Chapter 1

Light scattering

It is no exaggeration to state that everything we see is a result of scattering of light, and we see nothing that is devoid of scattering. Natural wonders ranging from the frequently seen sight of bright white clouds on the bright blue sky, to the blue moon seen once in a blue moon, are all examples of effects of scattering of light. Indeed, the phenomenon of scattering, accompanied by absorption, is responsible for the colour in life.

As a science, the study of light scattering and its applications is too vast to be put into a single treatise. It is no surprise, therefore, that various branches in pure and applied physics have materialised in modern physics. Each one deals with the effects and applications of light scattering in its own, independent premises. A few of these branches are astrophysics, ocean physics, atmospheric physics, radar physics, biomedical optics etc. Even more astonishing is the fact that a few phenomena observed in solid state physics, related to electron propagation in disordered crystals, are seen to have optical counterparts. Over the years, the science of light scattering has grown through exchange of ideas, observations and theories between these branches.

This thesis deals with some aspects of light scattering related to biomedical optics, and some related to laser optics.

1.1 Single scattering

Although the bulk of this thesis deals with light scattered from a collection of inhomogeneities, the effects of this collection are calculated from the knowledge of the effects of a single inhomogeneity. We familiarise ourselves with the fundamentals of the single scattering theory before graduating to the effects of multiple scattering. Detailed treatments on the phenomenon of scattering of light from a single particle are given in the literature[1, 2, 3].

Consider a wave incident upon a particle of an arbitrary shape. The total energy scattered by the particle in all directions can be considered to be the energy of the incident wave falling on a virtual area C_{sca}. This area is called the scattering cross-section of the particle. Likewise, the total energy absorbed by the particle can be put equal to the energy incident on an area C_{abs}, defining the absorption cross-section. The sum total of the two gives the extinction cross-section $C_{ext} = C_{sca} + C_{abs}$. For non-absorbing particles, $C_{ext} = C_{sca}$. These cross-sections depend upon the orientation of the particle and the polarisation state of the incident light. The geometric cross-section G of the particle is the projection of cross-sectional area of the particle on a plane perpendicular to the direction of the incident light. For a spherical particle of radius a, $G = \pi a^2$.

The essential problem in single scattering theory is to find out the scattered field as a function of direction, for a given incident field and a scatterer of arbitrary size, shape and orientation. (The size of a particle is always described relative to the wavelength of light inside the particle, unless otherwise specified. The orientation is specified with respect to an imaginary XYZ frame of reference, that is also used to define the direction of incidence and scattering.) Arbitrariness in shape is a characteristic that analytical theories cannot handle, so all the analytical treatises dealing with single scattering work with symmetric particles, *viz.* spherical, ellipsoidal, cylindrical etc. Since the experiments and simulations described in this thesis use only spherical particles, we get acquainted with some of the important characteristics of single scattering by a spherical particle.

The Mie theory[1], in rigour, computes the scattering properties of spherical particles of arbitrary size. The Mie theory expresses the fields inside and outside the spherical scatterer in terms of known functions, *viz*, the Riccati Bessel functions. In particular, the theory calculates the angular dependence of the scattered intensity for light in the two extreme cases of polarisation, parallel and perpendicular to the scattering plane, and from this the intensity for any polarisation can be calculated.

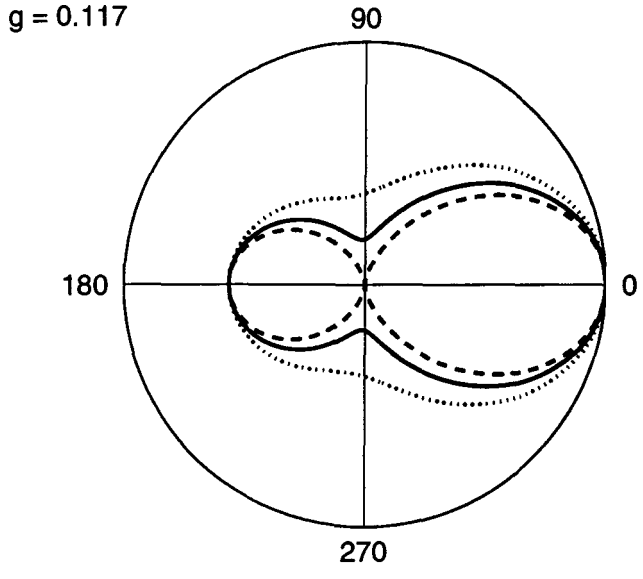

Figure 1.1: *A polar plot of the intensity distribution of light scattered from a particle of* $g = 0.117$. *The bold solid line is the intensity distribution for unpolarised light, while the dotted and dashed lines are for perpendicular and parallel polarisations respectively. The two polarisation states and their relevance in scattering problems will be discussed in a later section.*

The parameter that describes the size of the particle in comparison with the wavelength of the incident light is the size parameter x, given by $x = \mathrm{ka}$, where k is the wavevector and a is the diameter of the particle.

For particles smaller than the wavelength, the scattered intensity is distributed more or less uniformly in all directions, in which case the scattering is called isotropic. On the other hand, for particles larger than the wavelength, light is scattered preferentially in the

Diameter (μ)	Material	n	g
0.12	Polystyrene	1.59	0.117
0.195	PMMA	1.495	0.302
0.23	Polystyrene	1.59	0.45
0.3	Polystyrene	1.59	0.7

Table 1.1: *Refractive indices and anisotropy parameters of the particles used in the experiments described in this thesis. The parameters are calculated for a wavelength of* 0.612μ*, and assuming the surrounding medium to be water,* $n = 1.33$.

forward direction, and the scattering is called anisotropic. This anisotropy can be seen to be arising from the interference of wavelets scattered by small sections of the particle. For particles smaller than the wavelength, all the wavelets will be in phase in all directions, while for larger particles, the wavelets in the forward direction are in phase.

The figure 1.1 shows a typical polar plot of the scattered intensity with respect to the scattering angle (which is always defined relative to the angle of incidence). This scattering pattern has been calculated by the Mie theory.

If θ is the average scattering angle for a given particle, then $g = <\cos\theta>$, the average cosine of the scattering angle, is known as the anisotropy parameter which quantifies the anisotropy in scattering. g depends upon the size of the scatterer, the wavelength of the light and the refractive index contrast between the scatterer and the surrounding medium. It can be calculated from the Mie theory. Clearly, when $g = 0$, the scattering is isotropic and when $g = 1$, the scattering is totally forward. Table 1.1 gives the values of g for particles of different radii and refractive indices.

1.2 Multiple scattering

The introductory concepts of the theory of multiple scattering in a random medium are presented here. These concepts suffice for understanding the more advanced topics described in the later parts of this thesis. For detailed considerations of the theory of multiple

scattering, we refer the reader to the literature[4, 5, 6, 7].

In a medium consisting of a large number of scatterers, the incident field undergoes recurrent random scattering before it exits the medium. One cannot calculate exactly the form of the field scattered from an arbitrary multiply scattering medium. The total scattered intensity, as a function of either the distance of travel within the scattering medium or the angle of scattering, hinges crucially upon the interference effects that are manifest within the scattering medium. Accordingly, based on certain approximations concerning the length-scales of the medium, a few theoretical models have been proposed to explain the experimental observations regarding multiply scattering media. We get acquainted with these length-scales, and the implications of their magnitudes.

An important parameter in the theory of multiple scattering is the scattering mean free path, l,. It is defined as the average distance between two successive scattering events, and is given by

$$l_s = \frac{1}{n\sigma_s} \tag{1.1}$$

Here, n is the number of scatterers per unit volume and σ_s is the scattering cross-section of the individual scatterer. After travelling, on the average, a distance of l,, the wave can be assumed to have undergone scattering, but it hardly means the direction is randomised. As mentioned earlier, due to the anisotropy of scattering, the wave may propagate in the near forward direction even after several scattering events. To accomodate this characteristic of scattering, a transport mean free path is introduced, that is defined as the average distance that the light travels before its direction of propagation is randomised. The transport mean free path, denoted by l^* is given by

$$l^* = \frac{l_s}{1-g} \tag{1.2}$$

Here g is the anisotropy parameter defined in the earlier section. Clearly, for isotropic scattering, where $g \to 0$, the transport mean free path is approximately equal to the

scattering mean free path, which is understandable because the wave can be scattered into any angle with almost uniform probability over 4π steradian. The transport mean free path is the more relevant length scale in light transport through disordered media because scattering is, in general, anisotropic.

Generally, the process of scattering is not completely free of absorption, due to a finite absorption cross-section of the scatterer at the wavelength of interest. Neglecting absorption completely may lead to discrepancies between experimental observations and theoretical calculations. The length scales relevant to absorption are the inelastic mean free path l_i and the absorption mean free path l_{abs}. 'The inelastic mean free path is the distance over which the intensity of the wave reduces to $\frac{1}{e}$ times the initial intensity due to absorption. Within successive scattering events, the wave can travel along a zig-zag path of length l_i, without much displacement between the beginning and the ending points. The average distance between the begin and end points for paths of length l_i is defined as the absorption mean free path. It is given by

$$l_{abs} = \sqrt{\frac{1}{3} l_s l_i} \tag{1.3}$$

In most treatments, the random medium is taken to be in the form of a semi-infinite slab - of size L along the direction of incidence of light and infinite transverse to it. Such a medium is said to have an optical thickness $\tau = \frac{L}{l^*}$.

It should be noted that the average distance between two scatterers, which we call r, can be much smaller than l_s, since l_s is the distance between two scattering centres. The physical extent of the scatterers can be large to make these two distances different from each other.

1.3 Various regimes of multiple scattering

Depending upon these length scales, the degree of disorder or the strength of scattering of a given medium can be decided[7]. When the wavelength λ is much larger than the

distance between two scatterers r, the scatterers are not resolved by the wave and the wave sees an effective medium, with a certain effective refractive index. When the wavelength is small so that the discrete nature of the scattering centres can be resolved by the wave, one needs to take into account the effective transport mean free path l^* to decide the regime of scattering. When $l^* >> \lambda$, light transport is assumed to be diffusive. Interference effects can be neglected. When $l^* \sim \lambda$, the medium is said to be strongly scattering. Before the wave can complete one cycle of oscillation, it gets scattered. Here interference effects dominate light transport so much so that the effective transport can be brought to a complete halt. This can be regarded as a situation where the wave keeps getting scattered within the strongly scattering medium and is unable to exit the medium[8]. Thus, the light is localised inside the scattering medium, similar to the localisation of electrons inside a disordered conductor[9].

When $l^* >> \lambda$, one can assume that the light propagates diffusively within the medium and the diffusion equation can be used to study the light transport analytically, provided the medium satisfies certain conditions regarding its linear dimensions, extent of absorption and a few others. The diffusion equation, albeit its downright simplicity, has been used to explain a variety of observed phenomena like pulse propagation[10], coherent backscattering[11, 12, 13] and frequency correlations[14, 15, 16]. The equation reads, for the intensity **I** at a position r inside the medium, at time t,

$$\frac{\partial I(\mathbf{r},t)}{\partial t} = D\nabla^2 I(\mathbf{r},t) - \frac{v}{l_i} I(\mathbf{r},t) + S(\mathbf{r},t) \tag{1.4}$$

Here D is the diffusion coefficient, given by $\frac{1}{3}vl^*$, v is the speed of propagation of the light in the medium. The second term describes absorption, where l_i is the absorption length and v is the transport velocity of light in the medium, and $S(\mathbf{r}, t)$ is the source term. This diffuse regime is properly described by the condition $\lambda << l^* << L << L_{abs}$, where L_{abs} is the absorption length. The first inequality ensures that localization effects are absent or are small, the second inequality ensures multiple scattering and the third inequality ensures that absorption is negligible. Consider the propagation of light through

the slab of thickness L along +Z direction. The incoming beam decays exponentially due to multiple scattering, which scatters out the intensity from the beam. This decay is the Beer-Lambert decay given by

$$I(z) = I_0 e^{-\frac{z}{l_s}} \tag{1.5}$$

where I_0 is the incident intensity of the beam.

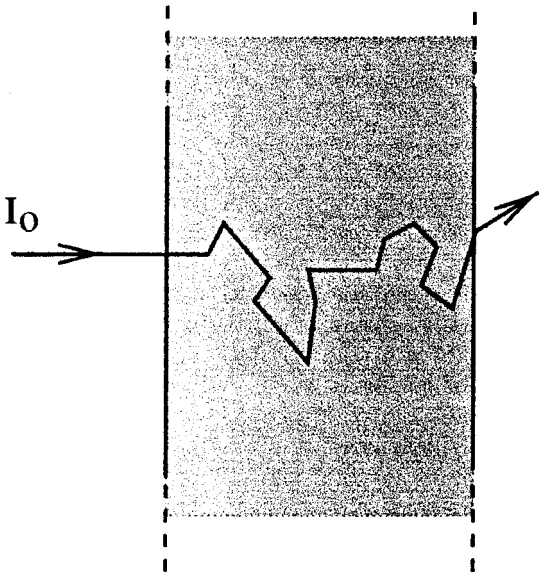

Figure 1.2: *The semi-infinite slab geometry showing the diffusion of a photon contained in a beam of intensity* **I,.** *The intensity transmitted is given by the equation 1.7*

According to diffusion theory, the incident beam is converted into diffuse light in a skin layer whose characteristic thickness is about one transport **mean free** path. Effectively, the diffusion approximation assumes that diffuse intensity enters the slab in a trapping plane located at a distance $z_0 \sim l^*$ outside the scattering medium. The exact value for the location of the trapping plane in the case of point scatterers is 0.7104 *l,.* Once inside the scattering medium, the intensity obeys equation 1.4. In the steady state, the time derivative vanishes. For the slab geometry shown in the figure 1.2, the equation to be solved is $I''(z) = 0$, along with the boundary conditions, $I(-z_0) = I_0$, and I(L + z_0) = 0,

where the prime refers to the derivative with respect to z. The solution reads,

$$I(z) = I_0 \frac{L + z_0 - z}{L + 2z_0} \tag{1.6}$$

Thus the transmitted intensity (T) is given by putting $z = \mathrm{L}$ in this expression, yielding,

$$T = \frac{I(z = L)}{I_0} = \frac{z_0}{L + 2z_0} \tag{1.7}$$

which indicates a $\frac{1}{L}$ behaviour. Thus, in the diffusion approximation for a slab geometry when the scattering medium is nonabsorbing, an Ohmic behaviour is observed for the transmitted light. For a bulk system, the solution of the diffusion approximation with the initial conditions $I(\mathbf{r}, 0) = \delta(\mathbf{r})$ and $S = 0$ reads

$$I(\mathbf{r}, t) = (4\pi D t)^{-\frac{3}{2}} e^{-(\frac{r^2}{4Dt}) - D\kappa^2 t} \tag{1.8}$$

The diffusion approximation works well in predicting the temporal broadening of pulses transmitted through random media[10]. In fact, the temporal pulse distribution predicted by the diffusion approximation can be used to extract the values of the diffusion coefficient, where the Mie theory cannot be applied[17]. Apart from pulse distribution, the diffusion theory has been employed in explaining coherent backscattering effect[11, 12, 13], continuous wave transmission and frequency, spatial and temporal correlations[14, 15, 16].

The biggest drawback of this approximation is that one needs a proper model for the source of light. In practical situations, a laser is used as a source, while the diffusion approximation starts off with a non-collimated source[18]. The position of this non-collimated source inside the entry face of the sample is also debatable. For a long time, the approximation was used with a diffuse source assumed to exist at a distance of one l^* inside the entry face. Recent literature questions this part of the diffusion approximation, and the question as to where exactly does collimated light become diffuse inside the random medium has become an activity by itself[18, 19, 20, 21]. Another important discrepancy with the diffusion approximation is its limited applicability in the case of strongly absorbing random media[22].

The diffusion theory, and other analytical theories for wave transport through random media, work on the assumption that light has a scalar nature. Analytical theories do not incorporate polarisation characteristics of light, and therefore, the phenomenon of depolarisation has not been studied analytically. Since depolarisation of light upon multiple scattering is the crux of the matter in this part of the thesis, we resort to numerical simulations. In our numerical studies, we employ Monte-Carlo simulations that simulate random photon paths on a disordered lattice. Monte-Carlo simulations have been used in the past to study transport of unpolarised and polarised light through scattering media[23, 24]. The techniques developed for simulating random walks in neutron scattering have been adapted here[25]. Numerical simulations provide a free handle on parameters like the scattering mean free path, the optical depth etc, and so different regimes of scattering can be investigated without making specific assumptions. This is not possible for analytical theories, that are based on fundamental assumptions about the degree of disorder. Though numerical simulations are computationally expensive, they are relatively easy to code, and provide transparent results.

Bibliography

[1] G. Mie, *Ann.Physik*, 25, 337 (1908).

[2] H. C. van de Hulst, Light Scattering by Small Particles, (Dover, New York, 1981).

[3] G. F. Bohren and D. R. Huffman, *Absorption* and Scattering of light by small particles, (Wiley, New York, 1983).

[4] P. Sheng, Introduction to Wave Scattering, Localization and Mesoscopic Phenomena, (Academic Press, San Diego, 1995).

[5] Scattering and Localization of Classical waves in random media in World Scientific Series on Directions in Condensed Matter Physics 8, Ed. Ping Sheng (World Scientific, Singapore, 1990).

[6] A. Ishimaru, Wave propagation and scattering in random media Vol I & II, (Academic, New York, 1978).

[7] M. C. W. van Rossum and Th. M. Nieuwenhuizen, Rev. Mod. Phys. 71, 313 (1999).

[8] D. S. Wiersma, P. Bartolini, A. Lagendijk and R. Righini, Nature, 390, 671 (1997).

[9] P. W. Anderson, Phys. Rev. 109, 1492 (1958).

[10] G. H. Watson Jr, P. A. Fleury and S. L. McCall, Phys. Rev. Lett. 58, 945 (1987).

[11] E. Akkermans, P. E. Wolf and R. Maynard, Phys. Rev. Lett. 56, 1471 (1986)

[12] M. J. Stephen and G. Cwilich, Phys. Rev. B, 34, 7564 (1986).

[13] M. B. van der Mark, M. P. van Albada and Ad Lagendijk, Phys. Rev. B 37, **3575 (1988).**

[14] S. Feng, C. Kane, P. A. Lee and A. D. Stone, Phys. Rev. Lett. 61, **834 (1988).**

[15] J. **H.** Page, H. P. Schreimer, A. E. Bailey and D. **A.** Weitz, Phys. Rev. E 52, **3106 (1995).**

[16] M. **J.** Stephen, Phys. Rev. B 37, **1 (1988).**

[17] M. S. Patterson, B. Chance and B. C. Wilson, App. Opt. 28, **2331 (1998).**

[18] Thorsten Spott and Lars O. Svaasand, Appl.Opt. 39, **6453 (2000)**

[19] A. A. Chabanov and A. Z. Genack, Phys. Rev. E, , 56, **R1338 (1997).**

[20] K. **K. Bizheva**, A. M. Siegel and D. A. Boas, Phys. Rev. E, 58, **7664, (1998).**

[21] Z. Q. Zhang, I. P. Jones, H. P. Schreimer, J. H. Page, D. A. Weitz, and Ping Sheng , Phys. Rev. E, 60, **4843 (1999).**

[22] D. **J.** Durian and J. Rudnick, J. Opt. Soc. Am., 14, **235 (1997).**

[23] P. Bruscaglioni, G. Zaccanti and Q. Wei, App. Opt. 32, **6142 (1993).**

[24] A. Sassaroli, C. Blumetti, F. Martelli, L. Alianelli, D. Contini, A. Ismaelli and G. Zaccanti, App. Opt. 37, **7392 (1998).**

[25] B. Davison and J. B. Sykes, Neutron *Transport* Theory, (Oxford University Press, **1958).**

Chapter 2

Polarisation in light scattering

2.1 Introduction

Since a considerable part of this thesis deals with the polarisation of scattered light and its preservation or destruction inside a random medium, we begin by introducing the most basic notions of polarisation of light, its representation and its measurement. Extensive texts are available that deal with the fundamentals of polarisation, its measurement, its representation using the Stokes vectors, and its relevance in light scattering[1, 2, 3, 4, 5].

Consider a plane monochromatic wave with angular frequency w, wave vector magnitude k propagating along the Z axis. The electric field at any instant of time can be written as

$$\mathrm{E} = \mathbf{A}\cos(kz - wt)\ \mathrm{B}\sin(kz - wt) \tag{2.1}$$

A and B are vectors independent of position. At a particular plane, say at $z = 0$, the tip of of the electric field vector traces a curve given by

$$\mathbf{E}(z = 0) = \mathbf{A}\cos(\omega t) + \mathbf{B}\sin(\omega t) \tag{2.2}$$

which is the equation of an ellipse. This is the most general state of polarisation of light called the elliptical polarisation. When one of the two constants **A** or B is zero, the electric field vibrates along a straight line, and the state of polarisation is called linear

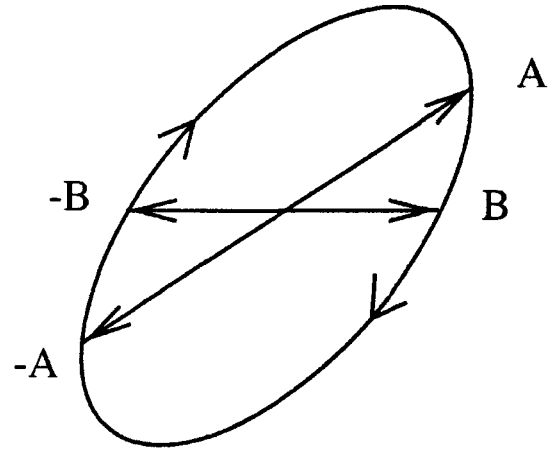

Figure 2.1: *Ellipse of vibration given by equation* ***2.2***

polarisation. When $|\mathbf{A}| = |\mathbf{B}|$, the electric field rotates along a circular path, and the light is said to be circularly polarised. The sense of vibration of the electric field vector along the ellipse depends upon the direction from which it is seen. In this thesis, we have used the convention according to which an elliptically polarised wave is right-handed if the electric field vector is rotating in the clockwise sense as viewed by an observer looking towards the source.

Apart from the handedness, the ellipse can be characterised by its ellipticity (the ratio of the length of the semiminor axis to the semimajor axis) and its azimuth (the angle made by the semimajor axis to an arbitrary direction of reference). (Refer to figure **2.2.)** The four parameters, *viz* handedness, ellipticity, azimuth and the intensity are called the four ellipsometric parameters of the polarised light.

These four parameters completely specify the state of polarisation of a wave. However, they are difficult to measure directly, and also not helpful in understanding the transformations of polarisation. Not all the four parameters are additive, which renders it difficult to discuss partially polarised light using this representation.

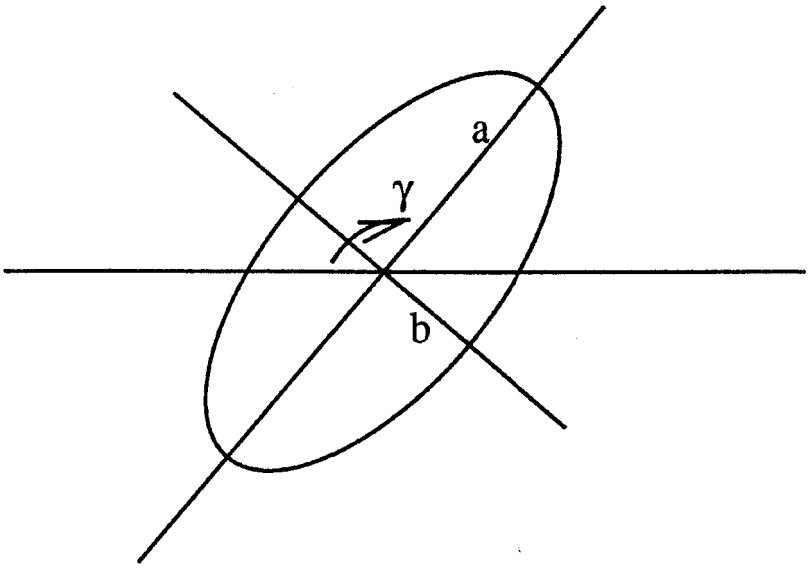

Figure **2.2:** *Ellipsometric parameters illustrated.*

2.2 Stokes vectors representation

An equivalent representation of polarised light is the Stokes vector **representation**[6], that uses a vector consisting of four Stokes parameters. Each of these parameters is easily measurable experimentally, and they are all additive in the sense that the parameters for the sum of two beams of light are given merely by adding the individual Stokes parameters. In essence, the Stokes parameters are a powerful representation of polarised light. Accompanied by the Mueller matrix method, that we shall introduce in the next section, the Stokes vector representation provides an elegant means to study the evolution of the polarisation state of light upon interaction with various optical elements, and most important, upon multiple light scattering in random media.

We introduce the four Stokes parameters along with their notations through the experiments by which they can be determined.

Assume that a detector measures the intensity of an arbitrary monochromatic beam of light regardless of its polarisation. Assume also that the various polarisers are ideal; they

do not absorb light. To define the polarisation states of the beam, we use a set of orthogonal axes $\hat{e}_{||}$ and $\hat{e}_{\perp}$, which we shall refer to as "horizontal" and "vertical" respectively.

Then, the electric field of the beam is

$$\mathbf{E} = \mathbf{E_0}\, exp(ikz - iwt) \tag{2.3}$$

$$\mathbf{E_0} = E_{||}\hat{e}_{||} + E_{||}\hat{e}_{\perp} \tag{2.4}$$

Now we make the following measurements on the beam.

1. Measure the intensity falling on the detector with no polarisers in the path. The measured value is

$$E_{||}E_{||}^{*} + E_{\perp}E_{\perp}^{*}$$

(We have omitted the factor $\frac{k}{2\omega\mu_0}$.)

2. Let a horizontal polariser be introduced in the path of the beam. The intensity transmitted will be $E_{||}E_{||}^{*}$. Let a vertical polariser be introduced in the path, replacing the horizontal polariser. The intensity measured now will be $E_{\perp}E_{\perp}^{*}$. The difference between the two measured intensities is

$$I_{||} - I_{\perp} = E_{||}E_{||}^{*} - E_{\perp}E_{\perp}^{*}$$

3. Let a polariser be placed in the path of the beam, with its axis aligned at $+45°$ to the horizontal. We introduce a new set of basis vectors $\hat{e}_{+}$ and $\hat{e}_{-}$ which are obtained by rotating our previous frame through $+45°$ and $-45°$ respectively. The new basis is then

$$\hat{e}_{+} = \frac{1}{\sqrt{2}}(\hat{e}_{||} + \hat{e}_{\perp}) \quad ; \quad \hat{e}_{-} = \frac{1}{\sqrt{2}}(\hat{e}_{||} - \hat{e}_{\perp}) \tag{2.5}$$

In this basis, the electric field vector $\mathbf{E_0}$ can be written as $\mathbf{E_0} = E_{+}\hat{e}_{+} + E_{-}\hat{e}_{-}$, where

$$E_{+} = \frac{1}{\sqrt{2}}(E_{||} + E_{\perp}) \quad ; \quad E_{-} = \frac{1}{\sqrt{2}}(E_{||} - E_{\perp}) \tag{2.6}$$

The amplitude of the transmitted wave through the polariser aligned at $+45°$ is $E_{+} = \frac{1}{\sqrt{2}}(E_{||} + E_{\perp})$, giving an intensity $\mathbf{I}+= (E_{||}E_{||}^{*} + E_{||}E_{\perp}^{*} + E_{\perp}E_{||}^{*} + E_{\perp}E_{\perp}^{*})/2$.

Similarly, the intensity transmitted through the polariser aligned at $-45°$ is $I = (E_{\|}E_{\|}^* - E_{\|}E_{\perp}^* - E_{\perp}E_{\|}^* + E_{\perp}E_{\perp}^*)/2$. The difference between the two intensities is

$$I_+ - I_- = E_{\|}E_{\perp}^* + E_{\perp}E_{\|}^*$$

4. Here, we need to introduce right and left circular polarisers. The respective basis vectors are given by

$$\hat{e}_R = \frac{1}{\sqrt{2}}(\hat{e}_{\|} + i\hat{e}_{\perp}) \quad ; \quad \hat{e}_L = \frac{1}{\sqrt{2}}(\hat{e}_{\|} - i\hat{e}_{\perp}) \tag{2.7}$$

In this basis, the electric field vector $\mathbf{E}_0$ can be written as $\mathbf{E_0} = E_R\hat{e}_R + E_L\hat{e}_L$, where

$$E_R = \frac{1}{\sqrt{2}}(E_{\|} - iE_{\perp}) \quad ; \quad E_L = \frac{1}{\sqrt{2}}(E_{\|} + iE_{\perp}) \tag{2.8}$$

The intensity transmitted through the right circular polariser is $I_R = (E_{\|}E_{\|}^* + iE_{\|}E_{\perp}^* - iE_{\perp}E_{\|}^* + E_{\perp}E_{\perp}^*)/2$. The intensity transmitted through the left circular polariser is $I_L = (E_{\|}E_{\|}^* - iE_{\|}E_{\perp}^* + iE_{\perp}E_{\|}^* + E_{\perp}E_{\perp}^*)/2$.

The difference between the two gives

$$I_R - I_L = i(E_{\|}E_{\perp}^* - E_{\perp}E_{\|}^*)$$

Through these four experiments, we have directly measured the four Stokes parameters, written as

$$I = E_{\|}E_{\|}^* + E_{\perp}E_{\perp}^* \tag{2.9}$$

$$Q = E_{\|}E_{\|}^* - E_{\perp}E_{\perp}^* \tag{2.10}$$

$$U = E_{\|}E_{\perp}^* + E_{\perp}E_{\|}^* \tag{2.11}$$

$$V = i(E_{\|}E_{\perp}^* - E_{\perp}E_{\|}^*) \tag{2.12}$$

The Stokes parameters are related to the ellipsometric parameters as follows :

$$I = c^2 \tag{2.13}$$

$$\mathrm{Q} = \mathrm{c}^2 \cos 27 \cos 27 \tag{2.14}$$

$$U = c^2 \cos 27 \sin 2\gamma \tag{2.15}$$

$$\mathrm{V} = c^2 \sin 2\eta \tag{2.16}$$

where,

$$c^2 = a^2 + b^2$$
$$|\tan \eta| = \frac{b}{a} \quad \left(-\frac{\pi}{4} \leq \eta \leq \frac{\pi}{4}\right)$$

Here, a and b are the semimajor and the semiminor axes of the ellipse, γ is the azimuth $(0 \leq \gamma \leq \mathrm{x})$, and $\frac{b}{a}$ is the ellipticity. The sign of V signifies the handedness of the ellipse; positive stands for right-handed, and negative stands for left handed. The ellipticity and azimuth can be directly calculated from the Stokes parameter as

$$\tan 2\gamma = \frac{U}{Q} \qquad \tan 2\eta = \frac{V}{\sqrt{Q^2 + U^2}}$$

We note that the parameters Q and U depend upon the choice of the horizontal and the vertical axes, and I and V do not. If the frame of reference on which these parameters are defined is rotated through an angle ψ, then the transformation from $(I$,Q,U,V) to (I_i, Q_i, U_i, V_i) in the new frame is given by

$$\begin{bmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & cos2\psi & sin2\psi & 0 \\ 0 & -sin2\psi & cos2\psi & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} \begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} \tag{2.17}$$

The following table shows the Stokes vectors for various types of polarisations.

Linearly polarised light

Angle with horizontal	0°	90"	+45°	-45"	γ
	$\begin{bmatrix} 1 \\ 1 \\ 0 \\ 0 \end{bmatrix}$	$\begin{bmatrix} 1 \\ -1 \\ 0 \\ 0 \end{bmatrix}$	$\begin{bmatrix} 1 \\ 0 \\ 1 \\ 0 \end{bmatrix}$	$\begin{bmatrix} 1 \\ 0 \\ -1 \\ 0 \end{bmatrix}$	$\begin{bmatrix} 1 \\ \cos 2\gamma \\ \sin 2\gamma \\ 0 \end{bmatrix}$

Circularly polarised light

Right	Left
$\begin{bmatrix} 1 \\ 0 \\ 0 \\ 1 \end{bmatrix}$	$\begin{bmatrix} 1 \\ 0 \\ 0 \\ -1 \end{bmatrix}$

In the case of an ideal, strictly monochromatic wave, the four parameters are not independent, and it can be seen that the relation $I^2 = \mathrm{Q}^2 + U^2 + \mathrm{V}^2$ holds. In more practical situations, we define the parameters for a quasi-monochromatic beam. This beam has a small finite spectral width, because of which the two orthogonal components of the electric field are functions of time, but vary slowly over time intervals of the order of the period $2\pi/\omega$. The correlation between these two components over long time intervals decides the polarisation state of light. If $E_{\|}$ and $E_{\perp}$ are completely uncorrelated, the light is said to be unpolarised. If they are completely correlated, the light is called polarised. If they are partially correlated, the light is partially polarised.

The Stokes parameters of a quasi-monochromatic beam are given by taking the time averaged quantities over an interval long compared with the period. Thus, the parameters are written as

$$I = < E_{\|}E_{\|}^{*} + E_{\perp}E_{\perp}^{*} > \tag{2.18}$$

$$Q = < E_{\|}E_{\|}^{*} - E_{\perp}E_{\perp}^{*} > \tag{2.19}$$

$$U = < E_{\|}E_{\perp}^{*} + E_{\perp}E_{\|}^{*} > \tag{2.20}$$

$$V = i < E_{\|}E_{\perp}^{*} - E_{\perp}E_{\|}^{*} > \tag{2.21}$$

In this case, we can see that $I^2 \geq Q^2 + U^2 + V^2$. The equality holds if the light is completely polarised. Accordingly, we can define the degree of polarisation of the light as $\frac{\sqrt{Q^2+U^2+V^2}}{I}$. The degree of linear polarisation can be defined as $\frac{\sqrt{Q^2+U^2}}{I}$, and the degree of circular polarisation as $\frac{V}{I}$. The sign of V signifies the preferential sense of rotation of the vibration ellipse of the beam: postive indicates a right handed ellipse. The quantity $\frac{U}{Q}$ specify the preferential azimuth and $\frac{V}{\sqrt{Q^2+U^2}}$ specifies the preferential ellipticity of the vibration ellipses.

Thus, being equipped with a powerful and mathematically simple means of representating and studying polarisation of light, we now proceed to learn a technique used to study the transformation of polarisation of a beam, upon interaction with optical elements like reflectors, polarisers, retarders etc, and most important for our context, scatterers.

2.3 Mueller matrix formalism

This section deals with a technique of measuring the change in polarisation of light upon reflection, refraction or scattering. The method is called Mueller matrix technique[7], which we shall now introduce. In the following discussion, we assume that the medium with which the light interacts is linear. This enables us to write a linear relation between the input and the output Stokes vectors of the beam, before and after the interactions. If $\mathbf{I}_i$ is the Stokes vector of the light before the interaction, and $\mathbf{I}_o$ is after the interaction,

$$\mathbf{I}_o = \mathbf{M}\mathbf{I}_i \tag{2.22}$$

$$\mathbf{M} = \begin{bmatrix} m_{00} & m_{01} & m_{02} & m_{03} \\ m_{10} & m_{11} & m_{12} & m_{13} \\ m_{20} & m_{21} & m_{22} & m_{23} \\ m_{30} & m_{31} & m_{32} & m_{33} \end{bmatrix} \tag{2.23}$$

M here is the 4x4 real-valued transformation matrix, whose elements m_{ij} depend upon the properties of the interacting element, frequency of light and in the case of light scattering, on the scattering angle. In general, all the sixteen elements of the Mueller matrix are independent. However, due to certain symmetry conditions that may apply to M, and due to certain optical properties of the interacting medium, the number of independent

matrix elements may be reduced. In the case of multiple optical elements arranged in a cascade, the total effect of the cascade is determined by the product of the individual Mueller matrices, in the correct order.

Examples : The Mueller matrix for an ideal linear polariser whose transmission axis makes an angle J with the direction of polarisation of the input beam is given by

$$\mathbf{M}(\xi) = \frac{1}{2}\begin{bmatrix} 1 & cos2\xi & sin2\xi & 0 \\ cos2\xi & cos^2 2\xi & cos2\xi sin2\xi & 0 \\ sin2\xi & cos2\xi sin2\xi & sin^2 2\xi & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \tag{2.24}$$

Now, if the input Stokes vector is given by $\begin{bmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{bmatrix}$ and the output beam is represented by $\begin{bmatrix} I_o \\ Q_o \\ U_o \\ V_o \end{bmatrix}$, we can see from above that the intensity transmitted by the ideal polariser, I_t is given by

$$I_t = \frac{1}{2}(I_i + Q_i \cos 2\mathrm{J} + U_i \sin 2\mathrm{J})$$

Consider an ideal retarder, an element that converts linear state of polarisation into elliptical, by introducing a phase difference between the two orthogonal components polarised parallel and perpendicular to the optic axis of the medium. If p is the angle made by the direction of polarisation of the light with the optic axis of the retarder, then the Mueller matrix for such a retarder is written as

$$\mathbf{M} = \begin{bmatrix} 1 & 0 & 0 & \\ 0 & \mathrm{C}^2 + S^2 \cos\delta & \mathrm{SC}(1-\cos\delta) & -S\sin\delta \\ 0 & \mathrm{SC}(1-\cos\mathrm{S}) & \mathrm{S}^2 + \mathrm{C}^2\cos\delta & C\sin\delta \\ 0 & \mathrm{S}\sin\delta & -C\sin\delta & \cos 6 \end{bmatrix}$$

where, $\mathrm{C} = \cos 2\beta$, $\mathrm{B} = \sin 2\beta$, and δ is the retardance introduced between the two orthogonal components, which depends upon the thickness of the retarder. It is a simple matter to verify through Mueller matrices, the construction of a circular polariser by using a properly ordered combination of a linear polariser and a linear retarder.

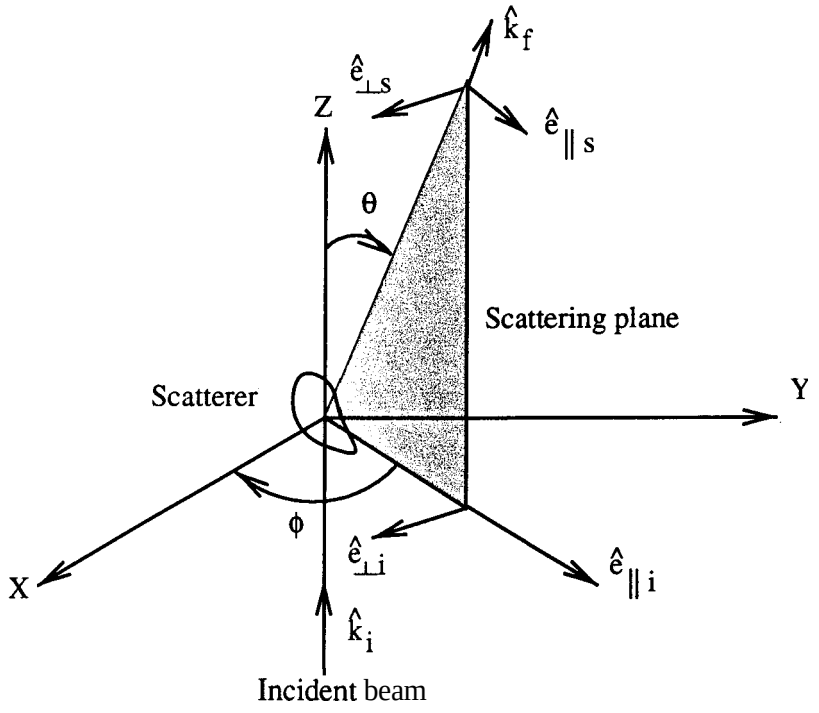

Figure **2.3:** *Geometry adopted for describing light scattering from a particle. The plane containing the incident and scattered wave-vectors ($\hat{k}_i$ and $\hat{k}_f$) is called the scattering plane. Parallel and perpendicular polarisations ($\hat{e}_{\parallel}$ and $\hat{e}_{\perp}$) are defined with respect to the scattering plane.*

2.4 Scattering particle Mueller matrix

The Mueller matrix elements for a scatterer depend upon the size the shape of the scatterer, the refractive index contrast between the scatterer and the surrounding medium, the angle of scattering and the azimuth of the scattering plane. Figure **2.3** illustrates the geometrical aspects of a scattering event, and we retain this geometry for all the discussions pertaining to a scattering event throughout this thesis, unless otherwise specified.

The figure **2.3** shows a particle of an arbitrary shape illuminated by a plane wave, propagating along the $+Z$ axis. The origin is chosen to be at any point inside the particle. The scattering direction and the incident direction together define the scattering plane. The scattering plane is uniquely determined by the azimuthal angle ϕ, except when the

scattering direction is parallel to the incident, i.e., total forward scattering. In that case, any plane containing the Z axis can be taken as the scattering plane.

The incident electric field $\mathbf{E}_i$, lying in the XY plane is resolved into components parallel. and perpendicular to the scattering plane, i.e., $E_{\|i}$ and $E_{\perp i}$ respectively. Similarly, the scattered electric field in the far-field region is resolved into components $E_{\|s}$ and $E_{\perp s}$. Since the interaction is assumed linear, the amplitude of the scattered field is a linear function of the amplitude of the incident field. This can be conveniently stated in matrix form as

$$\begin{bmatrix} E_{\|s} \\ r \end{bmatrix}_{scattered} = \frac{e^{ik(r-z)}}{-ikr} \begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} \begin{bmatrix} E_{\|i} \\ E_{\perp i} \end{bmatrix}_{incident} \tag{2.25}$$

where S_j $(j = 1, 2, 3, 4)$ are the elements of the amplitude matrix and are functions of the scattering angle 8 and azimuthal angle ϕ.

The relation between the Stokes vectors of the incident and the scattered light can be obtained from equation 2.22. The sixteen elements of the Mueller matrix are then expressed in terms of S_j's and their complex conjugates. For a scatterer of an arbitrary shape, seven of the sixteen elements of the matrix are independent. In the situation of light scattering by a collection of particles, the scattering matrix for the collection is merely the sum of the matrices of the individual particles. In that case, all the sixteen elements of the Mueller matrix are nonzero and independent.

In the case of spherical scatterers, $S_3 = S_4 = 0$. Mie theory computes the terms S_1 and S_2, which are functions of 8, in the form of an infinite sum which can be terminated after sufficient number of terms. The Mueller matrix consists of only four independent elements. The input and the output Stokes vectors are then related as

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix}_{output} = \frac{1}{k^2 r^2} \begin{bmatrix} S_{11}(\theta) & S_{12}(\theta) & 0 & 0 \\ S_{12}(\theta) & S_{11}(\theta) & 0 & 0 \\ 0 & 0 & S_{33}(\theta) & S_{34}(\theta) \\ & & 0 & -S_{34}(\theta) \quad \tilde{S}_{33}(\hat{\theta}) \end{bmatrix} \begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix}_{input} \tag{2.26}$$

where k is the wave-vector and r is the distance travelled before the scattering event. The

S_{ij}'s are given by

$$S_{11}(\theta) = \frac{1}{2}(|S_1|^2 + |S_2|^2) \tag{2.27}$$

$$S_{12}(\theta) = \frac{1}{2}(|S_2|^2 - |S_1|^2) \tag{2.28}$$

$$S_{33}(\theta) = \frac{1}{2}(S_2^* S_1 + S_1^* S_2) \tag{2.29}$$

$$S_{34}(\theta) = \frac{i}{2}(S_2^* S_1 - S_1^* S_2) \tag{2.30}$$

One just requires to calculate four independent matrix elements to characterise completely the effect of the scattering event on the incident light. Then, from the above mechanics applied recursively, one may study the evolution of polarisation state inside a random medium by keeping track of the Stokes parameters of the light during successive scattering events. The advantages of the Mueller matrix formalism are that, it gives us a simple means of determining the polarisation of the scattered light, given any arbitrary polarisation of the incident light. The elements of the Mueller matrix for a spherical scatterer can be exactly calculated by the Mie theory. Well-documented codes are available and easily accessible for the purpose[2, 8].

Bibliography

[1] H. C. van de Hulst, Light Scattering by Small Particles, (Dover, New York, 1981).

[2] C. F. Bohren and D. R. Huffman, Absorption and scattering of light by small particles, (Wiley, New York, 1983).

[3] W. A. Shurcliff, Polarised light, production and use, (Harvard University Press, Cambridge, Mass, 1962).

[4] D. Clarke and J. F. Grainger, Polarised light and optical measurements, (Pergamon, Oxford, 1971).

[5] C. Brosseau, Fundamentals of polarised light, (Wiley, New York, 1998).

[6] G. G. Stokes, Trans. Camb. Philos. Soc. 9, 399 (1852); reprinted in Mathematical and Physical Papers, Vol. III, (Cambridge University Press, Cambridge, 1901).

[7] H. Mueller, J. Opt. Soc. Am. **38,** 661 (1948).

[8] P. W. Barber and S. C. Hill, Light scattering by particles : Computational methods, (World Scientific, Singapore, 1990).