

Instrument Characterisation for Optical Activity Measurements in the Non-Paraxial Regime

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Except where acknowledged in the customary manner, the material presented in this thesis is, to the best of my knowledge, original and has not been submitted in whole or part for a degree in any university.

Connor Stewart

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Abstract

Optical activity provides essential information about the chirality of molecular structures in organic chemistry and has many applications in spectroscopy, crystallography and molecular biology. Collimated light has traditionally been used and its effect upon passing through these media is well understood. The plane of a linearly polarised beam rotates a constant amount independently of the incident polarisation [1]. The angle of rotation depends entirely on the beam path, concentration and structure of chiral molecules. The behaviour of linearly polarised light in the non-paraxial regime could be different [2] due to the possibility of it containing other components of polarisations. However, the helicity of the field remains preserved [1, 3] in a highly focused beam and recent studies link the helicity to the amount of optical rotation, suggesting that focused beams give rise to the same optical rotation as collimated ones.

The aim of this thesis is to develop an experimental setup that accurately measures the optical activity of a focused beam through a chiral sample. It is already known that light has to be collected in the forward direction [4]. Thus, our experimental setup has been characterised in such a configuration and the beam has been completely characterised over varying wavelengths and optical vortices.

The optical activity of a collimated beam with a fixed wavelength was measured through increasing concentrations of maltose and sucrose through varying beam path lengths. The same measurements were done for the non-paraxial case where the beam is focused. Cuvettes with various thicknesses containing the samples were translated through the focus of the beam and the Stokes parameters of the beam were calculated as a function of distance. The azimuthal angle was found to be independent of such translations and showed the same optical rotation as the collimated case.

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1

Introduction

1.1 Background and Scope

Optical activity has been a topic of great interest since its discovery in the early 19th century. In 1811, French physicist, François Jean Dominique Arago observed the rotation of the linear plane of polarised light as it passed through a quartz crystal [6]. Its discovery led to many advances in physics and chemistry. For example, in 1845, Michael Faraday demonstrated the link between light and electromagnetism with the discovery of magnetic optical activity. Many avenues in biochemistry and medicine were opened when Louis Pasteur first discovered enantiomeric pairs by observing the optical activity of tartaric acid in 1849 [7].

Optical activity is a property of a system describing its interaction with the circular polarisations of light. Optical activity has many manifestations, among them are *optical rotation* and *circular dichroism*. The first manifestation, optical rotation, is attributed to *circular birefringence*. A medium that has this property displays different indices of refraction for left and right circularly polarised light. Linearly polarised light can be decomposed into

left and right circularly polarised light and this refractive index differential will introduce a phase difference between the circular polarisations. The resulting recomposition is a different linear polarisation to the initial, the polarised light effectively “rotated” to a different linear polarisation. The second manifestation, circular dichroism, refers to a system’s differential absorption of left and right circular polarisations. The emergent beam would have an elliptical polarisation due to this differential.

It is clear to see that these definitions imply the importance of optical activity in the structure of chemicals. All systems that naturally exhibit optical activity have an important structural property called *chirality*. A structure or set of points is said to be chiral if it cannot be superimposed onto its mirror image. The development of the laser in the 1960s allowed for a stable source of coherent collimated light. This along with optical and electronic advances allowed for far more sensitive optical activity measurements. These measurements provide essential information about molecular structures. As a result, chirality has shown up in many areas such as pharmaceuticals, crystallography and biochemistry since a majority of chiral molecules are naturally occurring. Recently, it has also been used in the field of quantum optics [8].

1.2 Motivation

In 2013, the symmetry conditions of optical activity were theoretically derived [1]. The first condition, as discussed above, is that the medium the light propagates through must be chiral [7]. The second condition is that the medium must exhibit electromagnetic duality or the preservation of the *helicity* of light. If we described the electromagnetic field as a superposition of plane waves, the two eigenstates of helicity are analogous to the two circular polarisations of light. In the case of a solution of chiral molecules, every molecule the light interacts with must preserve helicity on its own. This means each molecule has to be electromagnetically dual [9, 10]. In general, chiral molecules are not dual. However, chiral molecules in a solution have random orientations. Thus, the system has spherical symmetry. In the definition of helicity, Λ , which is the projection of the total angular momentum, $\mathbf{J}$, onto the normalized linear momentum, $\mathbf{P}$,

$$\Lambda = \mathbf{J} \cdot \frac{\mathbf{P}}{|\mathbf{P}|} \quad (1.1)$$

whose eigenvalues are ± 1 [9], the total angular momentum is preserved after the interaction because the system has rotational symmetry. Therefore, helicity does not change in the forward scattering direction regardless of whether each individual molecule is electromagnetically dual or not [11]. This was later experimentally confirmed with optical activity measurements of scattering in the forward and perpendicular directions [4]. Measurements from different concentrations of the sugars: sucrose and maltose show that helicity is not preserved in the perpendicular direction and furthermore this was independent of the wavelength of light used. The change in circular polarisation demonstrated this. The circular polarisation remained the same in the forward direction further cementing the two conditions for optical activity.

In Gaussian optics, the *paraxial approximation* is a small angle approximation where the angle between the beam and the optical axis of the system is small. Almost all of the optical activity measurements since its first observation by Arago occurred in the paraxial regime. On the other hand, there have only been a few optical activity measurements in the non-paraxial regime. For example, in 2011, Löffler, Broer and Woerdman proved that orbital angular momentum has no effect on the optical activity of the beam [2]. In their paper, they demonstrated this with a cholesteric polymer arranged in a chiral crystalline form using a highly focused non-paraxial regime over a wavelength range of 500-700nm. However, it is still an open question if optical activity can be measured in the forward direction in the non-paraxial regime. This should be possible according to equation 1.1 because the beam preserved its helicity. However, the decomposition of the polarization in a non-paraxial beam shadows this assertion as each polarization could interact differently with the chiral medium. A focused beam has a different distribution of $\mathbf{k}$ vectors with respect to the same incident collimated beam before a high numerical aperture microscope objective. As a consequence, not only the distribution of $\mathbf{k}$ vectors changes but the polarisation relation changes as well. A simple model to account for those changes is the aplanic lens [12]. Answering this question is of main interest for any optical activity measurement in nanophotonics. This thesis aims to study the detection instrument requirements to measure the optical activity of a non-paraxial focused beam.

In this thesis, an experimental setup has been developed that analyses how to measure the optical activity of both a collimated and a non-paraxial focused beam for comparison. Sucrose and maltose are common sugars whose interactions induce well known optical activities that

have a predictable linear dependence on both the concentration and beam path if the beam is collimated [13, 14]. These sugars have been investigated in varying concentrations. The thickness of the cuvette containing the samples is also varied to explore different beam path lengths. The apparatus is setup in a way such that the beam's polarisation can be completely characterised. The eigenstates of the three normalised stokes parameters, s_1 , s_2 and s_3 , represent horizontal and vertical, diagonal and antidiagonal, left and right circular polarisations respectively. The superposition of a pair of these polarisations can produce any polarisation. However, it is not possible to measure the electromagnetic fields and phase directly to characterize the beam's polarisation. Instead, the polarisation characterisation is done through the intensity using a quarter and half waveplates converting the beam's polarisation to each of these three bases. Then, the stokes parameters are calculated by measuring the difference in the two perpendicular intensities for each of the previously mentioned bases after passing through a polarimeter or a polarised beam splitter. The beam can be set to any polarisation in a wavelength range from 760 to 810nm.

1.3 Layout of Thesis

Chapter 2 contains a history and review of the literature surrounding optical activity. I outline the relevant material concerning the paraxial approximation, angular momentum of light and light-matter interactions.

Chapter 3 explores the optical apparatus I have developed to measure optical activity in the paraxial regime. The measurements are taken using a polarimeter and serve as a control measurement for comparison in chapter 4.

Chapter 4 explores the optical apparatus I have developed to measure optical activity in the non-paraxial regime. This chapter explores three different methods of detection the first of which utilised the polarimeter. The second uses small area detectors to measure the beam after being split by a polarised beam splitter. The last uses two large area photodetectors to measure the split beam instead.

Chapter 5 concludes the thesis and outlines possible future research.

2

Background of Optical Activity

2.1 History of Optical Activity

In 1811, French physicist, François Jean Dominique Arago, first observed the rotation of the plane of linearly polarised light when passing through quartz [6]. Quartz is now known to form different trigonal crystals, chiral structures. Sir John F.W. Herschel followed up in this research in 1820 showing that two quartz crystals that were mirror images of each other rotated the plane of polarisation equally but in opposite directions [15]. In 1815, French physicist, Jean Baptiste Biot, also observed optical rotation through homogeneous fluids [16] and later in 1819 he observed similar phenomena in organic vapours such as turpentine [17]. It was assumed this was a property of organic substances. In the decades to come, Biot further investigated the optical activity of many more organic compounds, one of which was tartaric acid.

In 1816, Augustin-Jean Fresnel discovered the fact that orthogonally polarised beams do not interact [18]. Next year, he designed the Fresnel Rhomb, a prism that acted as a precursor

to the modern quarter waveplate converting linearly polarised light into circularly polarised light. This allowed him to identify that this manifestation of optical activity was a form of circular birefringence. For the phase difference case between the two circular polarisations due to differing refractive indices, the angle of optical rotation, ϕ in radians, for a path length L is given by:

$$\phi = \frac{2(n_L - n_R)L\pi}{\lambda} \quad (2.1)$$

where n_L and n_R are the refractive indexes for left and right circular polarisations respectively and λ is the wavelength of light.

In 1849, Louis Pasteur investigated a problem concerning tartaric acid [7, 19]. When derived through organic means it displayed the property of chirality and rotated the plane of polarisation. However, when synthetically created, it did not display this property. Like quartz, it could form into two different crystalline structures which were asymmetric and mirror images of each other. Each structure, when in solution rotated polarised light in opposite directions. The right handed orientations rotated light clockwise (dextrorotatory) and the left handed orientations rotated light anti-clockwise (levorotatory). When the two crystal structures are dissolved together in equal amounts, there is no net optical rotation. When created as a byproduct of the winemaking process, only one of the two structures emerged which has a net optical rotation. However, when created synthetically, there is no bias for each of the two mirrored structures and they were created in equal amounts producing no net optical rotation.

In 1852, George Gabriel Stokes defined parameters, now known as the Stokes Parameters, which characterise the polarisation of light [20]. Three of stokes parameters each represent the three bases consisting of two orthogonal polarisations: horizontal-vertical, diagonal-antidiagonal and left-right circular and they are labeled S_1 , S_2 and S_3 respectively. The fourth, S_0 , describes the intensity of the beam.

These observations led to better understanding of the structure of organic molecules. In 1874, there was a proposition that carbon based molecules displayed optical rotation due to the tetrahedral structure they form. If all four atoms chemically bonded to the carbon atom were different, the mirror image of that structure could not be superimposed onto itself. This was proposed independently by Jacobus Henricus van 't Hoff [21] and Joseph Achille Le Bel [22].

The term 'chirality' was not coined until 1893 by Lord Kelvin in the second lecture of

the Robert Boyle Lectures at the Oxford University Junior Scientific Club [23]. The formal definition of chirality referred to any geometrical structure or group of points whose mirror image could not be rotated or translated onto itself. The most prominent example of chiral structures are human hands for which such structures are named. Every chiral molecule has two stereoisomers called enantiomers that are mirror images of each other and are defined to have left and right handedness. From this point, chirality was closely linked with organic chemistry and was due to the way carbon chemically bonds with other elements. However, in 1914, Swiss chemist Alfred Werner produced the first synthetic chiral compound called Hexol [24], $[\text{Co}(\text{Co}(\text{NH}_3)_4(\text{OH})_2)_3]\text{Br}_6$, which has no carbon. This was one of the many results of his advancements in coordination chemistry.

Every optically active substance has a property called *specific rotation*, defined as the observed optical rotation per unit distance and unit concentration. Specific rotation is usually defined at room temperature (20°C), wavelength 589nm (also known as the sodium D line) and with collimated light. The equation for optical activity of a chiral solution is as follows [25]:

$$[\alpha]_{\lambda}^T = \frac{\alpha}{l \times c} \quad (2.2)$$

where α is the observed optical activity in degrees, l is the path length of the chiral solution in decimetres, c is the concentration of the chiral solution in grams per millilitre or grams per cubic centimetre, T is the temperature in degrees Celsius, λ is the wavelength of the beam in nanometres and $[\alpha]$ is the specific rotation of the chiral molecule. This measurement is most commonly carried out in a polarimeter. In this thesis, I have measured the optical activity of solutions of sucrose and maltose. The specific rotation of sucrose at 20°C and wavelength 589nm is $66.37 \text{ deg dm}^{-1} \text{ cm}^3 \text{ g}^{-1}$ [13]. Maltose is almost double in the same conditions with a value of $130.4 \text{ deg dm}^{-1} \text{ cm}^3 \text{ g}^{-1}$ [14].

As said in chapter 1, the conditions for optical activity have been theoretically derived [1]. The first condition is that the medium the light propagates through must have a chiral structure. The second condition is that the medium must preserve the helicity of the beam.

2.2 The Paraxial Approximation

As described in chapter 1, almost all of the optical activity observations since its first observation by Arago occurred in the paraxial regime. The goal of this thesis is to investigate

the instrumentation required to measure optical activity of a chiral sample in the non-paraxial regime. Therefore, the sample's optical activity is measured in a highly focused beam and is compared to its optical activity in a collimated beam. In other words, optical activity is measured and compared in the paraxial and non-paraxial regime.

In 1975, Melvin Lax, William H. Louisell and William B. Knight derived customary paraxial approximation of Maxwell's equations which have been mathematically used to characterise beams passing through commonly used optics [26]. When considering light whose electric field vector depends on its propagation, inconsistencies arise when the paraxial approximation is applied. Gaussian solutions are found in the direction of propagation despite Maxwell's equations saying otherwise. It was shown that the fields in Maxwell's equations can be expanded as a power series of the ratio between the beam width and the diffraction length if $\frac{w_0}{l} \ll 1$. With each higher order of the expansion, the solutions converge to an electric field along the direction of propagation. This provided an accurate description of paraxial wave optics.

The paraxial approximation has also been applied to high powered lasers. In 2015, laser pulses with very high intensity have been described in the paraxial approximation [27]. It was shown that it is applicable to femtosecond laser pulses. With first-order and second-order corrections it is applicable to longer pulses. Most of the time, laser radiation is a Gaussian beam but higher order modes spatially change this structure. These higher order corrections still describe the non-Gaussian beams if the pulse width is sufficiently small.

Following on from the research by Lax, Louisell and Knight, in 1985 Takenaka applied the power expansion method used in Lax's paper in elegant Hermite Gaussian beams [28] and later in 1998, Laabs applied it to standard Hermite Gaussian beams [29].

In 1979, G.P. Agrawal and D.N. Pattanayak looked into the angular spectrum representation of the electric field [30]. When a beam is focused to a subwavelength spot size, the paraxial approximation no longer holds. They studied a Gaussian beam of such spot size through a dielectric medium. Their numerical results from the paraxial regime agreed with zeroth order solutions to paraxial wave equation and their results from the focused beam agreed with the first order expansion derived by Lax.

2.3 Spin and Orbital Angular Momentum of Light

We can describe the total angular momentum of light to be the combination of its spin angular momentum and orbital angular momentum. Left circular and right circular polarisations are said to be the two spin states of a photon. In the paraxial regime, the eigenstates of helicity are analogous to the left and right circular polarisations. Orbital angular momentum is unrelated to polarisation and is instead a spatial property of the light's propagation. Light with well defined total angular momentum and non-zero orbital angular momentum can be described as “corkscrew light” which means the electromagnetic waves are twisted around its axis of propagation. Destructive interference occurs on the axis of propagation. The result of this is what is called an optical vortex. Its projection onto a flat surface is a ring with no light in the centre. This “hole” becomes larger as the orbital angular momentum is increased.

There are many methods to generate these optical vortices such as mode conversion, a q-plate [31], static spiral phase plates [32], deformable mirrors [33] and holograms [5]. This liquid crystal can be formed into any holographic image desired. These images force the liquid crystal to exhibit a shape with varying refractive indexes forcing any light passing through it to have a desired orbital angular momentum. In figure 2.1, the liquid crystal takes the form of a linear diffraction grating with a fork pattern. The beam's interaction with this structure cause it to diffract into multiple beams and the deflected beams will have non-zero orbital angular momentum.

In 2012, N. Bozinovic, S. Golowich, P. Kristensen and S. Ramachandran presented an alternate method of generating optical vortices using optical fibres [34]. By controlling the polarisation of the fibre, they were able to obtain a Gaussian beam of orbital angular momentum charge 1 or -1. They created optical vortices that are 97% pure after 20m of propagation through a fibre. In order for the fibres to carry light with non-zero orbital angular momentum, they must be able to carry higher order modes. The orbital angular momentum states were shown to be a linear combination of these higher order modes. The light is then passed through a longer fibre with initial twists and turns to attempt to disrupt the signal and the purity of the signal was barely compromised.

It has been shown that the orbital angular momentum of light has no effect on its optical activity. In 2011, dutch physicists, W. Löffler, D.J. Broer and J.P. Woerdman, demonstrated this using a polymer not with a chiral molecular structure but instead arranging it into a crystalline form with a chiral structure [2]. The crystal was split into different planes

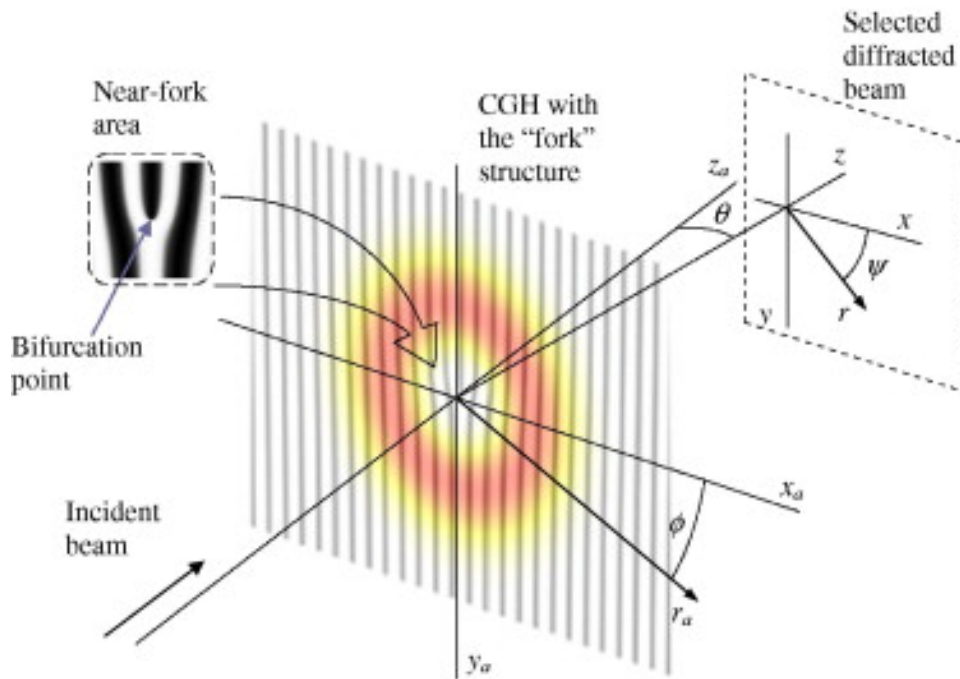


Figure 2.1: Computer generated holography of diffraction grating with fork structure at the centre. Beams carrying orbital angular momentum are deflected by angle θ propagating along the z_0 axis and the zeroth order beam remains along the z axis. Figure taken from [5].

containing chains of these polymers all parallel to each other. Each plane had the chains orientated at a slight angle to the chains in the adjacent plane. The light propagating normal to these planes sees rotating polymer chains producing a helix, a chiral structure. They measured that light with different optical vortices exhibited the same optical activity. Although not the focus of the paper, it is important to know that the light used was highly focused which inherently does not fulfill the paraxial approximation since the beam waist is small compared to the wavelength. This paper is one of few that measures optical activity in the non-paraxial regime.

2.4 Light-Matter Interactions and Circular Dichroism

There are many other instances of circular dichroism separate from optical activity but fewer in the non-paraxial regime. It is necessary to look at these interactions as they are a possibility in an optical activity measurement in the non-paraxial regime. It has been demonstrated that a non-chiral structure can produce circular dichroism [35]. A vortex beam is used to excite a subwavelength circular nanoaperture or *nanohole*. The nanohole is in the shape of a ring, a non-chiral structure. The rings symmetry allows its mirror image to be superimposed

onto itself. The beam had different circular polarisations and large circular dichroism was observed.

Following on from this, a similar experiment using the same nanoaperture in a gold film over a glass substrate showed the transformation of spin angular momentum (helicity) [3]. The sample scattered conserves the total angular momentum of the beam but breaks duality symmetry and therefore, helicity is not conserved. In a scattering case, the transformation of the helicity is entirely dependent on the scattering properties of the structure, so the helicity transformation depends on properties such as the aperture size in the gold film. This ultimately was a demonstration of the control of helicity in light further supported by the simplicity of the experiments design.

Another example of light-matter interaction is the visualisation of the angular momentum of light. In April 2013, a paper was published describing the formation of chiral nanoneedles onto the surface of a 2mm thick tantalum (Ta) plate directly by the orbital angular momentum and spin angular momentum of a 1064nm pulsed beam [36]. The needles themselves take on a spiral shape. Each needles handedness or chirality was determined by the helicity of the beam and the amount of times the needle “spiralled” or spiral frequency was determined by the orbital angular momentum. They concluded that beams with the same total angular momentum are degenerate and create nanoneedles with the same handedness and spiral frequency.

2.5 The Stokes Paramaters

The polarisation of light refers to the direction of the electric field. The simplest electromagnetic wave is a transverse, plane wave. For example, the electric field of a horizontally polarised wave is given by

$$\mathbf{E}_1(\mathbf{r}, t) = E_1 e^{i\mathbf{k} \cdot \mathbf{r} - i\omega t} \hat{x} \quad (2.3)$$

with wave vector $\vec{k}$ and complex amplitude E_1 . The electric field is restricted to the horizontal direction by $\hat{x}$. Similarly for a vertically polarised plane wave:

$$\mathbf{E}_2(\mathbf{r}, t) = E_2 e^{i\mathbf{k} \cdot \mathbf{r} - i\omega t} \hat{y} \quad (2.4)$$

with wave vector $\vec{k}$ and complex amplitude E_2 .

They can be combined to form a generally polarised beam propagating in the direction $\vec{k}$ since any polarisation can be constructed using a linear combination of the horizontal and vertical polarisations.

$$\mathbf{E}(\mathbf{r}, t) = (\hat{x}E_1 + \hat{y}E_2)e^{i\mathbf{k}\cdot\mathbf{r}-i\omega t} \quad (2.5)$$

The complex field amplitudes can be expressed in the forms $E_1 = |E_1|e^{i\phi_1}$ and $E_2 = |E_1|e^{i\phi_2}$ respectively. Note that if these phases ϕ_1 and ϕ_2 are equal, then the wave will have a linear polarisation that makes an angle θ to the x axis. θ is given by:

$$\theta = \arctan \frac{\text{Re}(E_2)}{\text{Re}(E_1)} \quad (2.6)$$

For arbitrary phases that are unequal, the resulting plane wave is elliptically polarised. If the amplitudes are equal and the difference between the phases is $\pm\frac{\pi}{2}$, then the plane wave is circularly polarised and its helicity depends on the sign of that phase difference. The electric wave becomes:

$$\mathbf{E}(\mathbf{r}, t) = E_0(\hat{x} \pm i\hat{y})e^{i\mathbf{k}\cdot\mathbf{r}-i\omega t} \quad (2.7)$$

Generally, the $\hat{x}$ and $\hat{y}$ are arbitrary and can be any two orthogonal vectors. For example, the two complex vectors

$$\vec{e}_{\pm} = \frac{1}{\sqrt{2}}(\hat{x} \pm i\hat{y}) \quad (2.8)$$

representing the positive and negative helicity can be used instead.

$$\mathbf{E}(\mathbf{r}, t) = (\mathbf{e}_+E_+ + \mathbf{e}_-E_-)e^{i\mathbf{k}\cdot\mathbf{r}-i\omega t} \quad (2.9)$$

This shows that any polarisation can also be expressed as a combination of left and right circular polarisations or any two orthogonal polarisations [37]. The superposition of circular polarisations is the key factor in the explanation of circular birefringence and circular dichroism.

In 1852 [20], George Gabriel Stokes defined the stokes parameters as mathematical representation of the polarisation beam regardless of its degree of polarisation. The four parameters are determined through intensity measurements and their values completely characterise the polarisation of the wave.

To find the total intensity, the amplitudes of the two orthogonal components must be

found. Using the transverse wave with arbitrary polarisation in equation 2.5, the complex amplitudes for the horizontal and vertical components are E_1 and E_2 respectively. In general, the horizontal and vertical amplitudes are given by the dot product of the total amplitude E and the directions $\hat{x}$ and $\hat{y}$ respectively. The intensity is then defined as the modulus of these amplitudes.

The first stokes parameter, S_0 is given by the sum of these dot products.

$$S_0 = |\hat{x} \cdot \mathbf{E}|^2 + |\hat{y} \cdot \mathbf{E}|^2 = |E_1|^2 + |E_2|^2 \quad (2.10)$$

The second stokes parameter, S_1 is given by the difference of these dot products.

$$S_1 = |\hat{x} \cdot \mathbf{E}|^2 - |\hat{y} \cdot \mathbf{E}|^2 = |E_1|^2 - |E_2|^2 \quad (2.11)$$

From equations 2.3 and 2.4, these two squared amplitudes are defined as the horizontal and vertical intensities of a transverse plane wave. So the stokes parameters become

$$S_0 = I_H + I_V \quad (2.12)$$

$$S_1 = I_H - I_V \quad (2.13)$$

where I_H and I_V are the horizontal and vertical intensities respectively.

This derivation can also be applied using different basis vectors such as the complex helicity vectors which would result in the last stokes parameter S_3 being given by

$$S_3 = I_L - I_R \quad (2.14)$$

where I_L and I_R are the left and right circular intensities respectively. Also S_0 is given by

$$S_0 = I_L + I_R \quad (2.15)$$

Lastly, this can be applied to the diagonal and antidiagonal vectors, which are the horizontal and vertical base vectors rotated by 45° and we obtain the remaining stokes parameter S_2 given by

$$S_2 = I_D - I_A \quad (2.16)$$

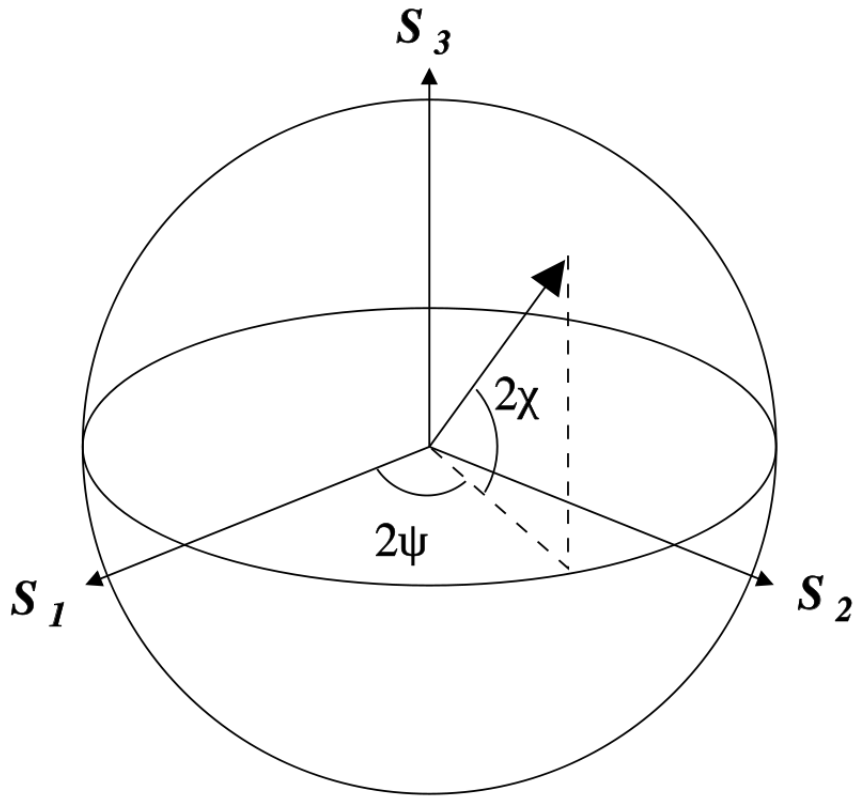


Figure 2.2: The Poincaré sphere which expresses three of the stokes parameters as a set of spherical coordinates.

for diagonal and antidiagonal intensities I_D and I_A . Note that S_0 is given by the sum of any of the pairs of orthogonal intensities. The stokes parameters are bound by the relation:

$$S_0^2 = S_1^2 + S_2^2 + S_3^2 \quad (2.17)$$

As a result the stokes parameters can be mapped onto spherical coordinates of a sphere of radius S_0 called the Poincaré Sphere as shown in figure 2.2.

The spherical coordinates are as follows:

$$S_0 = I \quad (2.18)$$

$$S_1 = Ip \cos 2\psi \cos 2\chi \quad (2.19)$$

$$S_2 = Ip \sin 2\psi \cos 2\chi \quad (2.20)$$

$$S_3 = Ip \sin 2\chi \quad (2.21)$$

where p is the degree of polarisation and $0 \leq p \leq 1$. I is the total intensity of the beam. ψ and χ are shown in figure 2.2.

The stokes parameters can be combined into a vector known as the *stokes vector* given by

$$\mathbf{S} = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} \quad (2.22)$$

This vector can span the entire space of both unpolarised and polarised light.

From the spherical coordinates, we can extract the azimuthal angle ψ from equations 2.19 and 2.20. By dividing equation 2.20 by equation 2.19 we get

$$\begin{aligned} \frac{S_2}{S_1} &= \frac{Ip \sin 2\psi \cos 2\chi}{Ip \cos 2\psi \cos 2\chi} \\ \frac{S_2}{S_1} &= \tan 2\psi \\ \psi &= \frac{1}{2} \arctan \frac{S_2}{S_1} \end{aligned} \quad (2.23)$$

Horizontal polarisation is defined the have an azimuthal angle of 0° . While diagonal and antidiagonal have azimuthal angles of 45° and -45° respectively.

We can also extract the ellipticity χ from the spherical coordinates by first adding the squares of equations 2.19 and 2.20.

$$\begin{aligned} S_1^2 + S_2^2 &= I^2 p^2 \cos^2 2\chi (\cos^2 2\psi + \sin^2 2\psi) = I^2 p^2 \cos^2 2\chi \\ \sqrt{S_1^2 + S_2^2} &= Ip \cos 2\chi \end{aligned} \quad (2.24)$$

Then dividing equation 2.21 by 2.24, we obtain:

$$\begin{aligned} \frac{S_3}{\sqrt{S_1^2 + S_2^2}} &= \frac{Ip \sin 2\chi}{Ip \cos 2\chi} \\ \frac{S_3}{\sqrt{S_1^2 + S_2^2}} &= \tan 2\chi \end{aligned}$$

$$\chi = \frac{1}{2} \arctan \frac{S_3}{\sqrt{S_1^2 + S_2^2}} \quad (2.25)$$

Lastly, the stokes parameters can be normalised by the total intensity. Starting with S_1 , the normalised stokes parameter s_1 labeled using a lower case 's' instead is given by

$$s_1 = \frac{S_1}{I} = \frac{S_1}{S_0} \quad (2.26)$$

But S_0 equals the sum of the horizontal and vertical intensities I_H and I_V . So s_1 is given by:

$$s_1 = \frac{I_H - I_V}{I_H + I_V} \quad (2.27)$$

Similarly, for normalised stokes parameters s_2 and s_3 .

$$s_2 = \frac{I_D - I_A}{I_D + I_A} \quad (2.28)$$

$$s_3 = \frac{I_L - I_R}{I_L + I_R} \quad (2.29)$$

Now, the relations between the normalised stokes parameters becomes:

$$s_1^2 + s_2^2 + s_3^2 = 1 \quad (2.30)$$

Solving equation 2.26 for S_1 gives:

$$S_1 = S_0 s_1 \quad (2.31)$$

Similarly for S_2 .

$$S_2 = S_0 s_2 \quad (2.32)$$

Subbing these into equation 2.23 gives:

$$\begin{aligned} \psi &= \frac{1}{2} \arctan \frac{S_0 s_2}{S_0 s_1} \\ \psi &= \frac{1}{2} \arctan \frac{s_2}{s_1} \end{aligned} \quad (2.33)$$

All of the normalised stokes parameters have values between -1 and 1. When $s_1 = 1$ and $s_2 = s_3 = 0$, the horizontal intensity I_H is maximised, the vertical intensity I_V is minimised and $I_D = I_A = I_L = I_R$. This means the beam is characterised as horizontally polarised. For

$s_1 = -1$ $s_2 = s_3 = 0$, the beam is vertically polarised. Similarly, $s_1 = s_3 = 0$ characterises the beam as diagonally and antidiagonally polarised for $s_2 = 1$ and $s_2 = -1$ respectively. Where as $s_1 = s_2 = 0$ characterises the beam as left or right circularly polarised for $s_3 = 1$ and $s_3 = -1$ respectively.

In this thesis, the normalised stokes parameters s_1 , s_2 and s_3 will be calculated from beam intensities to completely characterise the polarisation of the beam. The common sugars sucrose and maltose will rotate any linear polarisation. This angle of rotation can be calculated in the form of the change in the azimuthal angle ψ of the beam. The beam does not undergo any change in circular polarisation.

3

The Paraxial Regime

In this chapter, I illustrate the methods and solutions I have developed to build an experimental apparatus that measures the optical activity of a chiral sample in the paraxial regime. The aim is to verify whether the detection equipment of choice are suitable for optical activity measurements. The results gathered will be used as a control when compared to the measurements in the non-paraxial regime.

3.1 Experimental Setup

The laser used is a Toptica DL 100 tunable single mode diode laser with wavelength range of 765-810nm. Maximum operating power is 124mW occurring at 790nm. The beam is transported into the setup through an optic fibre. Its power is controlled by passing through multiple density filters that absorb varying amounts of the beam resulting in the same beam with a fraction of its original power. These filters absorb a large percentage of the intensity ranging from 50% all the way up to 99.997% and any combination of these.

Safety Class	Class 3B
Power Output	3 - 126 mW
Maximum Power Output (at 790.4nm)	126 mW
Operating Temperature	15 - 30 °C
Minimum Wavelength	762.3 nm
Maximum Wavelength	810.6 nm
Optical Spectrum Width	0.05 nm
Polarisation Ratio	100:1 (Linear)
Beam Diameter	3 mm
Weight	2.3 kg

Table 3.1: Toptica DL 100 pro design 790nm laser specifications.

The beam's polarisation is controlled using a half waveplate (WPH05M-808, Thorlabs) and quarter waveplate (WPQ05M-808, Thorlabs). The power of the incident beam was reduced by 99.997% using the density filters. The final beam output after the filters was approximately 0.6 μ W. After the half and quarter waveplates, the beam reflects off numerous mirrors (BB1-E03, Thorlabs) and is then redirected onto the paraxial regime path where it reflects off two mirrors before interacting with the chiral sample. Each mirror performs a unitary transformation on the beam's polarisation. To compensate for this effect, the polarisation is measured after the last reflection immediately before the sample and the waveplates are orientated in such a way that this initial polarisation is horizontal. This allows the waveplates to be placed anywhere in the setup before the sample as long as the polarisation of the beam is characterised after the last reflection.

The chiral sample is contained within a quartz cuvette (Hellma 101-OS) of beam path lengths 1, 2, 5 and 10mm. Aside from the 5mm cuvette, each one was found to exhibit no optical activity in the collimated path. However, the 5mm cuvette was defective in that it displayed optical activity, therefore it was not used. The cuvettes are placed and then fixed on a stand normal to the incident beam. The beam interacts with the sample and its polarisation is measured.

The collimated beam is collected by a polarimeter (PAX5710IR1 - TXP Polarimeter Module, Thorlabs). The polarimeter is used because of its ability to completely characterise a beam's polarisation by the normalised stokes parameters making it a great starting point. It can characterise beams of wavelengths in the range 700-1000nm which completely covers the range of wavelengths the laser can output (see table 3.1). The density filters reduced the beam's output power to such an extent as to not oversaturate the polarimeter. A simplified

form of the setup is shown in figure 3.1.

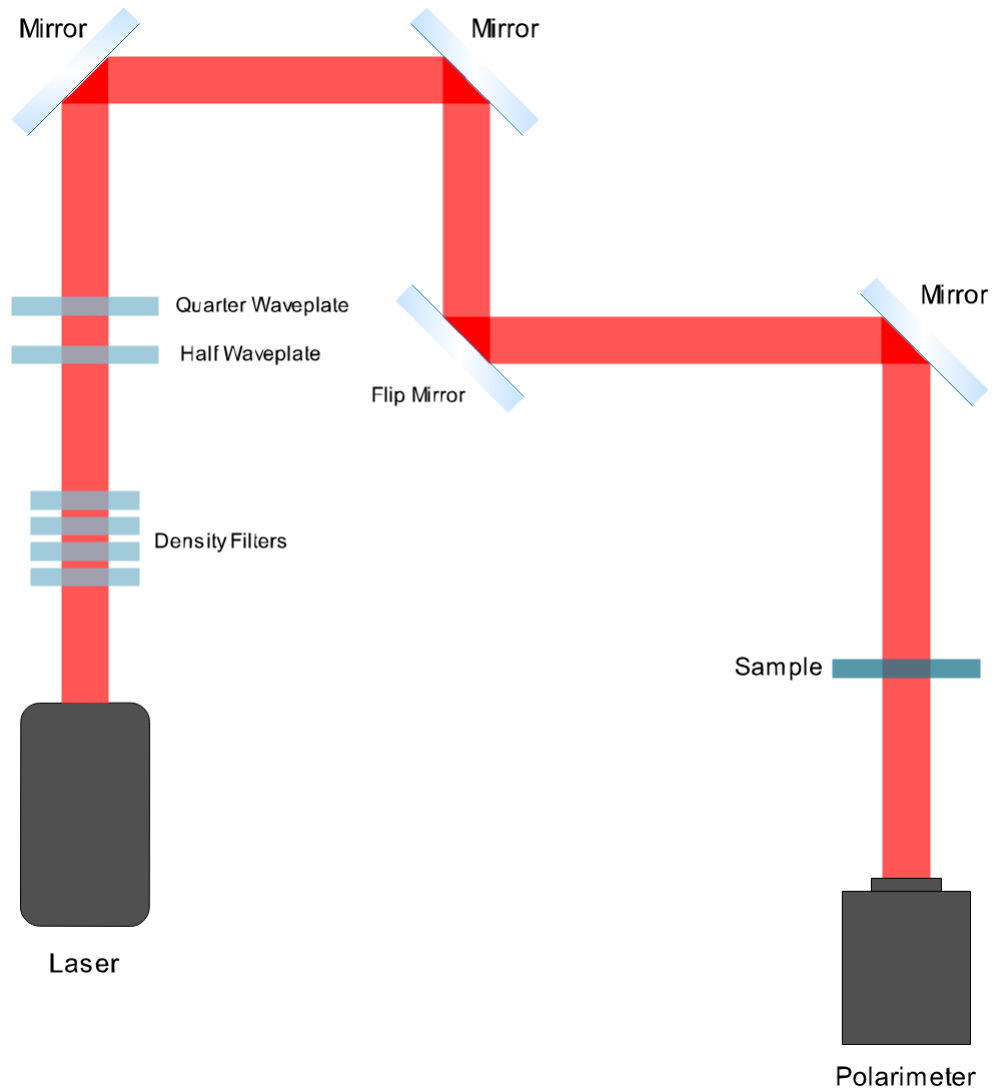


Figure 3.1: The experimental apparatus for the paraxial path only. The flip mirror is set such that the beam is deflected onto this path.

3.2 Results

The *azimuthal angle* of the polarisation of the beam, ψ is given in equation 2.33 for normalised stokes parameters s_1 and s_2 given by equations 2.27 and 2.28 respectively.

An azimuthal angle of 0 degrees and no ellipticity implies that the beam is horizontally polarised. Any change in the azimuthal angle would indicate optical activity. The TXP software automatically calculates the azimuthal angle from the stokes parameters. This angle has been verified using equation 2.33.

Each polarimeter measurement is a timed sequence of 1024 separate measurements over 30 seconds. The data recorded contains the time stamp, the normalised stokes parameters, degree of polarisation (%), azimuthal angle (degrees), ellipticity (degrees) and the power (db) of each of the 1024 measurements. In all the following cases in the paraxial regime, each point on the graphs is the average of three sets of measurements. The error values shown are calculated using the standard deviation of the azimuthal values of the sequence.

Optical rotation is calculated by taking the difference in azimuthal angles of pure water and solutions of sugars. Figures 3.2 and 3.3 plot the optical rotation of sucrose and maltose solutions respectively. A clear linear dependence of the optical activity on the beam path length emerges in both the samples of sucrose and maltose.

In figures 3.4 and 3.5, the azimuthal angle of each sugar is calculated as a function of concentration in a constant beam path of 2 mm. Once again, a clear linear dependence of the optical activity emerges but on the concentration of the sugars instead. These results are a clear demonstration of the linear dependence of optical activity on the beam path length and the concentration of the solution. However, the use of only three cuvettes of different thicknesses limits the accuracy of measuring the relationship between optical activity and beam path length. The linear regression approximation is still within error in both sugars.

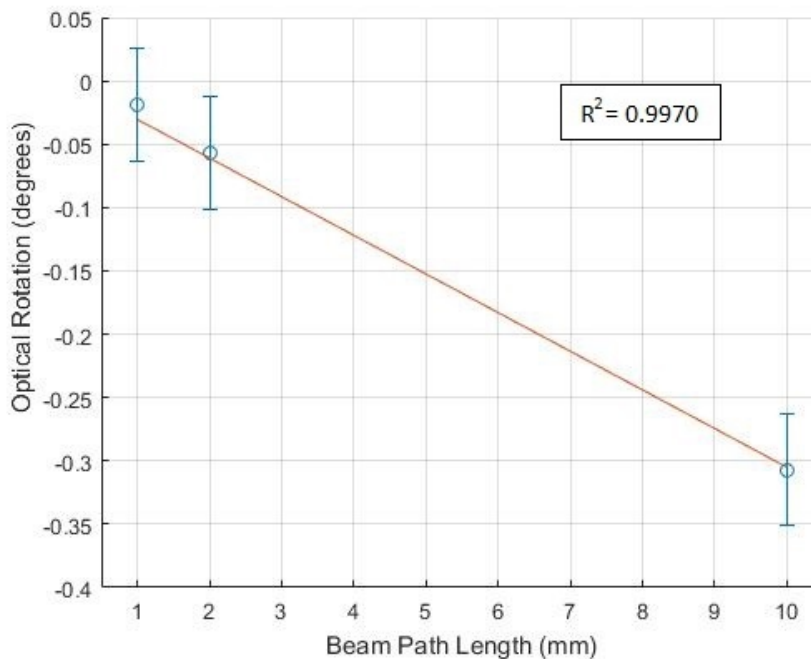


Figure 3.2: Optical rotation of sucrose solution with concentration 1 g/10mL in the cuvettes of thicknesses 1, 2 and 10mm. The coefficient of determination R^2 of the linear regression is 0.9970.

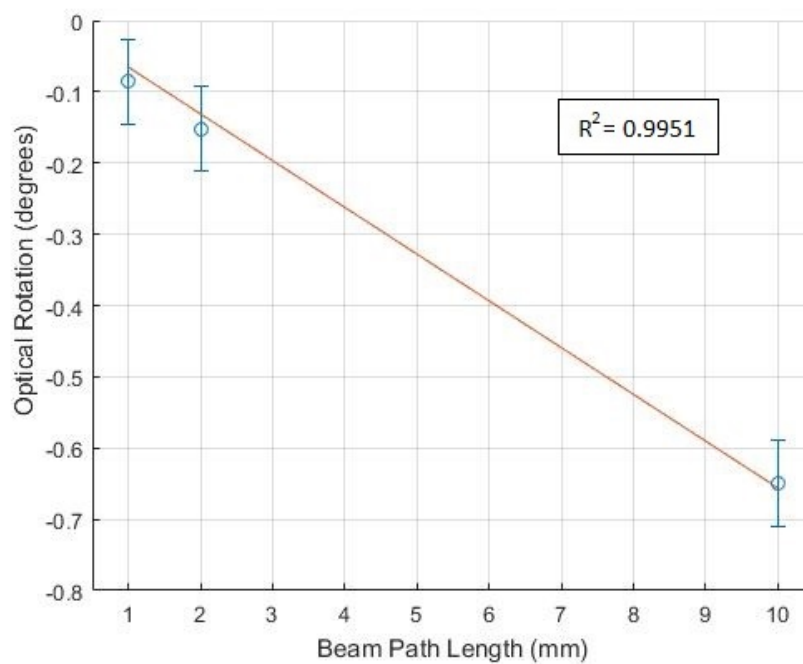


Figure 3.3: Optical rotation of maltose solution with concentration 1 g/10mL in the cuvettes of thicknesses 1, 2 and 10mm. The coefficient of determination R^2 of the linear regression is 0.9951.

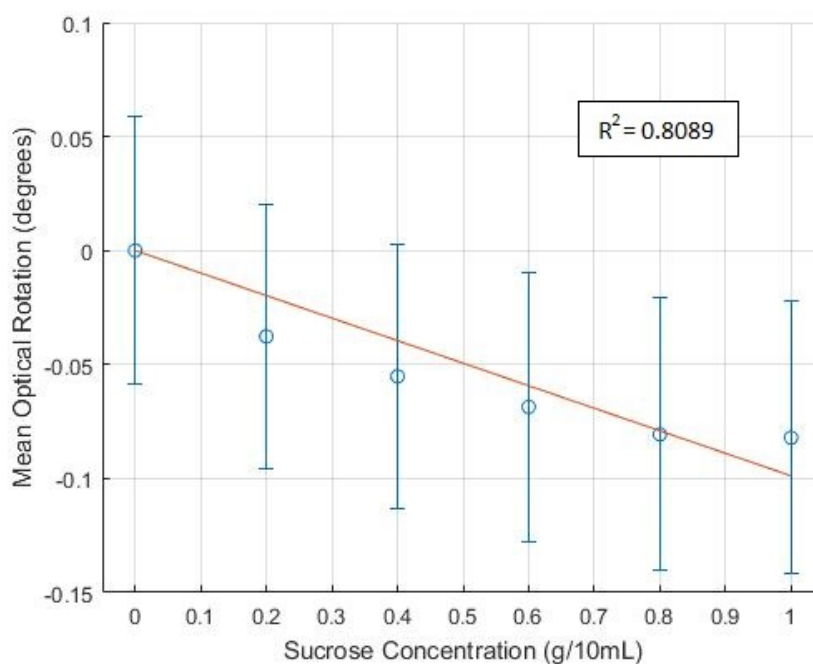


Figure 3.4: Optical rotation of sucrose solutions with various concentrations and water in the 2mm thick glass cuvette. The coefficient of determination R^2 of the linear regression is 0.8089.

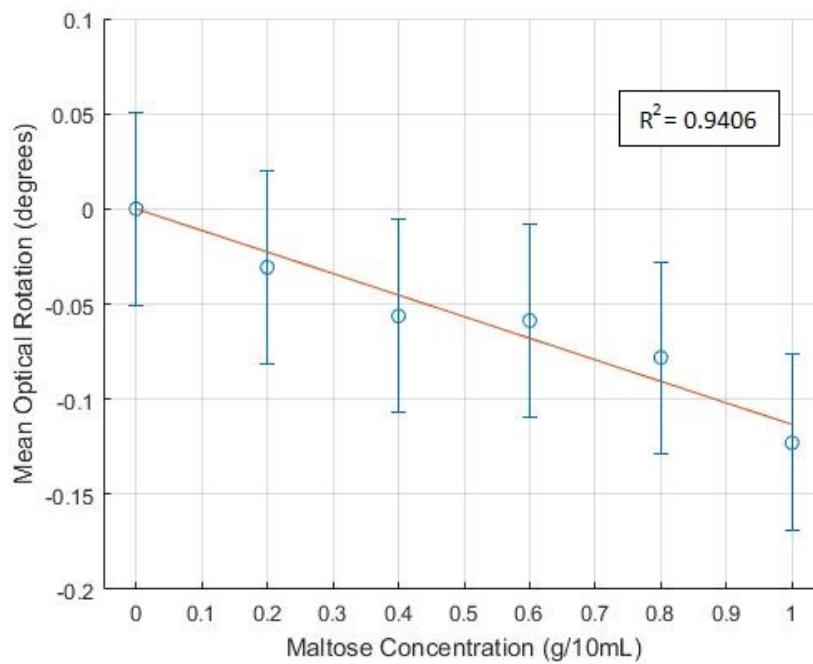


Figure 3.5: Optical rotation of maltose solutions with various concentrations and water in the 2mm thick glass cuvette. The coefficient of determination R^2 of the linear regression is 0.9406.

In conclusion, the polarimeter was sufficient to show the optical activity of sucrose and maltose. It shows its linear dependence on beam path length and concentration. Because of this, it is the first detection device used in the non-paraxial regime.

4

The Non-Paraxial Regime

In this section, I summarise the numerous methods using various equipment to measure optical activity of sucrose and maltose in the non-paraxial regime. In all methods, the sample and microscope objectives remain constant but the equipment used to capture the beam changes. The initial microscope objective (LMPLFLN50X, Olympus), called the condenser lens, has a numerical aperture of 0.5 and the final microscope objective (MPLFLN20X, Olympus), called the objective lens, has a numerical aperture of 0.45. The condenser lens has a much longer working distance than the objective lens. The specifications for each microscope objective are in table 4.1.

	Magnification	NA	Working Distance (mm)	Field Number	Transmittance 400-700nm
LMPLFLN50X	50x	0.5	10.6	26.5	>90%
MPLFLN20X	20x	0.45	3.1	26.5	>90%

Table 4.1: Olympus microscope objective specifications according to the manufacturer. Note the exact transmission at 790nm is not tabulated.

Optical activity does not depend on intensity and to confirm this in the non-paraxial regime the cuvettes of 1 and 2mm containing the samples are put into a translational stage along the axis of propagation. These are the same cuvettes used in chapter 3 that were shown to have no optical activity in the paraxial regime. The high numerical aperture microscope objectives use coatings and materials which can modify the beam's polarisation. Therefore, the polarisation of the beam was measured both before and after the objectives and it was found that they had no effect on the polarisation. This allows the sample to be moved into and past the focal point of the microscope objectives which occurs at about 10.6mm from the condenser lens in air and slightly further when a glass cuvette is present. The cuvette's glass walls are 1.25mm thick making the total size of each cuvette 2.5mm larger than the given beam path length. The 10mm thick cuvette was not used for this very reason reaching a total length of 12.5mm which is only 1mm less than the separation of the two microscope objectives, the sum of the working distances ($10.6\text{mm} + 3.1\text{mm} = 13.7\text{mm}$). No translation was possible and the focus of the beam would always be inside.

The existence of the cuvettes between the condenser and objective lenses changes the optical path length so the positions of the microscope objectives are changed to maintain a collimated transmitted beam. The cuvette's glass walls have a refractive index of 1.55 and water or any solution of sucrose and maltose has a refractive index of 1.33. For thicker cuvettes, the focus of the beam moves further away from the condenser lens. This means that the control measurement cannot be an empty path or even an empty cuvette. The control measurement must be a cuvette filled with water because water has the same refractive index as the solutions of sugars. Therefore, the relative distance between the condenser and objective lenses is set in order to obtain a collimated beam at the output of the objective lens. Thus, for the 2mm optical path cuvette the distance between the objectives is larger than the distance for the 1mm optical path. This is demonstrated in figure 4.1, showing that in a thicker cuvette, the beam path has a higher refractive index since there is less air and more water or sugar solution. Thus, in order for the beam to be collimated after the objective lens, the distance between the condenser and the objective lenses must be increased.

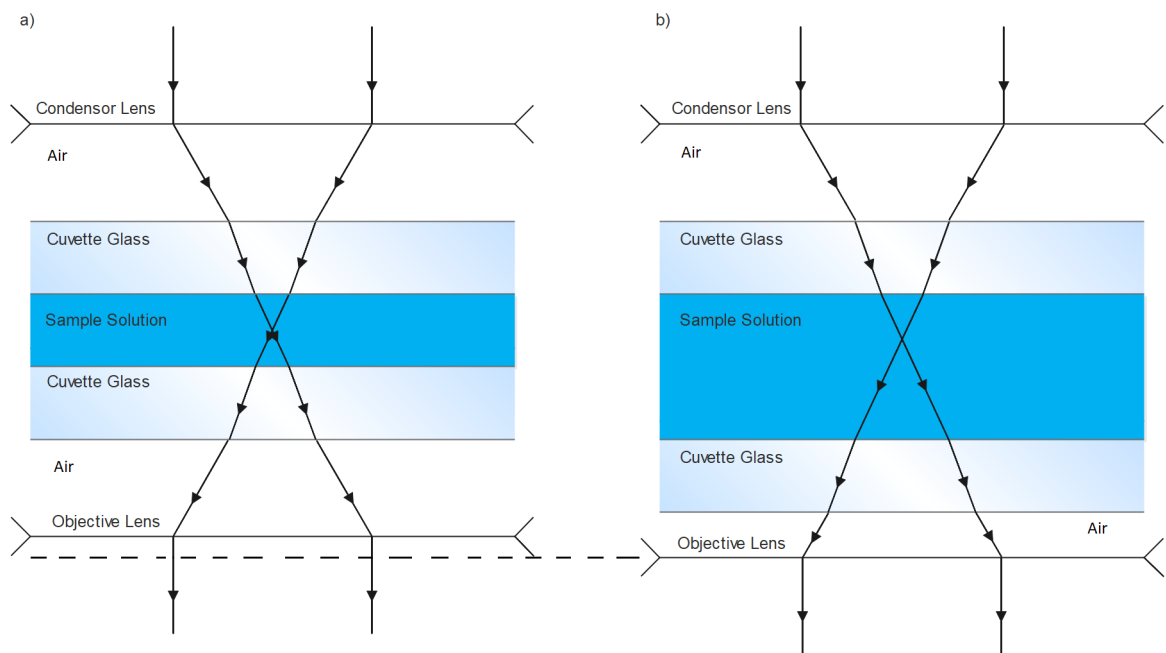


Figure 4.1: Ray diagram of the beam as it is focused into a cuvette, a), and another cuvette twice as thick, b). The sample solution and the cuvette glass have refractive indices 1.33 and 1.55 respectively. Therefore, in b), the distance between the condenser and objective lenses must be increased in order to offset the beam path consisting of more water or solution and less air.

4.1 First Setup - Polarimeter

Similarly from the collimated path, the first method for measuring the optical activity was using the polarimeter. The polarimeters capability of completely characterising the beam's polarisation with the stokes parameters was convenient and its capabilities were found to be sufficient based off the paraxial regime measurements. As before, the beams output power was greatly reduced to prevent oversaturation. The apparatus is shown in figure 4.2. The flip mirror is no longer deflecting the beam allowing it to continue down the optical axis of the condenser lens as compared with figure 3.1. The half and quarter waveplates now have two different configurations to characterise the beam's polarisation for both cuvettes. The waveplates are configured to change the polarisation to horizontal. After the beam interacts with the sample, it is collimated again and transmitted straight into the polarimeter. The focal point occurs around 3mm before the objective lens. Thus, when the cuvette containing the sample is put into the translation stage between the objectives, its initial position is as close to the objective lens as possible. The optical activity is measured in 50 micron steps back towards the condenser lens.

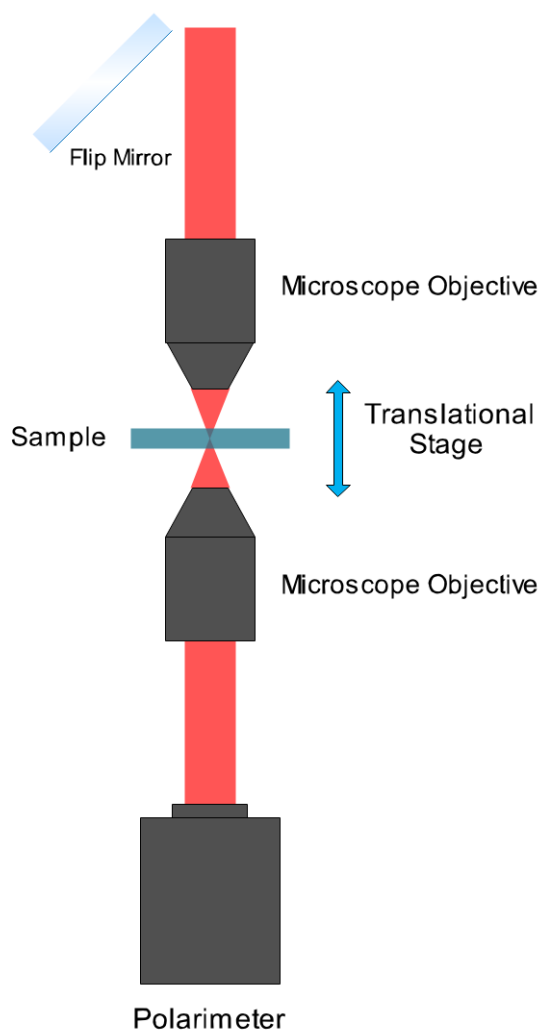


Figure 4.2: Schematic of the first apparatus which uses the polarimeter to characterise the beam's polarisation.

Figures 4.3 and 4.4 are plots of the azimuthal angle of the beam's polarisation as the 1mm cuvette is translated through the optical path. The initial translation stage position is labeled as 5mm, right at the end of the path between the objectives and it is translated back towards the condenser lens which is designated at 0mm. When the cuvette contains water, the azimuthal angle peaks at the 2mm mark and the 4.25mm mark. Similarly, when the cuvette contains the sugar solution, it peaks at the 1.75mm mark and 4mm mark. It is noteworthy that the distance between these two peaks in both samples are very similar at around 2.25mm. If the thickness of one of the glass walls is added to the size of the cuvette, a value of 2.25mm is also obtained. This means that the peaks in the azimuthal angle show when the focal point is in concurrent locations within the glass walls of the cuvette, the first interface of each wall or the second interface of each wall.

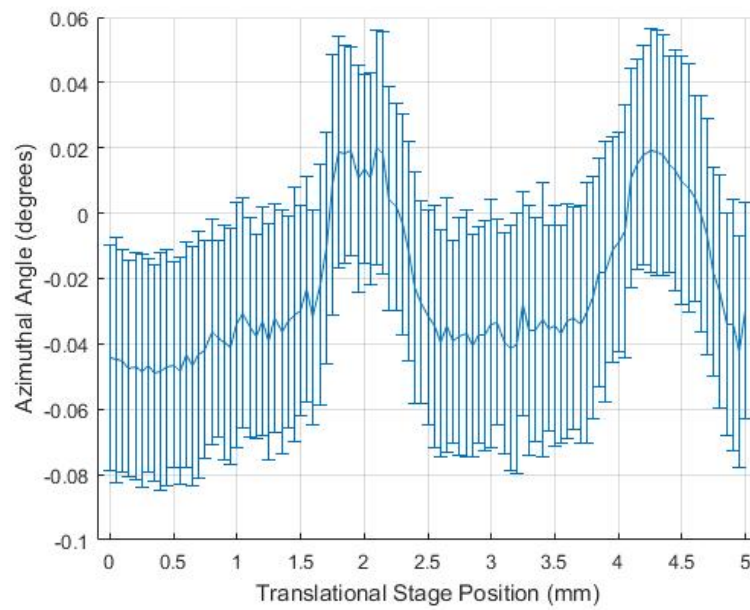


Figure 4.3: Azimuthal angle of the beam through water as a function of the displacement of the cuvette between the condenser and objective lenses. The cuvette containing the water is 1mm thick.

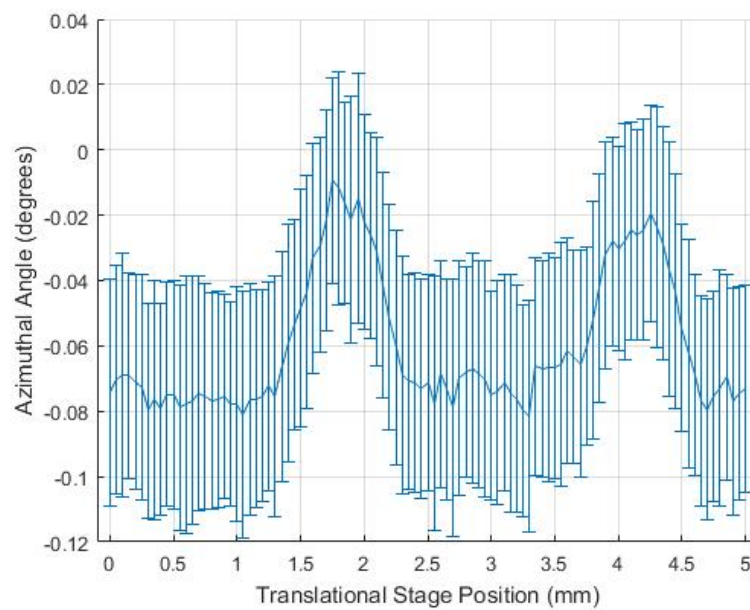


Figure 4.4: Azimuthal angle of the beam through a sucrose solution of concentration 1g/10mL as a function of the displacement of the cuvette between the condenser and objective lenses. The cuvette containing the sucrose solution is 1mm thick.

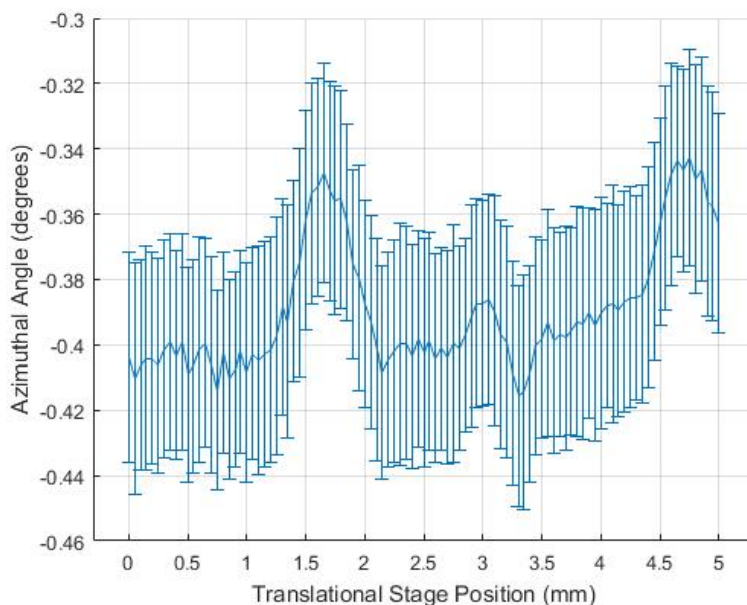


Figure 4.5: Azimuthal angle of the beam through a maltose solution of concentration 1g/10mL as a function of the displacement of the cuvette between the condenser and objective lenses. The cuvette is 2mm thick.

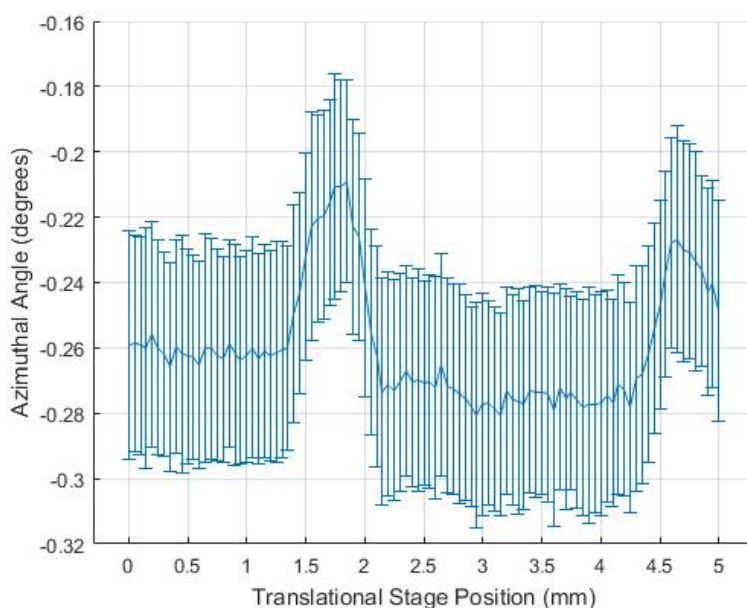


Figure 4.6: Azimuthal angle of the beam through a maltose solution of concentration 0.2g/10mL as a function of the displacement of the cuvette between the condenser and objective lenses. The cuvette containing the sucrose solution is 2mm thick.

Figures 4.5 and 4.6 plot the azimuthal angle of the beam as the 2mm cuvette is translated through the optical path. The azimuthal angle continues to peak even with a different sugar solution. The peaks in all cases are very similar in size between 0.05 and 0.06 degrees, even

for the azimuthal angle of the beam through water. However, this time they are 1mm further apart. This further supports the peaks existence in concurrent locations within the glass walls of the cuvette even if the cuvette is thicker.

The polarimeter was capable of showing the change in the azimuthal angle when increasing the concentration of the sugar solutions but the appearance of these peaks proved problematic. Figure 4.3 shows that they even appear in water, which is known to not exhibit any kind of optical activity. It seemed to indicate that the optical activity depends on the position of the focus of the beam within the glass walls. Because of this, further detection methods and equipment were required to address the peak appearance.

4.2 The Second Setup - Small Area Detectors

The next method for measuring the optical activity utilises a polarised beam splitter. The apparatus is shown in figure 4.7. The incident beam no longer has its power reduced by the density filters and its polarisation is set to diagonal after the last reflection using the waveplates. A polariser (GL10, Thorlabs) is used to ensure the purity of the beam's polarisation and is aligned to maximise the power output for a diagonally polarised beam.

After interaction with the sample, the beam passes through a half waveplate and a quarter waveplate. The beam then passes through a polarised beam splitter (PBS202, Thorlabs) orientated to transmit horizontally polarised light and reflect vertically polarised light. The polarised beam splitter transmits 95.75% of p-polarised beams and 0.05915% of s-polarised beams at wavelength 790nm. The two beams are focused and then collected using small area photodetectors (PDB440C, Thorlabs) which measure the beams intensity as a voltage. The small capture area allows for fast response times to see any fast fluctuations in the beam, rise time 10ns. An oscilloscope is used to display the detectors' electric signals. The oscilloscope is set at a measuring speed of 1000Hz running for 10 seconds. Thus, there are 10,000 acquisitions for each measurement. The oscilloscope allows the simultaneous measurement of both beams to observe these fast fluctuations of intensity in time in order to reduce errors and artifacts.

The waveplates and beam splitter combined are used to determine the three normalised stokes parameters s_1 , s_2 and s_3 . Therefore, the waveplates will have three configurations:

1. The first waveplate configuration orientates them in such a way that it does not change

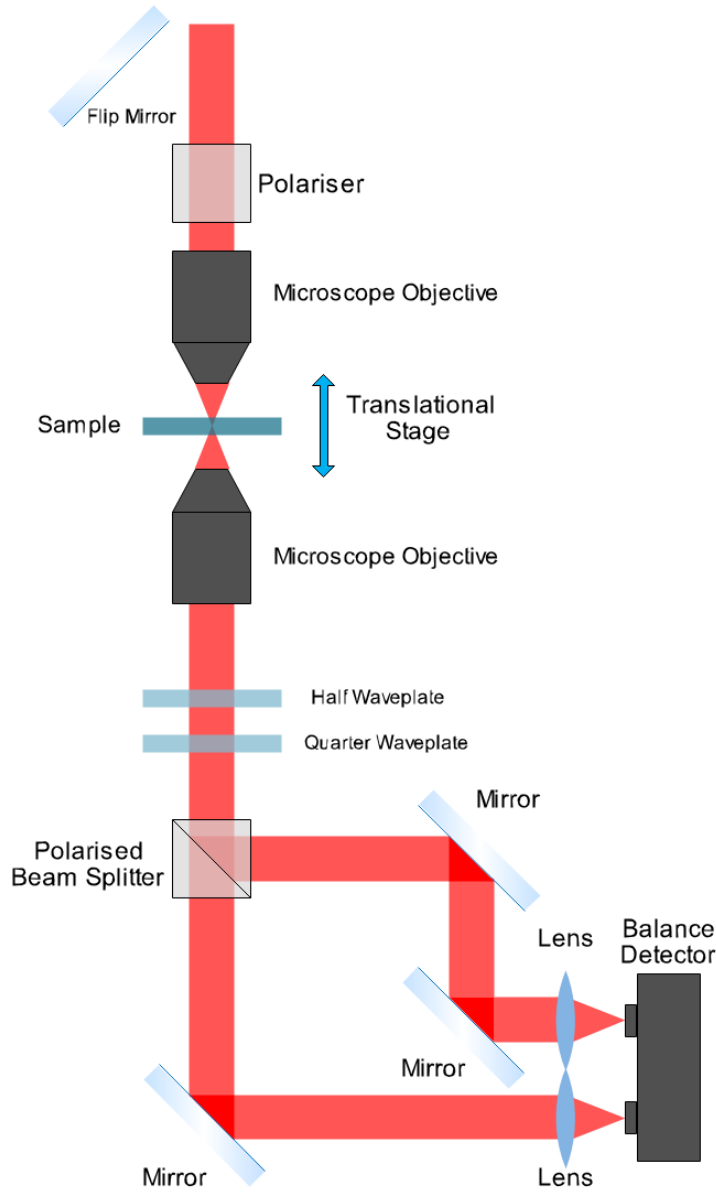


Figure 4.7: Schematic of the second apparatus which splits the beam in accordance with its polarisation and captures both beams using a small area photodetectors.

the polarisation of the beam. This keeps the polarisation in the horizontal-vertical basis meaning that the beam splitter will transmit horizontal polarisations and reflect vertical polarisations.

In case of the control polarisation (diagonal) in water, half the light is transmitted and half is reflected since diagonally polarised light is an equal superposition of horizontal and vertical polarisations. The measured intensities of the two split beams are used to determine s_1 by this equation:

$$s_1 = \frac{I_H - I_V}{I_H + I_V} \quad (4.1)$$

where I_H and I_V are the intensities of the transmitted and reflected beams respectively.

2. The second orientates the waveplates to change the basis of the polarisation of the beam to the diagonal-antidiagonal basis. This means that the polarised beam splitter will transmit diagonally polarised light and reflect antidiagonally polarised light.

In case of the control polarisation (diagonal) in water, almost all the light will transmit through the beam splitter and be picked up by one of the detectors and almost no light will be reflected. The measured intensities of the split beams are used to calculate s_2 by this equation:

$$s_2 = \frac{I_D - I_A}{I_D + I_A} \quad (4.2)$$

where I_D and I_A are the intensities of the transmitted and reflected beams respectively.

3. The last orientates the waveplates to change the basis of the polarisation of the beam to the left-right circular basis and the beam splitter will transmit left circular polarisations and reflect right circular polarisations.

Since all linear polarisations are an equal superposition of left and right circular polarisations, only with different phase, the intensity of the control polarisation (diagonal) in water will approximately be split in half by the beam splitter. The measured intensities of the two split beams are used to determine s_3 by this equation:

$$s_3 = \frac{I_L - I_R}{I_L + I_R} \quad (4.3)$$

where I_L and I_R are the intensities of the transmitted and reflected beams respectively.

It is necessary to measure these six intensities directly to observe whether the peaks in azimuthal angle in the polarimeter are real effects. According to equation 2.33, the azimuthal angle is calculated from the first two stokes parameters s_1 and s_2 . Peaks in the azimuthal angle would also reflect peaks in the stokes parameters. So, if these peaks are due to optical activity we would see that the intensity of the transmitted beam would increase and the intensity of the reflected beam would decrease and vice versa. Therefore, it is necessary to measure the intensities directly to observe this behaviour. Thus, this detection method uses two identical small area detectors, waveplates and polarised beam splitter instead of the polarimeter.

Figures 4.8 and 4.9 show the splitting of the intensities of the transmitted and reflected beams in the horizontal-vertical basis. So the transmitted and reflected beams represent

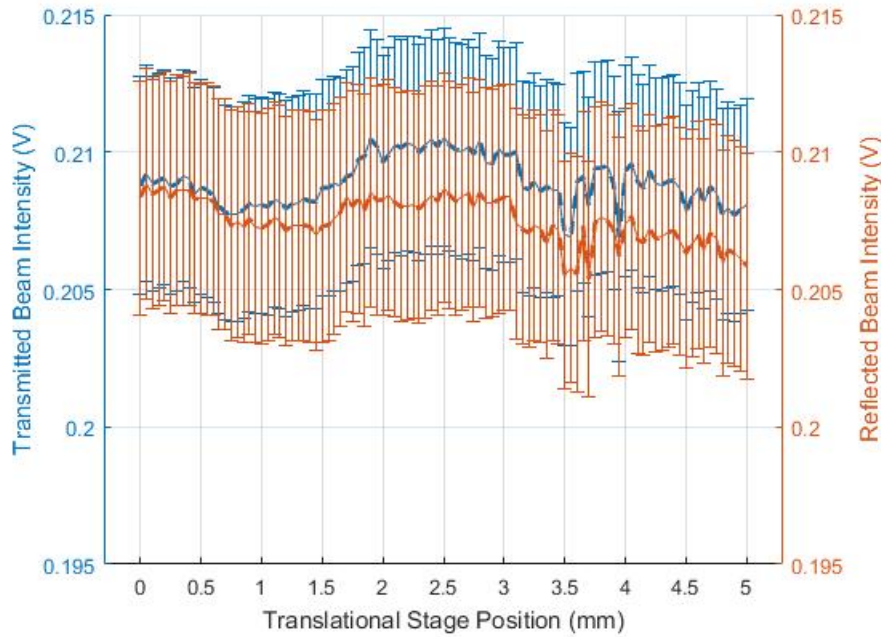


Figure 4.8: The intensities of the transmitted and reflected beams in the horizontal-vertical basis of a diagonally polarised beam through a 2mm cuvette containing water as a function of the displacement of the cuvette between the condenser and objective lenses in the non-paraxial regime. The thicker lines are the intensities and the thinner lines are the standard deviation error.

the horizontal and vertical intensities respectively. The beam's polarisation is aligned to be diagonal using the 2mm thick cuvette filled with water resulting almost equivalent intensities for both the transmitted and reflected beams. Then, the water is replaced by a sucrose solution. The horizontal intensity increases while the vertical intensity decreases which shows that the azimuthal angle of the beam has decreased. As above, these plots demonstrate the peaks shown in the polarimeter setup but instead now they are dips in intensity. Also, both the transmitted and the reflected intensities dip by a similar voltage resulting in them having the same intensity profile. This confirmed that these dips in intensities are artifacts and are not manifestations of optical activity. Any optical activity effects on the azimuthal angle of a linear polarisation would cause one intensity to rise and the other to fall by a similar amount. This artifact would manifest in a peak in one intensity and a trough in the other but this is not the case.

This artifact is further shown in figure 4.10. Instead of a cuvette, a glass microscope objective slide is put inside the translational stage to simulate a cuvette with one glass wall instead of two. Clearly, only one peak manifests. This supports the fact that the peaks occur when the focal point of the beam is inside the glass walls as there is only one glass wall in

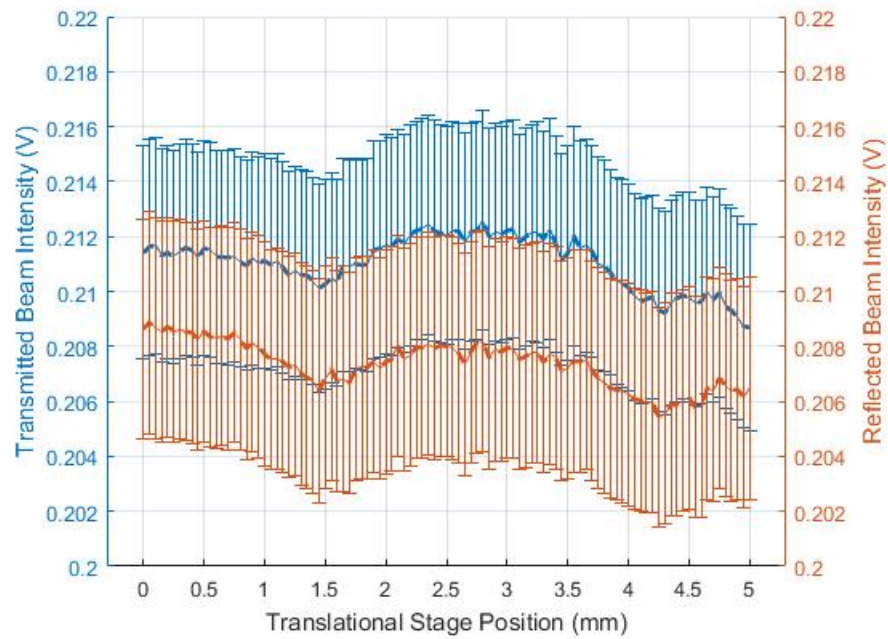


Figure 4.9: The intensities of the transmitted and reflected beams in the horizontal-vertical basis of a diagonally polarised beam through a 2mm cuvette containing a sucrose solution of concentration 1g/10mL as a function of the displacement of the cuvette between the condenser and objective lenses in the non-paraxial regime.

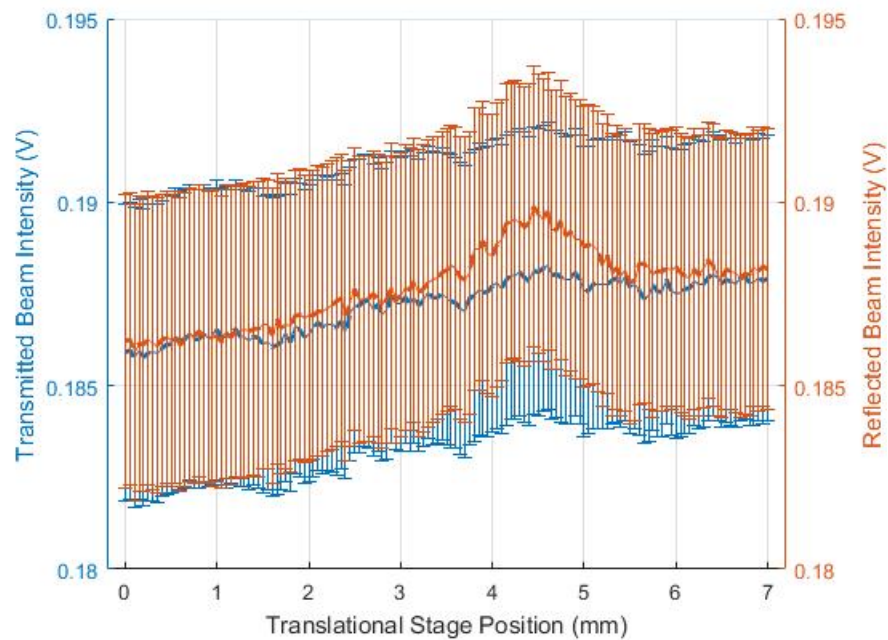


Figure 4.10: The intensities of the transmitted and reflected beams of a diagonally polarised beam through a microscope objective slide as a function of distance.

this case.

The small area detectors showed that the peaks are not manifestations of optical activity.

For each intensity measurement per point on the translational stage, both intensities dip in same direction by approximately by the same amount. Thus, the difference between the two intensities remained approximately the same. However, an alignment issue arose. A majority of the intensity measurements displayed a general upward or downward trend as the cuvette was translated. The cuvette placement between the condenser and objective lenses was likely imperfect with regards to whether the beam was indeed normal to the cuvette. The beam and the cuvette might not have been perfectly perpendicular to each other. So when the cuvette is translated and the focus changes position within the cuvette, the beam propagates in a slightly different direction. The detectors have a notably small area to collect light even if the beam is focused. Any small change in alignment due to translation would move the beams slightly off or onto the small area of the detectors and the intensity would increase or decrease as a result. Thus, one more method of detection is considered.

4.3 The Last Setup - Large Area Detectors

The last setup is very similar to the second apparatus but instead has two separate photodetectors (PDA36A-EX Si Amplified Photodetector, Thorlabs) at the end of the transmitted and reflected beam paths. The final schematic of the non-paraxial regime is shown in figure 4.11. The beam's polarisation is still set to diagonal. These photodetectors have a larger detection area and are large enough to fit the entire beam width after the focusing lenses. It also addresses the main alignment problem with the previous setup. Even if the beam is redirected due to imperfect alignment and the translation of the cuvette, it still will entirely fall within the area of detection of these new photodetectors and there is no reduction or increase in the intensity measurements. A third photodetector before the interaction (DET10A Si Biased Detector, Thorlabs) is used to measure any power fluctuations in the laser and back reflections from the cuvette's different displacements between the condenser and objective lenses. A cover slip is used to reflect a small percentage, about 2.2%, of the beam for this detector. The intensity of the beam from this detector remained stable throughout the following measurements and did not play any role in affecting the optical activity of the beam. An example of this is shown in figure 4.12.

Similarly to the previous setup, the half and quarter waveplates transform the beam's polarisation into the three bases: horizontal-vertical, diagonal-antidiagonal and left-right

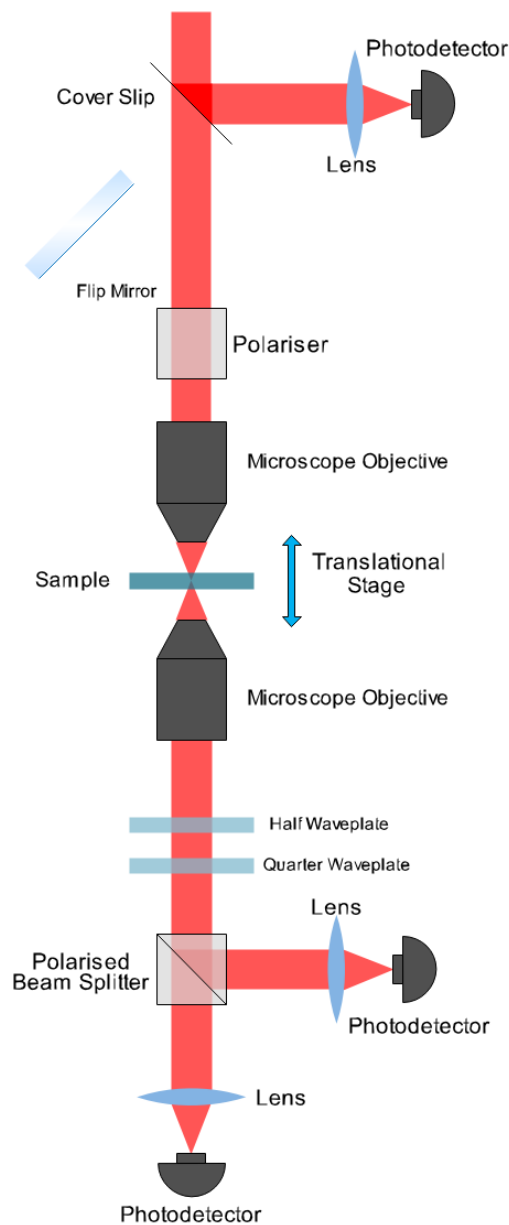


Figure 4.11: Schematic of the last apparatus which splits the beam in accordance with its polarisation and captures both beams using separate photodetectors. A third detector is placed at the beginning before the interaction to measure any power fluctuations in the beam.

circular. Thus, the polariser will transmit and reflect three base polarisations for a total of six intensities which are used to calculate the normalised stokes parameters. This completely characterises the beam's polarisation as shown in equations 4.1 to 4.3.

Figure 4.13 plots all the transmitted and reflected intensity measurements of a maltose solution of concentration 1g/10mL for the three polarisation bases. In the first plot, the vertical and horizontal intensities are fairly close to each other since diagonal polarisations contain equal amounts of horizontal and vertical. In the second plot, note that the y axes

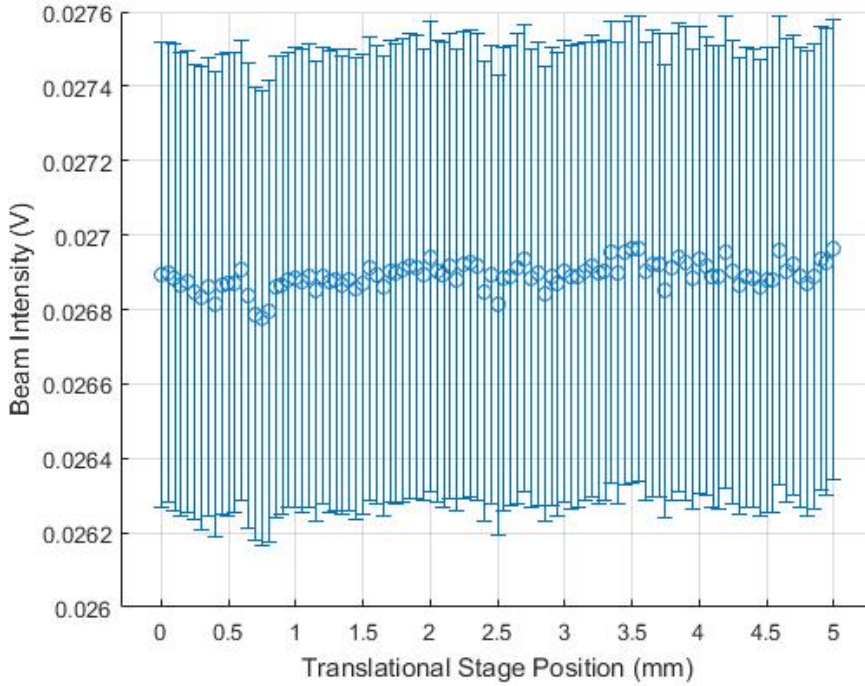


Figure 4.12: The intensity of the beam reflected off the cover slip before interaction.

on each side have different ranges. Almost all of the intensity is transmitted through the polarised beam splitter in the diagonal-antidiagonal basis since the beam is approximately diagonally polarised. In the last plot, the intensities are similar here too as the beam has almost no ellipticity and the transformed circular polarisations transmit and reflect equally. Figure 4.12 shows that the power of the beam remains stable throughout the translation of the cuvette with an error of only $\pm 1\%$. The intensity signals in these plots are on average flat which shows that the photodetectors' larger capture area solved the problem of having an increasing or decreasing profile as highlighted in the previous section. The peaks still remain so the six intensities are used to calculate the normalised intensities. These normalised intensities are no longer dependent on the position of the cuvette between the objectives as shown by the flat profile in figure 4.14. The normalised horizontal intensity I_{NH} is given by:

$$I_{NH} = \frac{I_H}{I_H + I_V} \quad (4.4)$$

and the normalised vertical intensity I_{NV} is given by:

$$I_{NV} = \frac{I_V}{I_H + I_V} \quad (4.5)$$

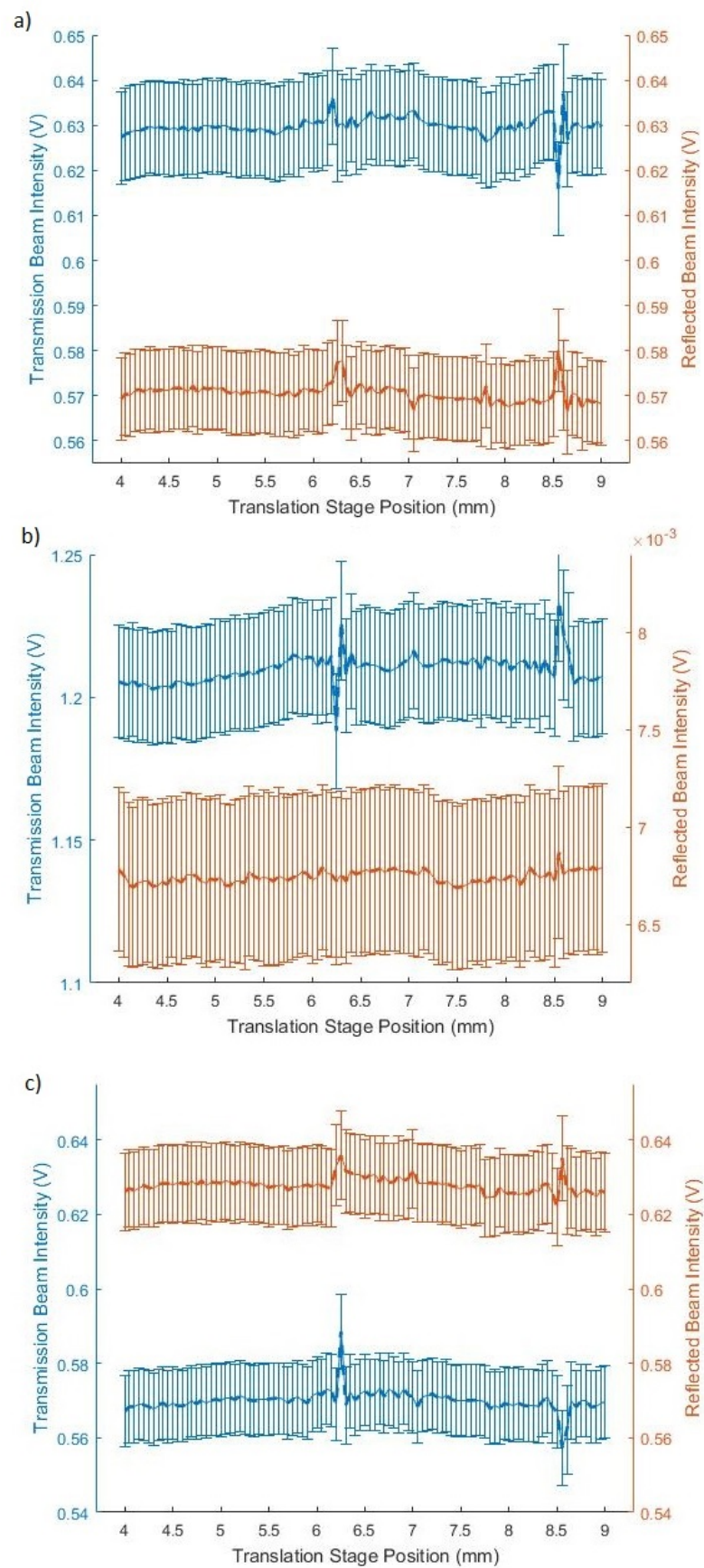


Figure 4.13: Caption next page.

Figure 4.13: The transmission and reflected beam intensities of maltose solution at concentration 1g/10mL in a 1mm thick cuvette in all three polarisation bases in the non-paraxial regime. a) The two intensities in the horizontal and vertical polarisation basis. b) The two intensities in the diagonal and antidiagonal polarisation basis. c) The two intensities in the left and right circular polarisation bases.

where I_H and I_V are the horizontal and vertical intensities respectively. By subtracting equation 4.5 from 4.4 in 4.1, it follows that the first normalised stokes parameter s_1 is given by:

$$s_1 = I_{NH} - I_{NV} \quad (4.6)$$

and similarly, s_2 is the difference between the normalised diagonal and antidiagonal intensities and s_3 is the difference between the normalised left and right circular intensities. The azimuthal angle is then calculated from s_1 and s_2 as shown in equation 2.33.

The stokes and azimuthal angle calculations for the maltose solution of concentration 1g/10ml are also done for a cuvette of thickness 2mm and the same measurements were done for a solution of sucrose in water of concentration 1g/10ml. These results are shown in figure 4.15. Unfortunately, the results are not satisfactory. As shown in the paraxial regime in figures 3.4 and 3.5, the azimuthal angle decreases as a result of the beams interaction with sucrose and maltose and only further decreases when the beam path length is increased. However, the opposite was shown and the change in azimuthal angle increased when the cuvette thickness increased. In the case of sucrose at 1g/10mL in a 2mm cuvette, the azimuthal angle actually increased. Although the error bars still allow for a decreasing linear trend in the relation between optical activity and beam path length, the average values are not satisfactory. On numerous measurements, the measured intensities would change when repeating the same measurements. For example, when measuring the transmission and reflected intensities of a 2mm cuvette containing water, when going back to repeat the measurements in the horizontal-vertical basis, the intensity voltages had changed as shown in figure 4.16. The detector placed before the interaction had shown that the power of the beam remained constant throughout.

The last measurements of optical activity follow on from the justification for the eliminating the peak artifacts. It was shown that the difference between the transmitted and reflected intensities was the same regardless of the cuvette's position in the optical path. As expected the optical rotation was independent of the intensity and therefore independent of the position of the cuvette. A cuvette translational stage position was arbitrarily chosen:

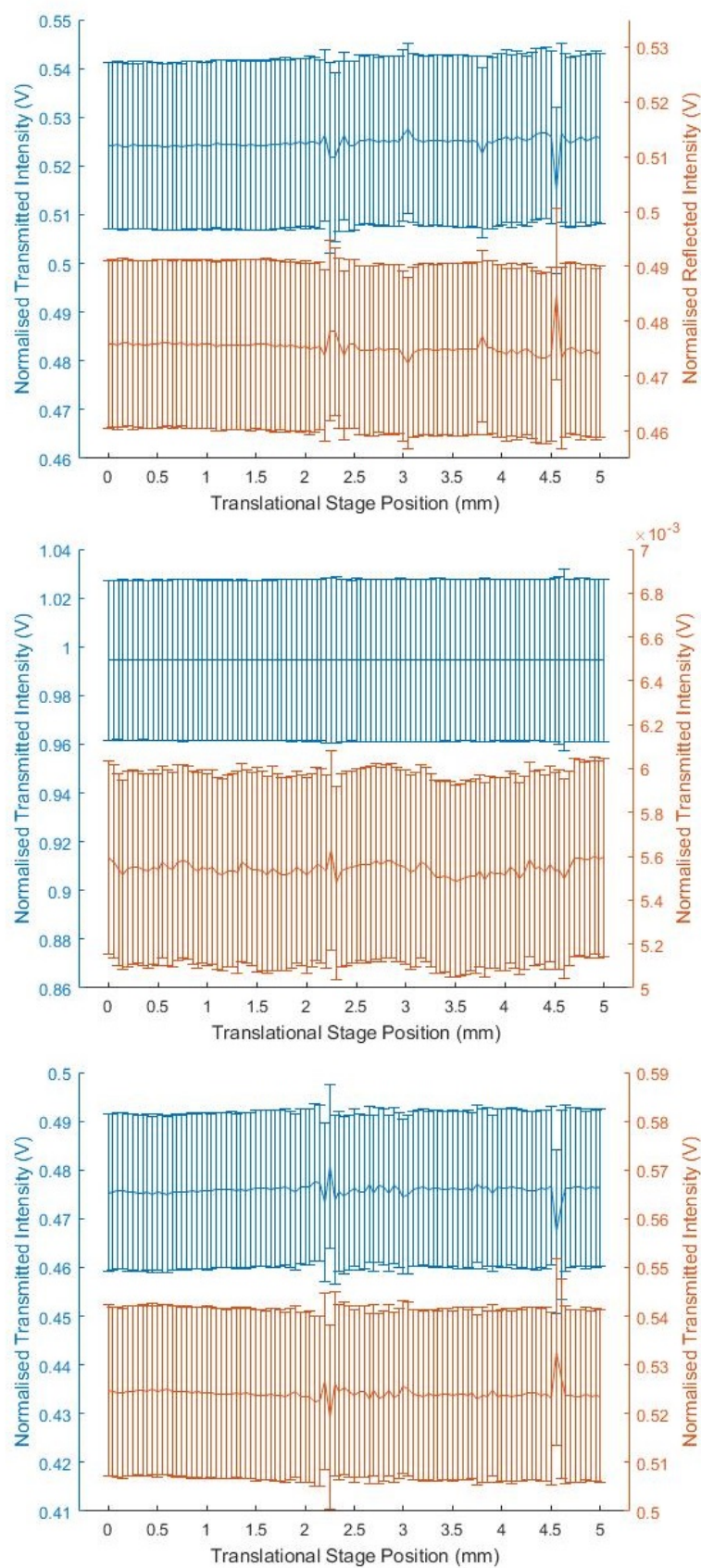


Figure 4.14: Caption next page.

Figure 4.14: The normalised transmission and reflected beam intensities of maltose solution at concentration 1g/10mL in a 1mm thick cuvette in all three polarisation bases in the non-paraxial regime. a) The two normalised intensities in the horizontal and vertical polarisation basis. b) The two normalised intensities in the diagonal and antidiagonal polarisation basis. c) The two normalised intensities in the left and right circular polarisation bases.

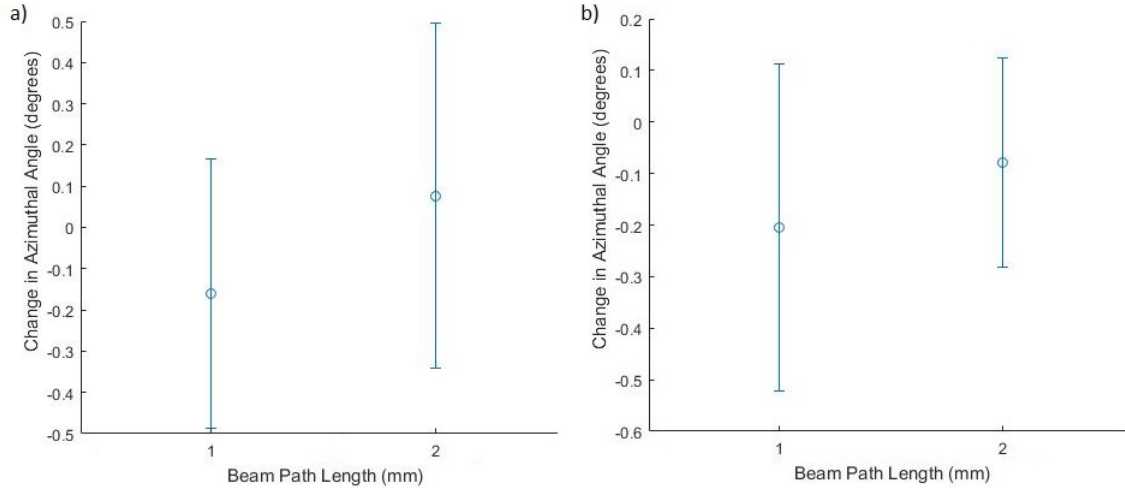


Figure 4.15: a) The difference between the azimuthal angles of a sucrose solution of concentration 1g/10mL and water in both the 1mm and 2mm cuvettes in the non-paraxial regime. b) Similarly the difference between the azimuthal angles of a maltose solution at concentration 1g/10mL and water in both the 1mm and 2mm cuvettes in the non-paraxial.

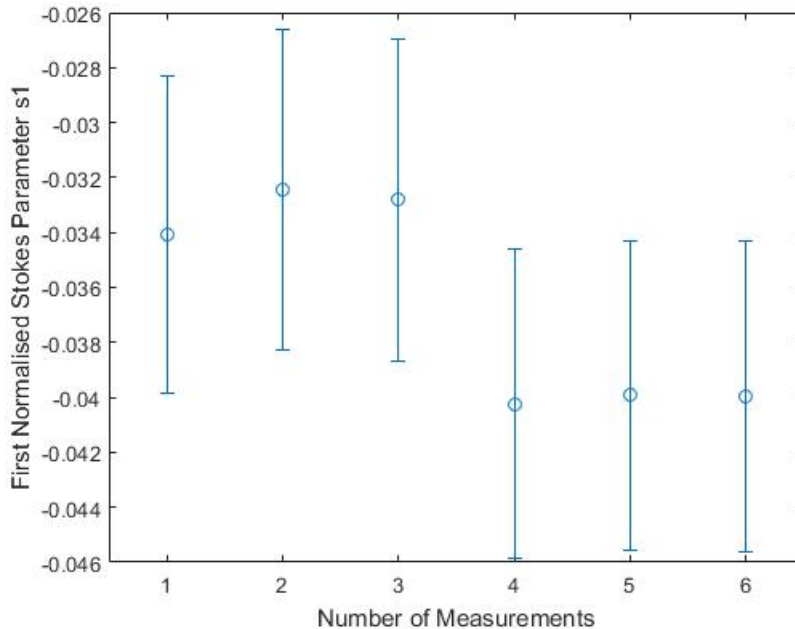


Figure 4.16: The first stokes parameter s_1 of water in a 2mm cuvette showing the equipment inconsistencies. The first three points indicate the first measurements and the next three points indicate the exact same measurement at a later time.

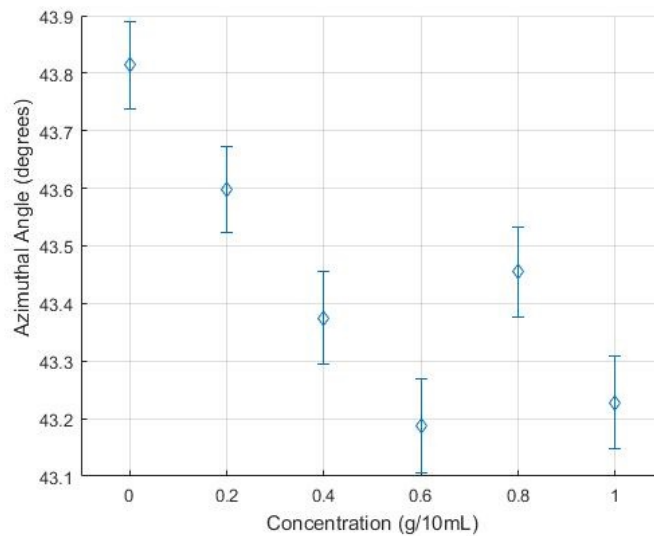


Figure 4.17: Azimuthal angle as a function of concentration of sucrose solutions in the non-paraxial regime.

3mm back along the optical beam path from the objective lens. The cuvette was placed in the translational staged and moved as close as possible to the objective lens without touching it. The translational stage value is rounded down to the nearest 0.1mm and moved to that position and then translational stage is moved 3mm back. The succeeding optical rotation measurements had taken place at this position.

In figures 4.17 and 4.18 the optical activity measurements of various concentrations of sucrose and maltose solutions in the non-paraxial regime are shown. The change in azimuthal angle of sucrose shows a linear relation with the concentration until it reaches 0.8g/10mL where it increases and then decreases again when reaching the highest concentration of 1g/10mL. On the other hand, the optical activity of maltose shows unexpected results that appear to rotate the polarisation of the beam in the opposite direction when compared to the results in the previous two setups and in the paraxial regime. One explanation is that the concentrations of the sugars were not accurate and the methods of their preparation could have been more accurate. However, this is unlikely to go so far as to cause the maltose solutions to rotate the polarisation of light in the opposite direction. As above, these are likely due to equipment inconsistencies over multiple measurements and the sample preparation. It still remains that there is a possibility that there is a difference between optical activity in the paraxial and non-paraxial regime.

The inconstancy of the results in figures 4.15, 4.17 and 4.18 of the non-paraxial beam path prevents any meaningful comparison with the results in figures 3.2, 3.3, 3.4 and 3.5 in

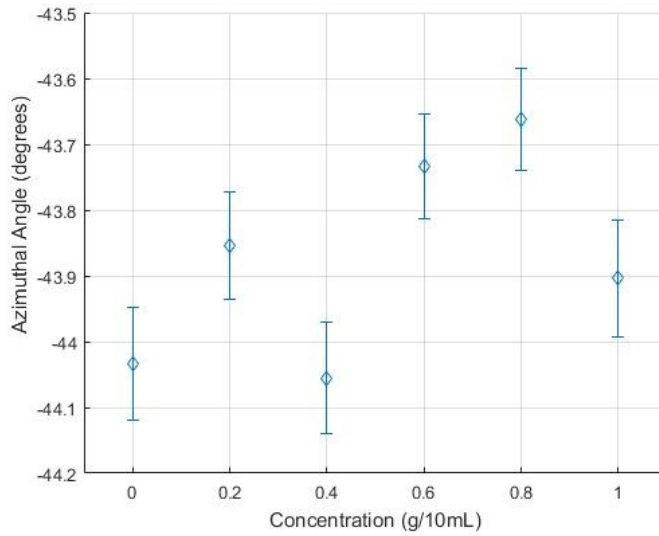


Figure 4.18: Azimuthal angle as a function of concentration of maltose solutions in the non-paraxial regime.

the collimated beam path. Therefore, further measurements need to be taken or new detection methods need to be considered.

4.4 Summary

In summary, the first setup involving the polarimeter showed unexpected peaks in the azimuthal angle. Thus, further detection methods had to be considered. The second setup using the small area detectors, waveplates and polarised beam splitter allowed the direct measurement of the transmitted and reflected intensities that are required to calculate the stokes parameters. It revealed that the peaks are not manifestations of optical activity since both the transmitted and reflected beams would either peak or dip in the same direction. However, the small capture size of the small area detectors showed that small misalignments in the beams due to the translation of the cuvette affected the intensity measurements. The final setup sought to remedy this issue by using photodetectors with larger capture areas. The peaks were eliminated by normalising the intensity measurements by the sum of the transmitted and reflected intensities. This normalisation method also eliminated the peaks when applied to the measurements in section 4.2. The final optical activity measurements in the non-paraxial regime were unexpected. So, more accurate results need to be taken by repeated measurements.

5

Conclusion

The discovery of optical activity has advanced the frontiers in many scientific fields such as organic chemistry, spectroscopy and molecular biology due to its link to chirality and molecular structure. More recently, optical activity has gained an interest in the field of optics once again due to its relations with more recent concepts concerning the nature of light such as interactions of the angular momentum of light, electromagnetic duality [1] and helicity of light [3].

In this thesis, I have focused on building an optical apparatus that measures and compares the optical activity of both a collimated and a non-paraxial focused beam. Various instruments and methods for detection have been explored. In the paraxial measurements, a commercial polarimeter was used to characterise the beam's polarisation with the stokes parameters. In the non-paraxial measurements, in addition to the polarimeter, a polarised beam splitter was used to measure the beam intensities required to characterise the beam's polarisation with the stokes parameters as well. I investigate the optical activity of the common sugars: sucrose and maltose that induce well known optical activities that have a linear dependence

on both the concentration and beam path length. Various concentrations of these sugars were measured and various cuvette thicknesses were measured to explore the beam path length relation. The samples were translated through the focal point of the focused beam to measure optical activity at different points in the beam. An optical artifact manifested in the form of peaks or dips in the measured beam intensities over different cuvette positions. This artifact was solved when applying various instruments and methods.

Based off preliminary results in the non-paraxial beam when using the polarimeter and small area photodetectors, there appears to be no difference in the beams interaction with the chiral solutions of sugars to its interaction when collimated. The beam splitter and multiple photodetectors show inconsistencies when compared to the paraxial measurements. However, it is highly likely this is due to equipment inconsistencies over multiple measurements. This can be avoided or at least controlled by taking more measurements and aligning the system more thoroughly and more often in such a way to minimise measurement inconsistencies.

The experimental nature of this thesis puts it as a sort of starting point to conduct more optical activity measurements in the non-paraxial regime. It measures optical activity in a specific kind of non-paraxial beam. The paraxial approximation restricts a beam to a very specific kind of propagation where the beam makes a small angle with the optical axis and any beam outside of this would be considered non-paraxial. This thesis shows the similarities between optical activity in a collimated beam and one kind of non-paraxial beam and it can't definitively make assumptions about the behaviours of other non-paraxial beams. It is limited in this sense.

5.1 Future Research

This does, however, open up the possibilities of studying optical activity in the non-paraxial regime in a broader scope. One simple extension is to use microscope objectives with higher numerical apertures and magnification. This study only considers the optical activity due to molecular chirality but as system without any chiral molecules can still display other forms of optical activity such as magnetic optical activity and non molecular manifestations of chirality.

This thesis does have implications when it comes to optical trapping. An optical trap uses highly focused and therefore, non-paraxial light to suspend particles. Optical traps

are very effective at isolating even the smallest systems. This implies that it is possible to investigate optical activity of optically trapped particles by measuring their interaction with a focused light source such as the laser being used to trap it. With sensitive enough equipment, optical activity would become another way of looking at a single particle.

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