, RMS error decreases with increasing number of samples. For this simulation, an ideal DRM was calculated with absolutely zero error. Random errors were introduced into the irradiance modulation of air (with varying sample points) having zero systematic errors and inverted to calculate the Mueller matrix. RMS error in the Mueller matrix is calculated and the entire process is repeated 1000 times to provide the statistics seen in Figure 3.7. For each box plot, the central red line is the median, the edges of the box are the 25th and 75th percentiles, the whiskers extend to the most extreme data points not considered outliers, and the outliers are plotted individually in red.

![Figure 3.7 Decreasing RMS error with increasing number of samples](image)

Figure 3.7 Decreasing RMS error with increasing number of samples
Oversampling also allows numerical techniques to be employed for calibration purposes using least square fitting. Nonlinear regression analysis is performed by defining the model function using the Stokes-Mueller formalism as in Equation 3.8 and assigning the defined 5 systematic errors along with any additional parameters as the coefficients to be determined as a result of performing the least square fitting.

This work uses readily available MATLAB functions for performing nonlinear regression, which is based on the Levenberg-Marquardt technique [35]. Seed values for the coefficients along with lower and upper bounds must be specified in order to constrain the regression coefficients to physically meaningful values. However, constraining the coefficients with lower and upper bounds may coerce the regression to converge to a local minimum rather than a global minimum.

### 3.6 Post-processing in Polarimetry

The Mueller matrix that is computed from the DRM may not readily reveal all the polarization characteristics of the sample under consideration. In such a case, post-processing becomes necessary in order to glean meaningful polarization information. This section briefly discusses the post-processing techniques adopted in the current research work.

#### 3.6.1 Physically Realizable Mueller Matrices

Not every 4 element vector composed of real numbers is a physical Stokes vector. The Stokes parameters are related as,

\[
S_0 \geq \sqrt{S_1^2 + S_2^2 + S_3^2},
\]  

(3.10)
Similarly, the elements of the Mueller matrix, transforming an input Stokes vector to an outgoing Stokes vector, is bound by constraints such that not every 4x4 matrix composed of real numbers is a physically realizable or meaningful Mueller matrix. Experimental noise may push the measured Mueller matrix into the nonphysical regime even for a well calibrated polarimeter. This presents mathematical challenges in processing the Mueller matrix; hence a post-processing technique is necessary to compute the “closest” physically realizable Mueller matrix to the estimated nonphysical Mueller matrix.

Cloude [22] and Anderson and Barakat [21] have discussed in detail the necessary and sufficient conditions for a Mueller matrix to be physically realizable. The closest physical Mueller matrix $M_{\text{phy}}$ to a given nonphysical Mueller matrix $M_n$ is determined such that,

$$\|M_{\text{phy}} - M_n\| = \min\{\|M - M_n\|, \forall M \in \mathcal{M}\},$$

(3.11)

where $\mathcal{M}$ is the set of all physical Mueller matrices and the operator $\|\ldots\|$ represents the Frobenius norm of a matrix. The Frobenius norm of a matrix $A$ is defined as,

$$\|A\| = \sqrt{\sum_{i=1}^{m} \sum_{j=1}^{n} |a_{ij}|^2} = \sqrt{\text{trace}(A^*A)},$$

(3.12)

where $a_{ij}$ is the element in the $i^{th}$ row and $j^{th}$ column of the matrix.

The algorithm outlined by Lu [36] can be used to estimate the closest physical Mueller matrix to a given nonphysical Mueller matrix. Accordingly, a complex valued Hermitian matrix called the N-matrix is constructed from the elements of the Mueller matrix. Since, the N-matrix of a nonphysical Mueller matrix has negative real eigenvalues, Cloude [22] suggested dropping the contribution associated with the negative eigenvalue and constructing the new N-matrix using only nonnegative
eigenvalues and their associated eigenvectors. The physical Mueller matrix can then be determined using the elements of the new N-matrix. This estimate was pointed out by Anderson and Barakat [21] to be the closest physical Mueller matrix with respect to the Frobenius norm.

3.6.2 Mueller Matrix Decomposition

The physically realizable Mueller matrix is still not insightful with regard to all the physical polarization properties of the sample. In order to glean meaningful polarization properties, the Mueller matrix can be decomposed into three unique constituent matrices using polar decomposition, as proposed by Lu and Chipman [37]. The decomposition for a nonsingular Mueller matrix $M$ can be written as,

$$M = M_{Dep}M_RM_D,$$  \hspace{1cm} (3.13)

where $M_{Dep}$ is the Mueller matrix of a pure depolarizer, $M_R$ is the Mueller matrix of a pure retarder and $M_D$ is the Mueller matrix of a pure diattenuator. The procedure for performing this decomposition as well as extracting the polarization properties such as linear and circular retardance, linear and circular diattenuation, depolarization index, linear and circular polarizance, extinction ratio and the orientations of the linear retardance and diattenuation, can be found discussed in detail by Lu [36].

3.7 Polarimetric Measurement Procedure

The polarimetric measurement procedure starting from aligning and calibrating the polarimeter to determining the Mueller matrix and polarization characteristics of the sample is summarized by means of a flowchart in Figure 3.8.
Figure 3.8 Flowchart of the polarimetric measurement procedure
3.8 Ellipsometry – Basics

Since this dissertation deals with measurements in reflection and ellipsometric calculation to yield optical constants of the sample under study, the basics of ellipsometry are introduced. Ellipsometry [20,25] is an optical technique that uses the change in the state of polarization of reflected light for the characterization of surfaces, interfaces and thin films. It is the preferred noncontact technique for the measurement of the complex refractive index and thickness of thin films. Figure 3.9 depicts an example of the change in the state of polarization upon reflection from a surface.

![Diagram of linearly polarized input state and elliptically polarized output state](image)

Figure 3.9 Elliptical state of polarization reflected from surface

A collimated beam of light with known (linear) state of polarization is obliquely incident on a sample and the (elliptical) state of polarization of the reflected beam of light is analyzed. A compensator and an analyzer at the detection end enable the measurement of the elliptical polarization. The ratio of the complex reflection coefficients of the sample for the two incident orthogonal states of polarization \(R_p\) and \(R_s\) can be related to
the structural and optical properties of the sample. The component of electric field parallel to the plane of incidence is p-polarized while the component of electric field perpendicular to the plane of incidence is s-polarized. The ratio of complex reflection coefficients can be written as,

\[ \rho = \frac{R_p}{R_s} = \tan \psi \, e^{i \Delta}, \quad (0 \leq \psi \leq 90^\circ, 0 \leq \Delta < 360^\circ) \quad (3.14) \]

where \( \tan \psi = |R_p|/|R_s| \) is the relative amplitude attenuation and \( \Delta = \arg (R_p) - \arg (R_s) \) is the differential phase shift between the p and s linearly polarized components upon reflection. The above complex ratio depends on the angle of incidence (\( \theta \)), the wavelength of light (\( \lambda \)), the complex refractive index \([n(\lambda) + i \, k(\lambda)]\) of the thin film and the thickness (d) of the thin film. Typically, the angle of incidence and the wavelength of light are known, and hence,

\[ \rho = \tan \psi \, e^{i \Delta} \equiv f(n, k, d). \quad (3.15) \]

Equation 3.15 is the fundamental equation of ellipsometry. The optical constants and thin film thickness can be determined when the electromagnetic theory of reflection is applied to the appropriate physical model along with the measured values of \( \psi \) and \( \Delta \).

Different instrumentation techniques for measuring the ellipsometric parameters (\( \psi \) and \( \Delta \)), the physical models of different surfaces, substrates, thin films, etc., and other ellipsometric topics are described in detail in references [38–41]. Generalized ellipsometry or Mueller matrix ellipsometry measures all the polarization properties of the sample by measuring the complete Mueller matrix of the sample [42–44]. The polarimetric system developed as part of the dissertation is a Mueller matrix polarimeter in reflection mode which parallels the operation of a Mueller matrix ellipsometer.
3.9 Conclusions

In this chapter a background of different polarimetric architectures including recent advances in the field was provided. The theory of polarimetry as well as the calibration and data reduction techniques for the specific case of a dual rotating retarder was discussed. With the advent of faster computer processing and the possibility that numerical techniques can easily account for many more systematic errors, it may be preferred over analytical calibration techniques.

Post-processing techniques such as estimating the closest physical Mueller matrix to any experimentally determined Mueller matrix, and decomposition of that Mueller matrix to calculate the polarization characteristics of the sample were discussed. The polarimetric measurement procedure was then outlined by means of a flowchart. Finally, ellipsometric concepts were introduced and the fundamental equation of ellipsometry was discussed.

The subsequent chapters deal with the development of the broadband polarimeter, its testing, measurements and results.
CHAPTER 4

UV-VIS-NIR MUELLER MATRIX POLARIMETER

4.1 Introduction

The Applied Microparticle Optics and Radiometry (AμOR) laboratory at the University of Alabama in Huntsville (UAH) has in the recent past developed a Mueller matrix polarimeter built around a Thermo Nicolet Nexus 870 FTIR spectrometer [45]. This instrument operates in the 3333 to 833 cm\(^{-1}\) waveband at a resolution of 4 cm\(^{-1}\) with high accuracy. In terms of wavelength, the operating spectral region corresponds to the 3 \(\mu\)m to 12 \(\mu\)m region and the resolution varies from 3.6 nm to 57.6 nm for the given spectral region. This polarimeter was successfully used to study optical components and biological samples by Krishnaswamy [45].

The above Fourier Transform Infra-Red SpectroPolarimeter (FTIR-SP) is now complemented by the present system. The new Mueller matrix spectropolarimeter (MMSP) developed from the ground up operates seamlessly in the 300 nm to 1100 nm wavelength region at a resolution 2 nm. The system can be configured in different incarnations: transmission, reflection, back-scattering, etc. The MMSP in transmission mode will be referred to as MMSP-T and in reflection mode as MMSP-R. This chapter
will discuss the design, development, calibration, testing and validation of the new MMSP briefly described in the conference proceeding [46].

4.2 Design Goals

The development process of the MMSP was dictated by certain performance specifications and operational constraints. The following outlines the specifications and constrains that were considered in the design stage of the polarimetric system.

1. The MMSP is to be developed from the ground up. The degrees of freedom that this provides complicate the process of specifying the optical, mechanical, electrical/electronic components to be used in the designing and development of the polarimeter.

2. The components chosen in the design of the MMSP should enable seamless operation in the UV-VIS-NIR spectral region in such a manner that the broadest operational waveband is made possible. This implies that the design should operate as deep into the UV and NIR as possible considering optical constraints. The MMSP should also operate at a high resolution over the entire spectral region.

3. The MMSP design should be such as to lend itself to various measurement configurations such as transmission, reflection, scattering, etc. This may be achieved using computer-controlled rotation stages or manual realignment.

4. The MMSP should operate with the highest achievable accuracy and precision. The repeatability and reproducibility of the MMSP must be good.
5. The user interface for setting operational parameters and data acquisition should be simple and flexible. Apart from the automated scanning option, the interface should accommodate features for performing diagnostics on the MMSP.

4.3 Instrument Design – Transmission Mode

Figure 4.1 depicts a schematic of the Mueller matrix spectropolarimeter in transmission mode (MMSP-T). The MMSP-T is composed of four major subsystems, namely, the optical subsystem, the motion control subsystem, the electronic subsystem and the software subsystem. The performance specifications of optical and opto-mechanical components provided by the manufacturer are listed in Appendix C.

![Figure 4.1 Schematic of the Mueller matrix spectropolarimeter in transmission mode](image-url)
4.3.1 Optical Subsystem

The core of the optical subsystem is the dual rotating retarders and fixed polarizers that constitute the PSG and PSA stages of the system. The polarizers P1 and P2 are Rochon prism polarizers from United Crystals [35]. These polarizers are constructed using two optically contacted pyramid-shaped prisms made of $\alpha$-Barium Borate ($\alpha$-BBO) with their axes aligned perpendicular to each other. The $\alpha$-BBO material is transparent in the 200 nm to 5000 nm spectral region and the polarizer is specified to have an extinction ratio of $1 \times 10^5$. The ordinary ray travels straight while the extraordinary ray is split by an angle that is dependent on the wavelength of light. The retarders R1 and R2 are double Fresnel rhomb quarter wave retarders from Artifex Engineering made of two optically contacted fused silica rhomb prisms [47]. Instead of using a birefringent material to produce the intended phase shift, the rhomb prism produces the phase shift as a result of four total internal reflections occurring within the prism. The retardance due to total internal reflection causes the retarder to be fairly achromatic in the broad 200 nm to 2000 nm spectral region.

A 150W UV-enhanced Xenon lamp having a usable wavelength range of 200 nm to 2500 nm provides the required light in the system. The broadband output of the Xe lamp is coupled into a grating based Oriel Cornerstone 260 monochromator [48]. This monochromator is capable of spanning a spectral range of 180 nm to 2400 nm at a resolution of 0.1 nm and has a focal ratio of f/3.9. Order sorting filters are used between the Xe lamp and the monochromator to prevent contamination of the chosen operating wavelength by different wavelengths with higher orders that satisfy the grating equation. The monochromator has a wavelength accuracy of 0.35 nm and this was verified by
measuring several arc lamp discharges and comparing them to the spectroscopic emission peaks provided in the CRC Handbook of Chemistry and Physics [49].

At the detector end of the system, a UV enhanced Silicon photodiode (PD) from Thor Labs having a responsivity in the 200 nm to 1100 nm region was selected [50]. This detector has an active area of 63.6 mm$^2$, a maximum dark current rating of 1.0 $\mu$A and a NEP rating of $9.1 \times 10^{-14}$ W/Hz$^{1/2}$.

Off-Axis Parabolic (OAP) mirrors are used for beam steering purposes. The OAP mirrors are made of Aluminum with a Magnesium Fluoride (MgF$_2$) coating which is highly reflective in the spectral region of interest. OAP1 is used to collimate the divergent output from the monochromator and couples it into the PSG while OAP2 focuses the collimated beam exiting from the PSA onto the Si photodiode. The OAP1 has an Effective Focal Length (EFL) of 8 inches and OAP2 has an EFL of 2 inches.

The overlap in the spectral response of all the above optical components ranges from 200 nm to 1100 nm. However, depending on the configuration of the system and the sample being measured, the operable spectral range may be reduced to 300 nm to 1100 nm since the throughput and the Signal to Noise Ratio (SNR) may be lower in the 200 nm to 300 nm region. The polarimeter operates best in the 300 nm to 1100 nm waveband.

4.3.2 Motion Control Subsystem

The retarders R1 and R2 were mounted on rotation stages from Newport which are driven by high torque DC servo motor drivers. These rotation stages provide an angular resolution of 0.0005°, an absolute accuracy of 0.023° and a maximum speed of
80°/s [51]. The LabVIEW drivers provided by Newport were integrated to design the necessary motion sequences of the retarders.

### 4.3.3 Electronic subsystem

In order to improve the overall SNR of the system, an optical chopper (OC) from Thor Labs [52] was used in tandem with a lock-in amplifier from Stanford Research Systems [53] and a Si photodiode. The optical chopper is set between the monochromator and the beam steering mirror OAP1. The optical beam was chopped at a frequency of 977 Hz (a prime number) so that there is no interference from any unwanted electrical signal or its harmonics. The analog lock-in amplifier is capable of measuring AC signals as small as nanovolts in the presence of much larger noise levels.

The voltage output from the lock-in amplifier is digitized using National Instruments data acquisition board (DAQ) [54] before it is fed to the computer. The DAQ has a 16 bit resolution with a maximum sampling rate of 200 kS/s.

### 4.3.4 Software Subsystem

Motion control and data acquisition are achieved using a unified user interface built on the LabVIEW platform utilizing the drivers provided, along with the monochromator and rotation stage’s motion controller. Apart from being able to set all the measurement parameters and starting the automated measurement process, the polarization irradiance modulation is displayed at each wavelength that is stepped through. Also, navigating to specific wavelengths, adjusting the monochromator slit width, rotating the retarders to specific position, etc. is allowed, which can be a useful feature in alignment, diagnostics and optimization of throughput. The acquired data is
automatically saved on the National Space Science and Technology Center (NSSTC) server.

Post-processing of acquired data is another part of the software subsystem and uses the algorithms outlined in Chapter 3 to perform calibrations to determine the systematic errors, construct the DRM, calculate the Mueller matrix of the sample and finally determine the polarization properties of the sample. The post-processing algorithms are coded in MATLAB. All the numeric results and plots are automatically saved in appropriate folders on the NSSTC server.

4.4 Instrument Design – Reflection Mode

Figure 4.2 depicts the schematic of the Mueller matrix polarimeter in reflection mode (MMSP-R). The subsystem designs discussed in the previous section remain the same for this configuration. The only difference is that the light from the PSG is incident on the sample at an angle of $\Phi = 70^\circ$ for the MMSP-R. A $70^\circ$ angle is chosen so as to equal the angle of incidence of conventional ellipsometers [38].
4.5 Optical Layout and Polarimetric Alignment

The optical layout begins with coupling the broadband light from the Xe lamp to the monochromator, followed by the collimating OAP1 mirror, and then by the focusing OAP2 mirror and the Si photodiode. The optical chopper and lock-in amplifier are set up so as to be able to maximize the signal when throughput is low. Thereafter, the four polarization elements P1, P2, R1 and R2 must undergo meticulous alignment. The polarimetric alignment of these elements is very crucial and has low tolerance for error. The procedure to achieve such alignment is as follows:
1. The transmission axis of the linear polarizer in the PSG (P1) defines the coordinate system for the rest of the polarization elements in the polarimeter. In this case the transmission axis is set to be vertical.

2. The PSA polarizer (P2) is optically aligned into the system and the transmission axis is rotated precisely until a minimum reading is recorded by the detector. The transmission axis of P2 is then orthogonal to that of P1.

3. The PSA retarder (R2) is introduced into the system and its fast axis is aligned with the transmission axis of P1 by rotating the retarder until the detector signal is minimized.

4. The PSG retarder (R1) is aligned exactly like R2.

5. Finally, P2 is rotated precisely 90° such that the transmission axis of P2 is aligned with that of P1. Now, the detector reads a maximum.

Once the preliminary alignment is completed, a calibration run is performed. The best polarimetric alignment is achieved by iteratively adjusting the orientations of the axes based on the systematic errors estimated during the calibration process such that the estimated errors are minimized.

Figure 4.3 is a photograph of the Mueller matrix polarimeter in transmission mode (MMSP-T). Figure 4.4 is a close-up view of the PSG and PSA. Figure 4.5 is a photograph of the Mueller matrix polarimeter in reflection mode (MMSP-R). Figure 4.6 is a snapshot of the user interface used to control the MMSP and acquire data.
Figure 4.5 Photograph of the Mueller matrix spectropolarimeter in reflection mode

1. PSG Polarizer
2. PSG Retarder
3. Sample Holder
4. PSA Retarder
5. PSA Polarizer
6. OAP Mirror
7. Si Photodiode
4.6 Performance Issues

There are a few issues that impede the optimal performance of any Mueller matrix polarimeter. These issues are explained and the solution, if any, adopted in this investigation to circumvent the problems are presented below:

1. The light exits the monochromator through a slit. Achieving perfect collimation (negligibly small divergence or convergence) with an extended source is not possible. Hence, a pinhole was placed at the exit slit of the monochromator which improved the collimation significantly. However, this was at the cost of reduced signal.
2. The Xe lamp has sharp spikes in its irradiance spectrum. At times this causes the detector in the lock-in amplifier to saturate. In order to avoid this, the slit width of the monochromator is decreased in steps until the detector reading is below a predetermined threshold. The threshold is based on intuition or a prior trial run.

3. The one drawback of choosing the Fresnel rhomb retarders is that they have an unusual aspect ratio. For a clear square aperture of 5 mm, the V-shaped prism is 140 mm long, and housed in a solid aluminum cylinder of 50 mm diameter and 140 mm length. It is an arduous exercise to align the retarder such that light enters and exits the prism without any clipping as the retarder is rotated through 360°.

4. The measurement scheme is such that the monochromator steps through a range of wavelengths and at each wavelength the retarders are rotated and 61 measurements performed at distinct orientations of the retarders. This is an extremely time consuming process where at every wavelength the system takes over 2 minutes for performing the measurements.

5. Toxic ozone is produced by the photochemical reaction between UV radiation from the Xe lamp and oxygen in air. This is a health hazard and the ventilation port available in the lab is used to blow away the lamp’s exhaust air.

### 4.7 Calibration and Validation

The MMSP is first calibrated and then the results of selected samples are validated against independently measured optical components or theoretically expected
values. This section provides the results of the calibration and validation for transmission and reflection.

4.7.1 Transmission Mode

A series of calibration measurements are performed followed by iteratively fine tuning the alignment of the axes of the polarization elements in order to minimize the alignment errors. The systematic errors are calculated using the Fourier decomposition technique discussed in Chapter 3. Figure 4.7 plots the estimated systematic errors. The three plots on the top row of Figure 4.7 correspond to the error in the alignment of R1, R2 and P2 respectively. As can be seen, the error in alignment of the three polarization elements is very close to 0°. Any alignment error that appears to be dependent on wavelength is likely due to crystal defects causing the crystal axis to depend on wavelength. The plots on the bottom row correspond to the magnitude of the retardance of R1 and R2, and the percentage difference in the measured intensity between the measurements made when the retarders are aligned at (0°, 0°) and at (360°, 360°). The magnitude of retardance of R1 and R2 are expected to be identical. The difference in the magnitudes is likely due to the difference in the angle of the V-shaped prism, since retardance due to total internal reflection depends on the incident angle. The manufacturer’s tolerance on the prism angle is not known. However, it is clear that even a small difference in the prism angle is capable of producing a significant difference in magnitude of retardance since the overall retardance is produced by 4 total internal reflections within the prism. The intensity difference plot is used as a diagnostic to look for any unusual spikes or drifts.
As described in Chapter 3, the estimated systematic errors are used to construct the DRM and thereafter the Mueller matrix can be calculated. Figure 4.8 is a plot of the normalized Mueller matrix where each element of the matrix is substituted by the spectral equivalent. Note that the diagonal elements of the matrix are close to 1 and the off-diagonal elements are close to 0. This correlates well with the expected 4x4 identity matrix for air.
Figure 4.8 Mueller matrix spectrum of air

Figure 4.9 is a plot of the above Mueller matrix with relaxed y-axes. This plot allows one to see the variation in the matrix elements and the deviation from the expected values.
Figure 4.9 Close-up view of the Mueller matrix spectrum of air

Figure 4.10 is a plot of the accuracy and precision in the Mueller matrix measurement for air as a function of wavelength. The percentage accuracy is defined in Equation 4.1, where \( I \) is a 4x4 identity matrix. The percentage precision is defined in Equation 4.2, where \( \text{std}(M_{k}(i,j)) \) is the standard deviation in element \((i,j)\) of \(k\) Mueller matrices. Three Mueller matrices were used for calculating the precision in Mueller matrix measurement.

\[
\% \text{ Accuracy} \equiv 100 - \left[ 100 \times \frac{\sum_{i=0}^{3} \sum_{j=0}^{3} [M(i,j) - I(i,j)]^2}{16} \right]^{\frac{1}{2}} \tag{4.1}
\]
\[
\text{Precision} \equiv 100 - \left[ 100 \times \frac{\sum_{i=0}^{3} \sum_{j=0}^{3} [\text{std}(M_{k}(i,j))]^{2}}{16} \right]
\]

(4.2)

From Figure 4.10 it can be seen that the measurement precision is over 99.5% in the entire spectral waveband. Also, the measurement accuracy is over 98% in the entire spectral waveband.

Figure 4.10 Accuracy and precision in the Mueller matrix measurement for air

Figure 4.11 is the plot of the polarization properties of air. Note that the retardance, diattenuation, polarizance and depolarization are close to zero. This is due to the fact that air does not possess any polarization-altering properties. The plot shows three successive measurements performed under similar measurement conditions (in red, green and blue). The three measurement results appear nearly identical and the overlaps in the plots indicate good repeatability. The features that can be seen are attributed to instrumental artifacts and small considering the possible range of values for retardance is between 0°
and $180^\circ$ and the possible range of values for diattenuation, polarizance and depolarization is between 0 and 1. The fluctuation in the orientation of the retardance and diattenuation can be ignored since the corresponding magnitude of retardance and diattenuation is very small and the orientation is determined by calculating the inverse tangent of the ratio of two quantities. When the magnitudes of these two quantities are very small, the inverse tangent of the ratio produces values approaching $\pm 45^\circ$ depending on the signs of the two quantities. Practically, the orientation is undefined under these conditions.

Figure 4.11 Polarization properties of air
In order to quantitatively validate the performance of the MMSP-T, measurements were performed on a Meadowlark retarder provided by Polaris Sensor Technologies (PST). The measured Mueller matrix of the retarder is shown in Figure 4.12. The transparency of the retarder made possible measurements from 350 to 1100 nm.

Figure 4.12 Mueller matrix of Meadowlark retarder

Figure 4.13 shows the polarization properties of the Meadowlark retarder. The retarder, as expected, has negligible diattenuation and depolarization. The post-processing algorithm constrains the magnitude of retardance ($\delta$) between 0° and 180° and hence, a
‘peak’ is seen at 550 nm. This folding can be manually rectified by substituting the values for $\delta$ by $360 - \delta$ in the 350 nm to 550 nm wavelength region. Note that the orientation of the fast axis of the retarder is consistently around -22° except close to the limit of 350 nm. The measured linear retardance is compared to that provided by Meadowlark at nine discrete wavelengths.

Figure 4.13 Polarization properties of Meadowlark retarder

Figure 4.14 shows the comparison of the measured and manufacturer specified linear retardance. The measured retardance is plotted in blue with error bars and the retardance values provided by Meadowlark at discrete wavelengths are plotted in red. The measured retardance values in the 510 nm to 700 nm spectral region are in agreement to within a
degree of retardance when compared to the values provided by Meadowlark. There is significant disagreement in retardance values below 480 nm. Given that retardance is inversely proportional to wavelength for a linear retarder, it can thus be inferred that the retardance value provided by Meadowlark at 407 nm is questionable and the retardance values determined by the Mueller matrix polarimeter are more reliable at shorter wavelengths. The Meadowlark retarder was also independently measured by PST using their polarimeter at a few discrete wavelengths. The retardance values measured by PST and by the MMSP-T were in agreement to within a degree of retardance.

Figure 4.14 Comparison of measured and manufacturer specified retardance of retarder
4.7.2 Reflection Mode

As mentioned earlier, a mirror is used in the sample space during calibration in the reflection mode such that light is incident on the mirror at a $70^\circ$ angle. The Mueller matrix of the mirror that goes into the model equation is derived from the complex refractive index and angle of incidence [23]. In addition to the five systematic errors discussed previously, three more errors should be accounted for, namely, the real part of refractive index ($n$), the imaginary part of refractive index ($k$) and the mirror angle (or the incident angle $\theta$). The numerical calibration technique, outlined in Chapter 3, is adopted where the eight parameters are estimated by nonlinear regression analysis by performing least squares fitting.

Figure 4.15 plots the eight systematic errors in addition to the difference in the reference intensity. Note that the mirror angle and the complex refractive index estimations appear to be non-physical. This is despite the fact that the regression was supplied with physical seed values. Constraining the regression parameters with lower and upper bounds returned either of the boundary values as the solution and these parameters produce huge errors in the estimated Mueller matrix. Hence, the regression was not bound by lower and upper values. One possible reason for the non-physical parameter estimation could be the fact that the Magnesium Fluoride (MgF$_2$) coating on the Al mirror is unaccounted for in the model equation used in the regression. Another reason is the presence of random errors that drive the optimization algorithm into non-physical parameter space. However, it can be seen that the non-physicality occurs in the three parameters at shorter wavelengths (less than 600 nm) and it is expected that the non-physicality in one parameter may compensate the effect of the other. Also, certain
artifacts that can be seen in this plot are propagated to subsequent calculations of the Mueller matrix and polarization properties. Despite this fact, the computed polarization properties are expected to be physical in nature and provide reliable qualitative information, if not quantitative.

Figure 4.15 Estimated systematic errors during calibration of MMSP-R

Figure 4.16 is a plot of the Mueller matrix of the Aluminum mirror at a 70° angle used for the calibration. Mueller matrix (MM DRM) calculated from inverting the DRM is plotted in red and the Mueller matrix (MM CALIB) calculated from the calibration parameters [23] (incident angle and complex refractive index) is plotted in blue. The two
plots overlap to a great extent with minor artifacts in the \(\text{MM}_{\text{CALIB}}\). The \(N\)-matrix corresponding to \(\text{MM}_{\text{CALIB}}\) does not have any negative eigenvalues and hence the Mueller matrix is physical and is capable of transforming a physical input Stokes vector to a physical output Stokes vector. Since \(\text{MM}_{\text{CALIB}}\) is physical, the polarization properties calculated from the decomposition of \(\text{MM}_{\text{CALIB}}\) are also expected to be physical. Since ability to estimate physical Mueller matrices and physical polarization properties are our primary concern, the calibration parameters (although non-physical) were used to construct the DRM and subsequently utilized to determine the Mueller matrix and polarization properties of the sample.

Figure 4.16 Mueller matrix of Aluminum mirror at 70° reflection
The normalized Mueller matrix is decomposed and the polarization properties are computed. Figure 4.17 is a plot of the polarization properties of the Aluminum mirror at a 70° angle. Note that, as expected from reflection off metal surfaces, the dominant polarization property of the mirror is linear retardance. The red and the blue plots correspond to two separate measurements performed under similar conditions. Note that the scale for the plots of linear diattenuation, circular retardance, circular diattenuation and depolarization index is small, thus showing apparently significant difference between the measurements. The overlap in the plot of linear retardance demonstrates that the overall correlation between the two sets of measurements is high.

Figure 4.17 Polarization properties of Aluminum mirror at 70° reflection
In order to qualitatively validate the calculated polarization properties, the complex refractive index for Aluminum as a function of wavelength [55] is used in Fresnel’s equations to calculate the complex reflection coefficients for the s and p polarization states. The diattenuation and retardance can be calculated from these coefficients. Figure 4.18 and 4.19 are plots of the diattenuation and retardance of the mirror as a function of wavelength and incident angle (70°±0.5° due to possible misalignment) respectively. The theoretically calculated and experimentally determined diattenuation values are comparably low. However, the theoretically calculated retardance values are much lower than the experimentally determined retardance values. A possible explanation is due to the fact that the Aluminum mirror is coated with Magnesium Fluoride (MgF$_2$) for enhancing the reflection of the mirror in the UV. MgF$_2$ is a birefringent material and contributes to the overall retardance produced by the mirror. Assuming that the MgF$_2$ coating has a thickness of 1µm±0.2µm and the incident angle is 70°±0.5°, the optical path travelled by the light beam is between 5µm and 7.5µm. With the refractive index data for the ordinary and extraordinary rays available [56], the retardance imparted by the MgF$_2$ is shown in Figure 4.20. It can be seen that adding up the retardance due to the reflection and the birefringence of MgF$_2$ gives the same order of magnitude of linear retardance as the experimentally determined retardance.

However, it must be noted that the MgF$_2$ film on the mirror surface is expected to be either in amorphous (non-crystalline) or polycrystalline form (composed of crystallites, each with a random orientation). Such a film is not likely to impart a net birefringence (or retardance). At this point it is not certain if the retardance is a contribution from the MgF2 film or due to an instrumental or a post-processing artifact.
In order to further validate the performance of the system in the reflection mode, measurements are performed on a microscope slide made of borosilicate glass at a 70°
incident angle. Figure 4.21 is a plot of the experimentally measured Mueller matrix of the microscope slide. Figure 4.22 is the plot of the polarization properties of the microscope slide. Note that, as expected from reflection off dielectric surfaces, the dominant polarization property is linear diattenuation.
In order to theoretically verify this, the refractive index of borosilicate is calculated from the dispersion relation and coefficients taken from the Schott glass catalog [57]. Thereafter, the Fresnel reflection coefficients for the s and p polarization states are calculated and subsequently the diattenuation and retardance of the microscope slide. The magnitude of retardance is equal to zero throughout the spectral region and range of incident angle since the imaginary part of the refractive index of the borosilicate material is zero in this spectral region. Figure 4.23 shows the diattenuation as a function of wavelength and incident angle (70°±1° due to misalignment). Note that the theoretically calculated and experimentally determined diattenuation values are roughly equal.
Figure 4.23 Diattenuation of microscope slide as a function of incident angle

4.8 System Limitations

The current system performs well in both transmission and reflection modes. However, transforming the system from one measurement mode to another (transmission to reflection) has been challenging. It does not seem feasible to perform broadband spectral measurements at non-specular angles due to reduced throughput of the system in such configurations.

4.9 Conclusions

This chapter described the design, development, calibration and validation of the MMSP-T and MMSP-R. The MMSP-T has been calibrated and the performance was quantitatively verified to be satisfactory. The MMSP-R has been calibrated and the performance was validated qualitatively, if not quantitatively. The unparalleled characteristics of the MMSP make it a one of a kind instrument.

Since the MMSP is calibrated and characterized, light scattering samples can be measured and polarization properties of such samples can be determined.
CHAPTER 5

POLARIZATION PROPERTIES OF MICROPARTICLE DEPOSITS

5.1 Introduction

The Mueller matrix spectropolarimeter (MMSP-T and MMSP-R) developed in this investigation has been used to successfully measure the polarization properties of several different samples: optical components like polarizers and retarders, electro-optic devices like liquid crystal modulators and magneto-optic Faraday rotators, optically active solutions, polymers in unstressed and stressed conditions for materials characterization, aluminum foil immersed in water for underwater detection, particulate matter (PM) deposits on microscope slides and hemispherical metal nanoparticles deposited on dielectric substrates. Measurements on such a wide range of samples demonstrate the versatility of the polarimetric system.

This chapter focuses on the polarimetric measurements performed on PM deposits on microscope slides in both transmission and reflection modes. Firstly, a measurement on PolyStyrene Latex (PSL) microspheres in the transmission and reflection configurations is presented along with results from theoretical calculations for thin films. Thereafter, measurements of PM deposits are presented in order to explore applications
of this system in discriminating between various subclasses of particles and thus to look for a pathway toward detection of biological agents.

5.2 Measurements on PSL microspheres

In order to gain perspective on the polarization properties of aerosol deposits on microscope slides, it is necessary to first look at the polarization properties of a much simpler system, spheres on a plane, both theoretically and experimentally. For this, PSL microspheres having a nominal diameter of 3 µm were deposited on borosilicate microscope slides for the initial experimental studies. Figure 5.1 is an image from a Zeiss confocal laser scanning microscope of the PSL deposit. Figure 5.2 is a plot of the polarization properties of this deposit. The blue curves in the plot correspond to the measurements made in transmission mode from 300 nm to 1100 nm while the red curves in the plot correspond to the measurements made in reflection mode from 400 nm to 1000 nm.

Figure 5.1 Microscope image of PSL microsphere on microscope slide
The primary polarization properties (diattenuation and retardance) of PSL microspheres in transmission mode are negligibly small. Also, the depolarization is similar in magnitude and comparable to a blank microscope slide implying that the PSL microspheres on the microscope slide do not significantly contribute to the depolarization. As can be verified from Mie theory [12], forward scattering from spheres do not have significant polarization characteristics. However, in reflection mode, PSL microspheres display significant spectral features in the linear retardance, linear diattenuation and depolarization.
In order to attempt to understand the experimental results, theoretical calculations are considered for comparison. A simple model of multiple reflections from a uniform PSL film of 3±0.25 µm thickness on a microscope slide was employed. A 0.25 µm variation in the film thickness was arbitrarily chosen in order to suppress the strong modulation in the curves that are due to interference phenomena. The refractive index of PSL is from Kasarova [58]. Fresnel reflection and transmission coefficients between the air – film interface and film – slide interface for a 70° incident angle were used to construct the reflection coefficient due to multiple reflections. Thereafter, the diattenuation and retardance experienced by the reflected light was calculated. Figure 5.3 is a plot of linear retardance due to the PSL film while, Figure 5.4 is a plot of linear diattenuation due to the PSL film.

![Figure 5.3 Linear retardance due to PSL film at 70° reflection](image-url)
The difference between the theoretical and measured values of diattenuation and retardance is due to the oversimplified model used in the theoretical calculations. The electromagnetic scattering models simulating sphere clusters on a plane are very complex [59] and beyond the scope of this dissertation. The solution to the problem of electromagnetic scattering by an ensemble of spheres is very involved [60,61].

The polarimetric signatures that can be seen in the measured polarization properties compared to the featureless theoretical calculations are of interest. These are the types of signatures that we look for in subsequent measurements of scattering materials of different morphologies.

5.3 Interpreting Polarization Properties of Scattering Samples

In order to understand the measured polarization properties of scattering samples such as PSL microspheres, consider Figure 5.5. In the absence of the PSL microspheres,
the microscope slide reflects part of the incident light and the polarization state of the reflected light is dependent on the polarization state of the incident light, the angle of incidence and the optical constants of the slide at a given wavelength through the Fresnel coefficients. The reflected polarization state can be determined by direct measurements. However, when there are scattering elements such as PSL microspheres on a slide, interpreting the polarization property of the sample system is complicated.

Figure 5.5 Schematic of the scattering process

The light incident on the scattering elements scatters light in $4\pi$ steradians and the polarization state of light at each scattering angle is different and dependent on the size of the scattering element and the complex refractive index ascribed to the scattering element at the particular wavelength of interest. The scattered light in the upper hemisphere is denoted by black arrows in Figure 5.5 and the component of the scattered light along the direction of the specular reflection from the surface of the slide is denoted by red arrows. Part of this latter scattering is directed into the slide, as is part of the incident beam and is denoted by grey arrows. This transmitted component may undergo multiple reflections within the slide and interact with the PSL microspheres multiple times (not depicted in
Figure 5.5). Additionally, the forward scattering component from the PSL microspheres will undergo a reflection from the slide surface and will roughly align with the specular reflection of the slide, as is denoted by blue arrows. Whether or not this component contributes to the measurement made at the specular reflection direction depends on the angle of incidence, and the areal density of the scattering elements. A higher areal density of scattering elements also leads to inter-particle scattering which is a complex phenomenon and is denoted by horizontal arrows. For low areal densities, the components associated with second order scattering by PM may be too weak to reveal any significant polarization features against the background of the strong specular reflection leaving the slide.

However, the measured polarization state of the sample in comparison to that of the blank slide may be interpreted as the cumulative polarization state that is “lost” or “taken away” from the slide due to the angular distribution of polarization dependent scattering from the PSL microspheres present. Similar to the radiometric quantity Total Integrated Scatter (TIS), the current polarization measurement or property may be interpreted as the Total Integrated Polarized Scatter (TIPS). The correlation between this measurement and the standard polarized Bidirectional Reflection or Scattering Distribution Function (pBRDF or pBSDF) is not known at this point and may be an interesting subject for future research. It must also be noted that the measurement of pure scattering at a non-specular reflection angle was not possible due to poor SNR. Also, the fact that a heavily loaded sample at the specular reflection angle (specular component itself absent due to the heavy loading) was immeasurable due to poor SNR, reinforces the TIPS interpretation.
5.4 Morphology of Subject PM and Sample Preparation

The range of PM samples on which measurements were performed include kerosene soot, two soil samples, three different species of pollen and *Bacillus subtilis* (BG). BG is a well known surrogate for anthrax spores and has been extensively used in research. The soot, soil and pollen samples represent major constituents of the ambient atmospheric aerosol, and can be considered interferants in the bio-aerosol (in this case BG) detection process.

Sample preparation forms an important aspect of performing such measurements since polarization artifacts merely due to sample preparation should be avoided or accounted for before interpreting the polarization features produced by the sample itself. The PSL microspheres were suspended and diluted (for a light loading) in water and samples for performing polarimetric measurements were prepared by drop-coating the microscope slides. Kerosene soot samples were prepared by exposing the microscope slide to a plume of soot from a kerosene lamp and lightly coating the surface of the slide. The byproducts of the combustion process electrostaically adhere to the smooth dry surface of the microscope slide. Soil samples are prepared by mixing the dry soil with water in a beaker and allowing the colloidal suspension to settle for a minute to allow heavier particles to precipitate. The slide was then drop-coated with the the lighter particles remaining in the suspension. Pollen species, being hydrophobic, could not be drop-coated using a water suspension. Hence, a drop of glycerol is smeared on the slide to form a thin layer over which pollen grains are lightly sprinkled. The glycerol film provides the required adhesion. BG samples were prepared by suspending BG in powder form in water to form a solution and drop-coating it on the slide.
Figure 5.6 is a photograph of the prepared samples. Some of the sample may not be readily visible in cases where the loadings are very light. The samples in the photograph from left to right are brown soil (soil collected from a construction site), red soil (soil collected from a baseball field), Nettle (*Urtica dioica*), Ragweed (*Ambrosia artemisifolia*), Pine (*Pinus ponderosa*), kerosene soot, BG and PSL.

The above described techniques for sample preparation do not afford the possibility of accurately quantifying the loading of the sample material on the slide. Also, it was not possible to prepare samples consistently and uniformly. However, the non-uniformity in sample preparation may work advantageous for the intended application since polarization properties as a result of artifacts in sample preparation is avoided. Also, polarimetric measurements performed on such samples and the polarization properties derived therefrom, amply demonstrate the usefulness of using the developed polarimetric system for the detection of bio-aerosols. This will be demonstrated in the following section.

The pollen grain sample’s size and shape are described in Table 5.1. Figure 5.7 depicts the shape, size and appearance of the pollen grains [62–65]. Nettle pollen,
Ragweed pollen and Pine pollen are shown in Figure 5.7(a), (b) and (c) respectively. The image of the Nettle pollen, Ragweed pollen and Pine pollen on microscope slide using a confocal laser scanning microscope is shown in Figure 5.7(d), (e) and (f) respectively. Note that the appropriate scale has been provided in the microscope images in the bottom right corner. Confocal laser scanning microscope images of BG, brown soil and red soil are shown in Figure 5.8(a), (b) and (c) respectively. BG is rod shaped and is only about 5 µm long [66]. Hence its shape is not discernible in the microscope image. The variation in the shape and size of the two soil samples is very large as can be seen in the microscope image. The crystals of mineral deposits can be clearly seen.

Table 5.1 Size and shape description of selected pollen samples

<table>
<thead>
<tr>
<th>Common name</th>
<th>Biological name</th>
<th>Typical size</th>
<th>Shape Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nettle</td>
<td><em>Urtica dioica</em></td>
<td>10µm</td>
<td>Grains spheroidal or irregular, thin walled and collapsing easily</td>
</tr>
<tr>
<td>Ragweed, Short</td>
<td><em>Ambrosia artemisifolia</em></td>
<td>17.6µm - 19.2µm</td>
<td>Grains spheroidal or slightly oblate with a spiky surface.</td>
</tr>
<tr>
<td>Pine, Western Yellow Ponderosa</td>
<td><em>Pinus ponderosa</em></td>
<td>45µm - 65µm</td>
<td>Grains characterized by the possession of two large, conspicuous, air-filled bladders.</td>
</tr>
</tbody>
</table>
5.5 Measurements of Aerosol Deposits

Polarimetric measurements of the PM deposits on microscope slides were performed in both transmission mode and reflection mode. This section compares and contrasts the results produced using the two measurement modes. The feasibility of
utilizing the polarimetric technique for detection of bio-aerosols is explored and presented.

5.5.1 Transmission Mode

The different species of PM were measured in the transmission mode and the polarization property as a function of wavelength as shown in Figure 5.9. The measurements performed on the three species of pollen grains were averaged. Similarly, the measurements performed on the two soil samples were averaged. This was done since the difference in the measurements between the various soil and pollen samples is comparable to the difference between multiple measurements of the same sample.

Figure 5.9 Polarization properties of PM species in transmission mode
Most of the polarization features of the aerosol species examined are negligible or comparable to the measurement made on a blank slide. A possible reason for this is that although forward scattering from scattering particles provide significant throughput for measuring irradiance, it has no significant polarization content. Hence, polarimetric measurements in transmission mode cannot be expected to provide the required polarization features that can be used for differentiating the various aerosol species.

5.5.2 Reflection Mode

The particulate matter species were measured in reflection mode with an incident angle of 70° as described in earlier chapters. Figure 5.10 is a plot of the polarization properties of the two soil samples. As can be seen, the polarization properties for the red soil and brown soil roughly parallel each other.
Figure 5.10 Polarization properties of soil samples in reflection mode

Figure 5.11 is a plot of the polarization properties of the three types of pollen species. Differences can easily be seen between the three pollen samples in one polarization property or the other.
Figure 5.11 Polarization properties of pollen samples in reflection mode

Figure 5.12 is the plot of the polarization properties of all species as a function of wavelength. The polarization properties of the different PM species display a significant amount of differentiating features in the reflection mode. The differentiating features that are seen in these measurements are a result of the differences in the size, shape and optical constants of the scattering material.

The average polarization properties of the three types of pollen along with error bars are plotted in green. The polarization properties for pollen are in stark contrast to the other samples. Although the linear retardance of pollen is comparable to the blank slide (in yellow), the linear diattenuation of pollen gradually decreases from about 0.7 at 400 nm to about 0.2 at 1100 nm. This is in contrast to the linear diattenuation of all the
other PM species, as well as the blank slide. The decrease in linear diattenuation is accompanied by an increase in circular diattenuation although, of much smaller magnitude. Also, the depolarization due to pollen increases from about 0.2 at 400 nm to greater than 0.5 at 1100 nm. The average polarization properties of the two soil samples along with the error bars are plotted in blue. The soil sample displays an increase in the linear retardance from about 4° at 300 nm to about 9° at 1100 nm. The increase also appears to be quite linear in nature. Soot (in black), on the other hand, has a decreasing linear retardance varying from about 8° at 600 nm to about 5° at 1100 nm. The narrower spectral range over which the polarization properties of soot is measured is due to fact that at the shorter wavelengths, the throughput was too low to be able to make reliable measurements or estimations of polarization properties. Finally, BG (in red) has linear retardance that is non-linear over the measured spectral region spanning 300 nm to 1100 nm.
The differences in the polarization properties of the various aerosol species that have been highlighted demonstrate that it may be feasible for the polarimetric technique to be able to discriminate between the different scatterers.

### 5.6 Conclusions

This chapter first presented the measurement of PSL microspheres in transmission and reflection along with theoretical calculations using a simplified model for comparison to the reflection measurement. Polarimetric signatures were clearly seen in the measurements performed in reflection.
The morphology of the particulate matter and the manner in which the samples were prepared was discussed. The measurements performed in transmission mode do not yield any significant polarization properties that can be exploited to discriminate between the different PM species. However, in reflection mode, significant differences in certain polarization properties were discovered, for the first time to our knowledge, which may be utilized for discriminating between the different aerosol species.
CHAPTER 6

MUELLER MATRIX ELLIPSOMETRY OF NANOPARTICLES

6.1 Introduction

Plasma oscillations are setup on the surface of metal nanoparticles and resonance conditions occur when the frequency of incident electromagnetic radiation matches the natural oscillation frequency of the surface electrons against the restoring force of the positive nuclei. These resonances are referred to as Localized Surface Plasmon Resonances (LSPRs). The interaction of electromagnetic radiation with metal nanoparticles (NPs) is of great interest due to the multitude of applications that may result, c.f. [6–8] and references contained therein. A valuable tool for locating the spectral positions of LSPRs is ellipsometry. Ellipsometric measurements determine the amplitude ratio ($\psi$) and phase shift ($\Delta$) between the parallel and perpendicular polarization states. Ideally, the complex refractive index can be calculated from the spectral data for $\psi$ and $\Delta$, and the peaks in $k$ (imaginary component of refractive index) identify the positions and strengths of the LSPRs. Ellipsometric measurements can be performed over a range of wavelengths and incident angles in order to achieve additional insights or a more robust estimate of the optical constants. However, conventional ellipsometers cannot measure polarization properties (such as circular retardance, circular
diattenuation and depolarization) that describe the interaction of electromagnetic radiation with metallic nanoparticle systems. As a result, the polarization properties exhibited at and around LSPR peaks may not be completely understood from conventional ellipsometry alone. Mueller matrix (generalized) ellipsometry may provide additional and vital polarization information on the metal nanoparticle system [9].

The broadband Mueller matrix polarimeter, when configured in reflection mode, can parallel the operation of a conventional ellipsometer. However, the Mueller matrix polarimeter is capable of measuring all 16 elements of the Mueller matrix from which all the polarization properties including depolarization can be calculated.

This chapter includes measurements of hemispherical gallium (Ga) nanoparticle deposits on sapphire substrates as a means of demonstrating the versatility of the developed system and its utility in measuring optical properties of metal nanoparticle systems. This chapter first provides the necessary background of the nanoparticle system, its optical characteristics determined by in situ ellipsometric measurements performed by Duke University (DU) and the complete polarization properties determined from Mueller matrix ellipsometric measurement data.

6.2 Background

The optical properties of metal nanoparticles have been gaining interest in recent years for applications ranging from sensing to enhancement of emission and absorption. Typical metals considered for such studies have been Au and Ag, both of which have LSPR peaks primarily in the visible or IR region. Ga NPs, on the other hand, can be tuned into the deep UV as a result of the high plasma frequency of Ga. Additionally, the
LSPR for Ga NPs have broad tunability, stability across a wide temperature range, excellent plasmon resiliency when oxidized and simplicity of deposition even at room temperatures [7].

Ga NPs were deposited on sapphire substrates by collaborators at DU using a Veeco GEN II plasma-assisted molecular beam epitaxy system equipped with a UVISEL-Jobin-Yvon *in situ* spectroscopic ellipsometer operating at an angle of incidence of 70° from normal in the 1.5 to 6.5 eV photon energy range (corresponding to a spectral range of 190 nm to 827 nm) [8]. The schematic of the above setup housed at DU is shown in Figure 6.1.

The *in situ* ellipsometric data is used to calculate the ratio of the complex reflection coefficients (\(R_p\) and \(R_s\)) of the sample for the two incident orthogonal states of polarization. The ratio of complex reflection coefficients can be written as [20],

\[
\rho = \frac{R_p}{R_s} = \tan \psi e^{i\Delta}, \quad (0 \leq \psi \leq 90^\circ, 0 \leq \Delta < 360^\circ),
\]

(6.1)

where \(\tan \psi = |R_p|/|R_s|\) is the relative amplitude attenuation and \(\Delta = \arg (R_p) - \arg (R_s)\) is the differential phase shift between the \(p\) and \(s\) linearly polarized components upon reflection [25]. The pseudodielectric function [6] which is related to \(\rho\) above can be calculated in real time using the equation,

\[
\langle \varepsilon \rangle = \varepsilon_r + i\varepsilon_i = (n + ik)^2 = n_0^2 \sin^2 \varphi \left[ 1 + \left( \frac{1-\rho}{1+\rho} \right)^2 \tan^2 \varphi \right],
\]

(6.2)

where \(n_0\) is the refractive index of the incident medium and \(\varphi\) is the angle of incidence.
Since the Ga NPs on the sapphire substrate are approximately hemispherical in shape, the asymmetry in the dimension of the NPs and anisotropy with respect to the two different semi-infinite surrounding media causes the LSPR to split into two modes, a low energy peak corresponding to the longitudinal (in-plane) LSPR mode and a high energy peak that corresponds to the transverse (out-of-plane) LSPR mode. Figure 6.2 is a tilted SEM image of the Ga NPs deposited on a sapphire substrate illustrating Ga NP’s truncated spheroid shape [6]. Figure 6.3 is a photograph of the sample with Ga NPs on sapphire substrate. The darker regions on the edges of the sample are the bare sapphire substrate while the rest of the sample has the Ga NPs. Also note that samples are scratched and stained in certain areas.
Figure 6.2 SEM image of the Ga NP on sapphire showing truncated spheroid shape [6]

Figure 6.3 Photograph of sample with Ga NPs on sapphire substrate

The temporal evolution of the pseudodielectric function of the Ga NPs during the deposition process reveals that a resonance feature appears just as the deposition begins. This plasmon peak continuously redshifts with increasing average particle size (as the deposition time increases), as shown in Figure 6.4 [6]. The upper panel in Figure 6.4 is the AFM imagery of three samples with the corresponding size distribution histogram.
below it. The NP size increases from (c) to (a). The plot below is absorption as a function of photon energy. The real-time pseudodielectric data allows the deposition to be monitored so the plasmon resonance can be tuned to specific photon energies.

Figure 6.4 Plot showing redshift in LSPR peaks with increase in average Ga NP size [6]
6.3  Gallium Nanoparticle Sample Characteristics

The three samples measured were labeled A5, A6 and A8. Each had a different
deposition time and hence they have different nanoparticle size distributions. As a result
of the different size distributions, the LSPR peaks occur at different photon energies. The
three samples are characterized as follows:

\[ t_{A6} < t_{A5} < t_{A8}, \]  \hspace{1cm} (6.3)
\[ r_{A6} < r_{A5} < r_{A8}. \]  \hspace{1cm} (6.4)

Here, \( t_{A6} \), \( t_{A5} \) and \( t_{A8} \) are the deposition times for samples A6, A5 and A8 respectively,
while \( r_{A6} \), \( r_{A5} \) and \( r_{A8} \) are the mean radii of samples A6, A5 and A8 respectively. The
size statistics and microscope images were provided by DU. Figure 6.5 shows the SEM
images of samples A5 and A8 as well as their corresponding size distribution histograms.
Note that the SEM image of A5 is for a magnification of 150000x while the SEM image
of A8 is for a magnification of 100000x. The size statistics and SEM image was not
available for sample A6.
Figure 6.5 SEM images of A5 and A8 with corresponding size distribution histogram

Figure 6.6 shows the AFM images (provided by DU) of the three samples A5, A6 and A8. Table 6.1 summarizes the sizes and LSPR peak photon energies (along with wavelength equivalents) of the three samples. The mean radius for sample A6 was unavailable. However, it is known (also clearly seen in the AFM images) that the mean radius of A6 is smaller than that of sample A5.
Figure 6.6 AFM images of samples A5, A6 and A8

Table 6.1 Size distribution summary and LSPR peak photon energy for A5, A6 and A8

<table>
<thead>
<tr>
<th></th>
<th>A5</th>
<th>A6</th>
<th>A8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean Radius (nm)</td>
<td>26</td>
<td>&lt; 26</td>
<td>16 &amp; 70</td>
</tr>
<tr>
<td>LSPR (eV)</td>
<td>3.82</td>
<td>4.26</td>
<td>1.98</td>
</tr>
<tr>
<td>LSPR (nm)</td>
<td>324.6</td>
<td>291.0</td>
<td>626.2</td>
</tr>
</tbody>
</table>

The ellipsometric measurements performed by DU reveal that the transverse mode (or lower energy mode) of the LSPR peak occurs in sample A5 at 324.6 nm, in sample A6 at 291.0 nm and in sample A8 at 626.2 nm. The size distribution for sample A8 is bimodal and the LSPR peak of 626.2 nm corresponds to the transverse mode in the 70 nm sized nanoparticles. The 16 nm sized nanoparticles is expected to have a LSPR peak in the deep UV, beyond the measuring capability of DU’s \textit{in situ} spectroscopic ellipsometer. Similarly, the longitudinal mode (or higher energy mode) of the LSPR peak is blue-shifted and beyond the measuring capability of the ellipsometer.

While the Ga NPs were grown on one surface of the Sapphire substrate, the other surface was coated by DU with Titanium (Ti) using e-beam evaporator. The thickness of the Ti film was estimated by DU to be around 2 $\mu$m.
6.4 Mueller Matrix Ellipsometric Measurements

The developed polarimetric system was configured in the reflection mode with a 70° incident angle paralleling the operation of a conventional ellipsometer. This section first presents measurement results on the Sapphire substrate. Thereafter, measurements are performed on the nanoparticle samples and the results, including pseudodielectric function, Mueller matrix and polarization properties, are analyzed.

6.4.1 Measurement on Sapphire Substrate

Mueller matrix measurements were performed on the bare Sapphire substrate at 70° reflection. The polarization properties are plotted in blue for the bare Sapphire substrate in Figure 6.7. Note that the substrate has a significant amount of diattenuation which is fairly constant, between 0.86 and 0.87.
A Crystal Violet (CV) dye coating was applied to the Ga nanoparticle samples by our collaborators for the purpose of Surface Enhanced Raman Scattering (SERS) studies. In order to understand the influence this coating has on polarization properties, a CV coated Sapphire substrate was measured. The thickness of this coating on the substrate was not known. The polarization properties of this sample are plotted in red in Figure 6.7. The prominent polarization features in linear retardance, orientation of linear retardance, linear diattenuation, linear polarizance and the small feature in the depolarization around 590 nm coincide with the absorption peak of CV. Figure 6.8 is a plot of the normalized absorption coefficient of CV [67].
Using the linear retardance and linear diattenuation for bare sapphire (plotted in blue) in Figure 6.5, the complex refractive index was calculated using the ellipsometric equation (Equation 6.1) and its relation to the pseudodielectric function (Equation 6.2). This experimentally determined refractive index was compared against the tabulated refractive index [68] for verification. The real parts of the refractive indices are plotted in Figure 6.9 with the experimental values plotted in blue and the tabulated values plotted in red. There is a good correspondence between the theoretical and experimental values except for certain anomalies between 300 nm and 400 nm, possibly due to low SNR, scattering or, more likely, the presence of the Ti coating on the opposite side of the wafer. The refractive index of Ti decreases significantly below 400 nm, approaching 1 around 300 nm [69]. The imaginary part of the refractive index was near zero throughout as expected (discussed later along with Figure 6.11).
6.4.2 Pseudodielectric Function of Ga Nanoparticle Samples

The polarimetric measurements performed on samples with Ga nanoparticles produces, as before, the Mueller matrix which is decomposed to extract the polarization properties of the sample. The linear retardance and linear diattenuation thus calculated is used in the pseudodielectric function (Equation 6.2) to calculate the complex refractive index. Figure 6.10 is a plot of linear retardance and linear diattenuation for samples A5, A6 and A8. The standard deviation is represented by the shaded area along the width of each curve. The shaded area is not readily visible since the standard deviation is about the width of the line. Figure 6.11 is a plot of the calculated real and imaginary parts of the refractive index for the bare sapphire and samples A5, A6 and A8. The shaded area represents the standard deviation. In this case the standard deviation is about the width of the line.
Figure 6.10 Plot of the retardance and diattenuation for samples A5, A6 and A8
Figure 6.11 Complex refractive indices of sapphire substrate, samples A5, A6 and A8

The imaginary part of the refractive index is plotted as broken lines while the real part of the refractive index is plotted as solid lines. The complex refractive index of sapphire is plotted in black and it can be seen that while the real part is between 1.7 and 1.8 in the entire spectral region, the imaginary part is consistently zero. This means that the sapphire substrate has negligible absorption, as expected. It can also be clearly seen that the peaks of the imaginary part of the refractive indices for the samples A5, A6 and A8 occur at the spectral region where their corresponding real parts undergo anomalous dispersion. This is also as expected and consistent with theory.
Figure 6.12 is a plot comparing the imaginary part of the refractive index for samples A5, A6 and A8, as determined by the *in situ* spectroscopic ellipsometer at DU (labeled DUKE) with that of the *ex situ* Mueller matrix ellipsometric measurements determined here (labeled UAH). Shaded area represents standard deviation. Again, the standard deviation is about the width of the line. The redshift observed in the measurements performed *ex situ* is likely due to a thin layer of oxide of the metal [8] or the CV dye coated over the Ga NP samples. The redshift for Ga NPs has previously been observed to be about 0.4 to 0.5 eV [6]. Since the absorption increases when the resonance condition is satisfied, the peak position in each absorption curve corresponds to the occurrence of LSPR peaks in the respective samples. Table 6.2 summarizes the peak positions from Figure 6.12 along with the expected LSPR peak position. The LSPR peaks for samples A5 and A6 are very close to the range of expected values considering that the measurements were performed at a 10 nm resolution. The LSPR peak position for sample A8 is not as redshifted as expected. A likely reason could be the difference in the thickness of CV coating on sample A8 as compared to sample A5 and A6. However, the CV thickness information was unavailable and this hypothesis has not been confirmed.

The other important observation is the negative values for absorption which is non-physical. This is likely due to islands of metal nanoparticles on a dielectric plate allowing multiple reflections to occur within the plate. The theoretical model (pseudodielectric function) used for the calculation assumes a uniform thin film on a semi-infinite medium.
Figure 6.12 Comparison of the pseudodielectric function for samples A5, A6, and A8

Table 6.2 Comparison of LSPR peaks of samples A5, A6, and A8

<table>
<thead>
<tr>
<th></th>
<th>A5</th>
<th>A6</th>
<th>A8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean Radius (nm)</td>
<td>26</td>
<td>&lt; 26</td>
<td>16 &amp; 70</td>
</tr>
<tr>
<td>DU LSPR (nm)</td>
<td>324.6</td>
<td>291.0</td>
<td>626.2</td>
</tr>
<tr>
<td>Expected LSPR (nm)</td>
<td>362.5 – 373.4</td>
<td>321.2 – 329.7</td>
<td>784.7 – 837.7</td>
</tr>
<tr>
<td>UAH LSPR (nm)</td>
<td>360</td>
<td>330</td>
<td>730</td>
</tr>
</tbody>
</table>

6.4.3 Polarization Properties of Ga Nanoparticle Samples

It is important to identify features, if any, in the plots of the Mueller matrices and polarization properties corresponding to the LSPR peaks in order to understand and correlate the occurrence of the resonance peaks with such properties.
Figure 6.13 shows the spectral plots (ranging from 250 nm to 1100 nm) of all the 16 elements of the Mueller matrix of samples A5, A6 and A8. Shaded areas represent the standard deviation. The standard deviation is about the width of the line.

Figure 6.13 Mueller matrix plots of samples A5, A6 and A8

Figure 6.14 is a plot of the polarization properties of the samples A5, A6 and A8. Once again, shaded areas represent the standard deviation. The most significant polarization properties that show up are linear retardance, linear diattenuation and depolarization.
Sample A8 can be seen as having rather broad peaks in the Mueller matrix elements and polarization properties compared to samples A5 and A6. This is likely due to the larger spread in the size distribution.

A polarization property that has not previously been quantified for nanoparticle systems such as the one under consideration is depolarization. The Mueller matrix ellipsometer has successfully measured the depolarization of Ga NP samples.

Figure 6.15 allows a closer view of the depolarization plot. Shaded area represents the standard deviation. The standard deviation is about the width of the line. The peaks in the depolarization curve for samples A5 and A6 occur at 590 nm and 520 nm, respectively. The more interesting feature is the minima in the depolarization curves for
samples A5 and A6. The minima in depolarization index for sample A5 occurs at 360 nm coinciding with the measured LSPR peak of 360 nm. The minima in depolarization index for sample A6 occurs at 320 nm close to the measured LSPR peak of 330 nm.

![Depolarization Indices of Samples A5, A6, and A8](image)

Figure 6.15 Depolarization indices of samples A5, A6, and A8

The exact reason for the depolarization peak and minima and its correlation, if any, to LSPR peaks is not known. Theoretical and experimental investigation by our collaborators, also involving us, is ongoing which may help understand this phenomenon in greater detail. Perhaps the depolarization is expected to increase with decreasing wavelength, but the onset of LSPR (or absorption) suppresses the multiple scattering and hence the depolarization.

Sample A8 does not seem to show any significant peaks or dips in the depolarization index. This may be due to the spectrally broad resonance occurring at 730 nm. There are likely depolarization peaks that are outside the spectral region spanned by the Mueller matrix ellipsometer.
Note that Figure 6.15 is the plot of depolarization ranging from 250 nm to 1100 nm. The structures seen below 300 nm and near 1100 nm cannot be deemed reliable due to low SNR associated with instrument cut-off.

6.5 Conclusions

The Mueller matrix ellipsometric measurement of Ga nanoparticles deposited on a Sapphire substrate reveals the LSPR peaks. Hence, the developed system is able to parallel the measurements of a conventional ellipsometer. The measured LSPR peaks are redshifted compared to the in situ measurements which is likely due to modification of the sample surfaces. The Mueller matrix ellipsometric measurements also reveal previously unknown polarization features, especially depolarization. The position of the depolarization peak is away from the LSPR peak and at a higher wavelength.

Thus, this novel system is able to measure all the Mueller matrix elements and extract the complete polarization properties of Ga NPs from 300 nm to 1100 nm for the first time. The additional polarization information provided by the MMSP-R provides direction for furthering experimental and theoretical work in the area of nanoplasmonics.
CHAPTER 7

CONCLUSIONS AND RECOMMENDATIONS

7.1 Dissertation Accomplishments

This dissertation has described the successful development of a Mueller matrix spectropolarimeter (MMSP) that operates in the UV-VIS-NIR spectral region. This MMSP can be configured in both a transmission mode (MMSP-T) and a reflection mode (MMSP-R). The MMSP has been developed from the ground up and it operates from 300 nm to 1100 nm at high resolution with good accuracy and precision. The MMSP is capable of measuring the complete Mueller matrix of a variety of samples and is extremely versatile. The Mueller matrix is decomposed to provide all the polarization properties, namely, linear diattenuation, linear retardance, linear polarizance, circular diattenuation, circular retardance, circular polarizance, depolarization index and the orientation axes of the linear diattenuation and linear retardance- all as a function of this broad wavelength region for each sample. This makes the developed MMSP a one of a kind measuring system.

The MMSP-R has been used to measure the polarization properties of various atmospheric particulate matter species (dust, soot, pollen and BG) deposited on microscope slides. It has provided for the first time differentiating polarization features
between the various atmospheric particulate matter species in the UV-VIS-NIR spectral region. This can be exploited in the near future for developing sensors for bio-aerosol detection.

The MMSP-R has also been used to measure the complete Mueller matrix of a metal nanoparticle system from which all the polarization properties of the sample were extracted. This revealed some previously unknown features in polarization properties, especially, depolarization. The Mueller matrix and polarization properties information are available in the broad spectral region spanning UV-VIS-NIR. The additional polarization information will provide new directions for further theoretical work. The data from the MMSP-R was used to calculate the pseudodielectric function which locates the LSPR peak of metal nanoparticle systems.

The major accomplishments of this dissertation are summarized below:

1. In reflection mode, measurements performed on atmospheric particulate matter clearly provide, for the first time, differentiating polarization features. This demonstrates the potential of utilizing the polarimetric technique for the discrimination of bio-aerosols in the midst of ambient atmospheric aerosol clutter.

2. Measurements performed on Gallium nanoparticle samples by the MMSP-R provide the complete Mueller matrix of the sample and all the polarization properties. It reveals previously unknown features in polarization properties, especially, depolarization. The additional polarization information provided by the MMSP-R provides new direction for furthering theoretical work in the area of
nanoplasmonics. This is expected to lead to better understanding of the interaction between electromagnetic radiation and nanoparticle systems.

3. The data from the MMSP-R can be used to calculate the pseudodielectric function of the Ga NP sample, thus paralleling the operation and measurement of a conventional ellipsometer. The peak in the pseudodielectric function (k) locates the LSPR peak of the metal nanoparticle system.

4. The MMSP-T is able to measure the polarization properties of a variety of samples, such as, optical components, electro-optical devices, optically active materials, polymer materials, etc. This demonstrates the versatility of the MMSP and the range of applications that is made possible.

5. In order to achieve these applications, the MMSP was developed from the ground up. The components of the system are chosen such that it operates seamlessly from 300 nm to 1100 nm at a high spectral resolution with good precision and accuracy. The system is completely automated and the Labview interface used for controlling the instrument and data acquisition has an easy to use and intuitive Graphical User Interface (GUI).

6. Calibration, data reduction and post-processing are achieved by means of easy to understand Matlab code. The calibration has been achieved by either the Fourier decomposition technique or the non-linear regression technique for estimating the systematic errors in the MMSP, based on the measurement mode. The data reduction is able to compensate for the estimated systematic errors.

7. The MMSP measures the complete Mueller matrix of the sample. The Mueller matrix is decomposed to provide all the polarization properties, namely, linear
diattenuation, linear retardance, linear polarizance, circular diattenuation, circular retardance, circular polarizance, depolarization index and the orientation axes of the linear diattenuation and linear retardance, as a function of the broad wavelength region at high spectral resolution. This makes the developed MMSP a one of a kind instrument.

7.2 Recommendations for Future Research

The research on micro-structured surfaces is by no means comprehensive or absolutely conclusive. Improvements can be made in several areas for optimum operation and enhanced performance. Some suggestions for future research are:

1. A better alternative to the quarter wave retarder, if any, should be found which has a larger clear aperture but a much shorter prism length while retaining the “achromaticity” of the Fresnel Rhomb over the broad spectral region. This will make the optical alignment far easier while greatly increasing the throughput of the system.

2. Rather than having a narrow band of wavelengths exit the monochromator and pass through the PSG and PSA, it may be better to have the broadband source pass through the PSG and PSA and have a spectrometer and a linear CCD at the detector end to perform the spectral measurements. This would significantly reduce the time taken for acquiring data. The original design and budget did not accommodate this feature in the current system.
3. The most beneficial upgrade to this system would be a rotation stage with an arm on which the PSA and spectrometer are mounted. This would enable measurements to be performed at any scattering angle.

4. Improve the calibration of the system in reflection such that reliable calibration parameters can be estimated leading to better system performance.

5. With respect to bio-aerosol detection, improvements are required for consistent sample preparation. It should be possible to quantitatively estimate the areal density of the particulate matter deposit on the slide. This is important for correlating the observed polarization features to the density of the scatterer.

6. The research must depart from considering isolated species of particulate matter and should consider binary, ternary and higher order mixtures of varying proportions in order to conclusively determine the feasibility of such a bio-aerosol sensor.

7. Application in the area of Mueller matrix ellipsometry of metal nanoparticles can be furthered by considering several samples of varying size distributions and different metals (Au, Ag, etc). The results provided by this system can then be compared to results from conventional ellipsometers. The capability of performing measurements at various incident angles is expected to provide significant new information.

8. The MMSP-R can also be used to measure the complete Mueller matrix and polarization properties of dielectric films, semiconductor films as well as metal films.
7.3 Conclusions

The Mueller matrix spectropolarimeter (MMSP-T and MMSP-R) developed in this research provides the ideal platform for studying UV-VIS-NIR spectral polarization properties of a variety of samples. In transmission mode, the system has been valuable in measuring the polarization properties of optical components and electro-optic devices, and characterization of materials like polymers, etc. The instrument demonstrates enormous potential for bio-aerosol detection by successfully measuring and differentiating between several species of atmospheric particulate matter. The instrument also has provided interesting insights into the polarization properties of metal nanoparticles, at and around Localized Surface Plasmon Resonance (LSPR) peaks.
APPENDIX A

Light Scattering – Qualitative description

This appendix provides a qualitative description of the light scattering process [11,12,70]. Electromagnetic radiation incident on a scattering medium sets the electric charges into oscillatory motion. This acceleration of charges leads to the radiation of electromagnetic energy in all directions. This secondary radiation is referred to as ‘scattered’ radiation. Apart from reradiating the electromagnetic energy, the oscillating charges may transform part of the incident electromagnetic energy into other forms (such as thermal energy), in a process termed absorption. The scattering process may be elastic or inelastic. The scattering is elastic when energy (and hence the wavelength) of the incident photon is conserved and only the direction is changed, for instance, Rayleigh scattering and Mie scattering. The scattering is inelastic when the scattered photon has a wavelength which is blue-shifted or red-shifted with respect to the incident photon, for instance, Brillouin scattering or Raman scattering. In this context, elastic scattering (and more specifically, Mie scattering) is of interest.

In order to understand electromagnetic scattering by a single particle, the particle is conceptually subdivided into a collection of tiny dipoles driven to radiate (or scatter) by the incident electromagnetic field. Each dipole reradiates coherently in all directions and the total scattered field in a particular direction is given by the superposition of the individual scattered field, taking into account their phase differences. Thus, the scattering by a particle is dependent on its size and shape, the scattering angle, the composition of the particle (optical constants), and the polarization state and frequency of the incident electromagnetic field [71].
The simplest particle is an isotropic, homogeneous sphere. The scattering by a sphere can be determined using various approximations like Mie theory, Fraunhofer theory, geometrical optics, anomalous diffraction, coupled-dipole method, T-matrix method, etc. Calculation of scattering of non-spherical particles and aggregates is much more complex [10,72].
## APPENDIX B

1. Commonly encountered Jones vectors and Stokes vectors

<table>
<thead>
<tr>
<th>State of polarization</th>
<th>Jones vector</th>
<th>Stokes vector</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linear horizontal polarization</td>
<td>$\begin{bmatrix} 1 \ 0 \end{bmatrix}$</td>
<td>$\begin{bmatrix} 1 \ 1 \ 0 \ 0 \end{bmatrix}$</td>
</tr>
<tr>
<td>Linear vertical polarization</td>
<td>$\begin{bmatrix} 0 \ 1 \end{bmatrix}$</td>
<td>$\begin{bmatrix} 1 \ -1 \ 0 \ 0 \end{bmatrix}$</td>
</tr>
<tr>
<td>Linear 45° polarization</td>
<td>$\frac{1}{\sqrt{2}} \begin{bmatrix} 1 \ 1 \end{bmatrix}$</td>
<td>$\begin{bmatrix} 1 \ 0 \ 1 \ 0 \end{bmatrix}$</td>
</tr>
<tr>
<td>Linear 135° polarization</td>
<td>$\frac{1}{\sqrt{2}} \begin{bmatrix} 1 \ -1 \end{bmatrix}$</td>
<td>$\begin{bmatrix} 1 \ 0 \ -1 \ 0 \end{bmatrix}$</td>
</tr>
<tr>
<td>Right circular polarization</td>
<td>$\frac{1}{\sqrt{2}} \begin{bmatrix} 1 \ i \end{bmatrix}$</td>
<td>$\begin{bmatrix} 1 \ 0 \ 0 \ 1 \end{bmatrix}$</td>
</tr>
<tr>
<td>Left circular polarization</td>
<td>$\frac{1}{\sqrt{2}} \begin{bmatrix} 1 \ -i \end{bmatrix}$</td>
<td>$\begin{bmatrix} 1 \ 0 \ 0 \ -1 \end{bmatrix}$</td>
</tr>
<tr>
<td>Unpolarized light</td>
<td>-----</td>
<td>$\begin{bmatrix} 1 \ 0 \ 0 \ 0 \end{bmatrix}$</td>
</tr>
<tr>
<td>Partial polarization (DoLP=0.5)</td>
<td>-----</td>
<td>$\begin{bmatrix} 1 \ 0.5 \ 0 \ 0 \end{bmatrix}$</td>
</tr>
</tbody>
</table>
2. Commonly encountered Jones matrices and Mueller matrices

<table>
<thead>
<tr>
<th>Optical component</th>
<th>Jones matrix</th>
<th>Mueller matrix</th>
</tr>
</thead>
</table>
| Free space                                           | \[
\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}
\] | \[
\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}
\] |
| Isotropic absorbing material with transmittance \(k\) | \[
\begin{bmatrix} \sqrt{k} & 0 \\ 0 & \sqrt{k} \end{bmatrix}
\] | \[
\begin{bmatrix} k & 0 & 0 & 0 \\ 0 & k & 0 & 0 \\ 0 & 0 & k & 0 \\ 0 & 0 & 0 & k \end{bmatrix}
\] |
| Linear horizontal polarizer                          | \[
\begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix}
\] | \[
\begin{bmatrix} 1 & -1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}
\] |
| Linear vertical polarizer                            | \[
\begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix}
\] | \[
\begin{bmatrix} 1 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 1 \end{bmatrix}
\] |
| Linear polarizer at 45°                              | \[
\frac{1}{2}\begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix}
\] | \[
\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}
\] |
| Right circular polarizer                             | \[
\frac{1}{2}\begin{bmatrix} 1 & i \\ -i & 0 \end{bmatrix}
\] | \[
\begin{bmatrix} 1 & 0 & 0 & -i \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & 1 \end{bmatrix}
\] |
| Left circular polarizer                              | \[
\frac{1}{2}\begin{bmatrix} 1 & -i \\ i & 1 \end{bmatrix}
\] | \[
\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & -1 & 0 \end{bmatrix}
\] |
| Linear quarter wave retarder with fast axis at 0°     | \[
\begin{bmatrix} 1 & 0 \\ 0 & i \end{bmatrix}
\] | \[
\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{bmatrix}
\] |
| Linear half wave retarder with fast axis at 45°       | \[
\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}
\] | \[
\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}
\] |
| Depolarizer                                          | -----        |                |
APPENDIX C

Manufacturer Specification

1. 150 W UV Enhanced Xenon Arc Lamp (Newport 6254)

2. Oriel Cornerstone 260 ¼ m Monochromator (Newport 74125)

<table>
<thead>
<tr>
<th>Specification</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Focal Length</td>
<td>260 mm</td>
</tr>
<tr>
<td>F/number</td>
<td>F/3.9</td>
</tr>
<tr>
<td>Usable Wavelength Range</td>
<td>180 nm to 24 μm, with interchangeable gratings</td>
</tr>
<tr>
<td>Wavelength Accuracy</td>
<td>0.35 nm</td>
</tr>
<tr>
<td>Resolution (triple grating instrument)</td>
<td>0.15 nm</td>
</tr>
<tr>
<td>Resolution (double grating instrument)</td>
<td>0.10 nm**</td>
</tr>
<tr>
<td>Wavelength Precision</td>
<td>0.08 nm</td>
</tr>
<tr>
<td>Maximum Slew Rate</td>
<td>205 nm/s***</td>
</tr>
<tr>
<td>Weight</td>
<td>(9.5 kg)</td>
</tr>
</tbody>
</table>

*With 1200 l/mm grating, 10 μm x 2 mm slit.
**Optical resolution step size limit is 0.08 nm.
***With 1200 l/mm grating.
3. Rochon Prism Polarizer (United Crystals) [73]

- Common Materials: Calcite, a-BBO, YVO4, Quartz, MgF2
- Structure: Optical Cement, or Optical Contact
- General Aperture: 10×10~20×20mm
- Extinction Ratio: 1×10⁻⁵
- Transmitting wavefront distortion: less than λ/8 @633nm
- Transmittance: >90% @633nm
- Transparent Range: 200-5000nm
- Beam Splitting Angle: 2.5°, 5°, 7.5° @633nm
- Damage Threshold: 10W/cm²(CW); 100MW/cm²(Pulse) Calcite
- Coating: AR-coating available upon request

4. Fresnel Rhomb λ/4 Retarder (Artifex Engineering 50.260.00113)

<table>
<thead>
<tr>
<th>Material:</th>
<th>UV Grade Fused Silica</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dimension Tolerance:</td>
<td>+0.0/-0.2mm</td>
</tr>
<tr>
<td>Clear Aperture:</td>
<td>&gt;80%</td>
</tr>
<tr>
<td>Flatness:</td>
<td>λ/10@ 632.8 nm</td>
</tr>
<tr>
<td>Surface Quality:</td>
<td>20-10 scratch and dig</td>
</tr>
<tr>
<td>Wavelength Range:</td>
<td>200-2000 nm for Fused Silica</td>
</tr>
<tr>
<td>Bevel:</td>
<td>0.2mm to 0.5mm</td>
</tr>
</tbody>
</table>
5. Precision Rotation Stage (Newport URS150)

<table>
<thead>
<tr>
<th>Model</th>
<th>URS150BCC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Travel Range</td>
<td>360 °</td>
</tr>
<tr>
<td>Resolution</td>
<td>0.0005 °</td>
</tr>
<tr>
<td>Minimum Incremental Motion</td>
<td>0.002 °</td>
</tr>
<tr>
<td>Uni-directional Repeatability, Guaranteed</td>
<td>0.002 °</td>
</tr>
<tr>
<td>Absolute Accuracy, Guaranteed</td>
<td>0.023 ° or ± 0.0115 °</td>
</tr>
<tr>
<td>Maximum Speed</td>
<td>80 °/s</td>
</tr>
<tr>
<td>Wobble, Guaranteed</td>
<td>50 or ± 25 μrad</td>
</tr>
<tr>
<td>Eccentricity, Guaranteed</td>
<td>3 or ± 1.5 μm</td>
</tr>
<tr>
<td>MTBF</td>
<td>20000 h @ 25% load and 30% duty cycle h</td>
</tr>
<tr>
<td>Thread Type</td>
<td>M6</td>
</tr>
<tr>
<td>Base Material</td>
<td>Hardened steel with aluminum body</td>
</tr>
<tr>
<td>Motor</td>
<td>UE34CC</td>
</tr>
<tr>
<td>Feedback</td>
<td>Worm mounted rotary encoder, 8,000 cts/rev, Index pulse</td>
</tr>
<tr>
<td>Limit Switches</td>
<td>Two independently adjustable optical limit switches</td>
</tr>
<tr>
<td>Origin</td>
<td>Optical, fixed at position 0°.</td>
</tr>
<tr>
<td>Worm Gear Ratio</td>
<td>1:90 + 1:2.75 drive belt</td>
</tr>
<tr>
<td>Bearings</td>
<td>Large diameter ball bearings</td>
</tr>
<tr>
<td>Drive Mechanism</td>
<td>Worm/gear</td>
</tr>
<tr>
<td>Drive Type</td>
<td>DC Servo</td>
</tr>
<tr>
<td>Cable Length</td>
<td>3 m</td>
</tr>
<tr>
<td>Weight</td>
<td>3.4 kg (7.5 lb)</td>
</tr>
<tr>
<td>Maximum Torque (Mz)</td>
<td>2 Nm</td>
</tr>
<tr>
<td>Transversal Stiffness (ko)</td>
<td>5 μrad/Nm</td>
</tr>
<tr>
<td>Normal Load Capacity (Cz)</td>
<td>300 N</td>
</tr>
</tbody>
</table>
6. UV Enhanced Mounted Silicon Photodiode (Thor Labs SM1PD2A)

**Electrical Characteristics:**

- **Spectral Response:** 200 - 1100 nm
- **Active Area:** \( \varnothing 9.0 \text{mm} \ (63.6 \text{mm}^2) \)
- **Rise Time \((R_L = 50 \Omega)\):** 45 ns (5V bias)
- **Fall Time \((R_L = 50 \Omega)\):** 45 ns (5V bias)
- **NEP @900\text{nm}:** \(5.5 \times 10^{-14} \text{ W/}\sqrt{\text{Hz}}\) (@5V bias)
- **Dark Current:** 1\(\mu\text{A} \) max. (5V)

**Maximum Ratings:**

- **Damage Threshold CW:** 10W/cm\(^2\)
- **Damage Threshold Pulse:** 5\(\mu\text{J/cm}^2\)
- **Max Bias Voltage:** 10V

**Spectral Response:**

![Spectral Response Graph]

7. Multifunction DAQ (National Instruments PCI-6014)

- Ideal for beginning users and OEMs
- 16 analog inputs at up to 200 kS/s
- 16-bit input resolution
- 8 or 10 digital I/O lines
- Digital triggering
- Up to 4 analog input signal ranges
8. Lock-In Amplifier (Stanford Research Systems SR510)

<table>
<thead>
<tr>
<th>Signal Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Inputs</strong></td>
</tr>
<tr>
<td>Voltage</td>
</tr>
<tr>
<td>Current</td>
</tr>
<tr>
<td><strong>Impedance</strong></td>
</tr>
<tr>
<td>Voltage</td>
</tr>
<tr>
<td>Current</td>
</tr>
<tr>
<td><strong>Full-scale sensitivity</strong></td>
</tr>
<tr>
<td>Voltage</td>
</tr>
<tr>
<td>Current</td>
</tr>
<tr>
<td><strong>Maximum inputs</strong></td>
</tr>
<tr>
<td>Voltage</td>
</tr>
<tr>
<td>Current</td>
</tr>
<tr>
<td><strong>Noise</strong></td>
</tr>
<tr>
<td>Voltage</td>
</tr>
<tr>
<td>Current</td>
</tr>
<tr>
<td><strong>Common Mode Rejection</strong></td>
</tr>
<tr>
<td>Range</td>
</tr>
<tr>
<td>Rejection</td>
</tr>
<tr>
<td><strong>Gain stability</strong></td>
</tr>
<tr>
<td>Accuracy</td>
</tr>
<tr>
<td><strong>Signal filters</strong></td>
</tr>
<tr>
<td>60 Hz notch, 50 dB (Q=10), adjustable from 45 Hz to 65 Hz</td>
</tr>
<tr>
<td>120 Hz notch, 50 dB (Q=10), adjustable from 100 Hz to 130 Hz</td>
</tr>
<tr>
<td>Tracking band pass (Q=5). Filter adds 20 dB to dynamic reserve</td>
</tr>
<tr>
<td><strong>Dynamic reserve</strong></td>
</tr>
<tr>
<td>LOW (20 dB), 5 ppm/°C</td>
</tr>
<tr>
<td>NORM (40 dB), 50 ppm/°C</td>
</tr>
<tr>
<td>HIGH (60 dB), 500 ppm/°C</td>
</tr>
<tr>
<td>(100 mV to 5 mV sensitivity)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Reference Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Frequency</strong></td>
</tr>
<tr>
<td>0.5 Hz to 100 kHz</td>
</tr>
<tr>
<td><strong>Input impedance</strong></td>
</tr>
<tr>
<td>1 MΩ, AC coupled</td>
</tr>
<tr>
<td><strong>Trigger</strong></td>
</tr>
<tr>
<td>Signal</td>
</tr>
<tr>
<td>Pulse</td>
</tr>
<tr>
<td>Mode</td>
</tr>
<tr>
<td>Acquisition time</td>
</tr>
<tr>
<td>SR slew rate</td>
</tr>
<tr>
<td>Phase control</td>
</tr>
<tr>
<td>Phase noise</td>
</tr>
<tr>
<td>Phase drift</td>
</tr>
<tr>
<td>Phase error</td>
</tr>
<tr>
<td><strong>Orthogonality</strong></td>
</tr>
<tr>
<td>90° ± 1°</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Demodulator</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Stability</strong></td>
</tr>
<tr>
<td>5 ppm/°C (LOW reserve)</td>
</tr>
<tr>
<td>50 ppm/°C (NORM reserve)</td>
</tr>
<tr>
<td>500 ppm/°C (HIGH reserve)</td>
</tr>
<tr>
<td><strong>Time constants</strong></td>
</tr>
<tr>
<td>Pre</td>
</tr>
<tr>
<td>Post</td>
</tr>
<tr>
<td><strong>Offset</strong></td>
</tr>
<tr>
<td>Up to 1° full scale (10° on expanding)</td>
</tr>
<tr>
<td><strong>Harmonic rejection</strong></td>
</tr>
<tr>
<td>−55 dB (band pass filter in)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Outputs and Interfaces</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Channel 1 outputs</strong></td>
</tr>
<tr>
<td>X (Rcos), X Offset, X Noise, R, R Offset, X5 (ext. D/A)</td>
</tr>
<tr>
<td><strong>Channel 2 outputs</strong></td>
</tr>
<tr>
<td>X (Rsin), Y (Rsin), X offset, Y offset, Y (ext. D/A)</td>
</tr>
<tr>
<td><strong>Output meters</strong></td>
</tr>
<tr>
<td>2% precision analog meter</td>
</tr>
<tr>
<td><strong>Output LCD</strong></td>
</tr>
<tr>
<td>4-digit LCD display shows same value as the analog meter</td>
</tr>
<tr>
<td><strong>Output BNC</strong></td>
</tr>
<tr>
<td>±10 V corresponds to full-scale input (&lt;1 Ω output impedance)</td>
</tr>
<tr>
<td><strong>Reference output</strong></td>
</tr>
<tr>
<td>4-digit LCD display for reference phase or frequency</td>
</tr>
<tr>
<td>X1 to X4</td>
</tr>
<tr>
<td>4 analog inputs, 13-bit, ±10.24 V</td>
</tr>
<tr>
<td>X5, X6</td>
</tr>
<tr>
<td>2 analog outputs, 13-bit, ±10.24 V</td>
</tr>
<tr>
<td>X output*</td>
</tr>
<tr>
<td>X (Rcos), ±10 V, ±1 Ω output impedance</td>
</tr>
<tr>
<td>Y output*</td>
</tr>
<tr>
<td>Y (Rsin), ±10 V, ±1 Ω output impedance</td>
</tr>
<tr>
<td><strong>Ratio</strong></td>
</tr>
<tr>
<td>Ratio output equals ±1° signal output divided by the denominator of the input</td>
</tr>
<tr>
<td><strong>Internal oscillator</strong></td>
</tr>
<tr>
<td>Range</td>
</tr>
<tr>
<td>Accuracy</td>
</tr>
<tr>
<td>Stability</td>
</tr>
<tr>
<td>500 ppm/°C (amplitude)</td>
</tr>
<tr>
<td><strong>Distortion</strong></td>
</tr>
<tr>
<td>2% THD</td>
</tr>
<tr>
<td><strong>Amplitude</strong></td>
</tr>
<tr>
<td>10 mVrms, 100 mVrms, 1 Vrms</td>
</tr>
<tr>
<td><strong>Computer interfaces</strong></td>
</tr>
<tr>
<td>RS-232 standard, GPIB optional</td>
</tr>
<tr>
<td>All instrument functions can be controlled and read through the interfaces</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>General</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Power</strong></td>
</tr>
<tr>
<td>35 W, 100/120/220/240 VAC, 50/60 Hz</td>
</tr>
<tr>
<td><strong>Dimensions</strong></td>
</tr>
<tr>
<td>(SR510) 17° × 3.5° × 17° (WHD)</td>
</tr>
<tr>
<td>(SR530) 17° × 5.25° × 17° (WHD)</td>
</tr>
<tr>
<td><strong>Weight</strong></td>
</tr>
<tr>
<td>12 lbs. (SR510), 16 lbs. (SR530)</td>
</tr>
<tr>
<td><strong>Warranty</strong></td>
</tr>
<tr>
<td>One year parts and labor on defects in materials and workmanship</td>
</tr>
</tbody>
</table>

* SR530 only
REFERENCES


[51] “Precision Rotation Stage, DC Motor, Continuous 360° Motion, URS150,” <http://search.newport.com/?q=*x2=sku&q2=URS150BCC> (15 April 2012).


