COMBINING RAS AND TAS TECHNIQUES FOR APPLICATION OF THE METHOD FOR ABSORBING PROBES

By

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Abstract

Transmission Anisotropy Spectroscopy (TAS) or/and Reflection Anisotropy Spectroscopy (RAS) are a powerful non-destructive and sensitive optical probe of surface that is capable of operating with in a wide range of environments. In this thesis, by applying these optical techniques attempt was made to determine the degree of anisotropy of a biaxial Potassium Titanyl Phosphate (KTP) crystal and its value is of the order of 10^{-3} and 10^{-4} for reflection and transmission respectively. It is also seen that the absorption correction of KTP crystal is very small. Therefore the coupled techniques are also powerful for the determination of absorption loss of a transmitting probes. The methods used in this work can also be used in situ real-time measurements.

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Introduction

Interaction of electromagnetic waves with crystals results in optical properties. Anisotropic crystalline has many properties which are direction dependent. Potassium Titanyl Phosphate (KTP or $KTiOPO_4$) is one of these crystals and this paper deals about its surface properties. Potassium Titanyl Phosphate (KTP or $KTiOPO_4$) is a nonlinear optical material suitable for use in many optical systems.

Structurally, Potassium Titanyl Phosphate (KTP) is orthorhombic and belongs to the acentric point group C_2. Its complicated structure is characterized by chains of TiO_6 octahedral linked at two corners by alternating long and short Ti-O bonds. KTP possesses optical properties that allow it to be used for both intra- and extracavity laser applications. It is optically transparent from 350 nm to 3500 nm. The optical spectrum is structure-free except for traces of OH-absorption bands observed at 2800 nm and 3800 nm. The refractive indices vary slowly with changes in wavelength and temperature [33].

The nonlinear optical coefficients of KTP are comparable to those of Ba_2NaNb_{5015} and KTP can be phase matched at 1060 nm using either Type I or Type II interactions. In Type II interactions, KTP has large angular and temperature bandwidths as well as high nonlinear coefficients and damage thresholds. It has a high conversion efficiency for second harmonic generation (SHG) of laser light (NdYAG laser) with

fundamental wavelengths between 994 nm and 2500 nm. This material is also well suited for use as an optical parametric oscillator (OPO). KTP's wide tuning range and high conversion efficiencies mean that short crystals can be used in this application. Another application well suited for KTP is quasi phase matching (QPM). In this process z-oriented waveguides of KTP are periodically poled and pumped with diode lasers to generate blue to near UV wavelengths.

KTP also possesses electro-optical properties that are comparable to those of $LiNbO_3$ for bulk modulator applications. KTP is also a superior material for waveguide electro-optical modulators. When these properties are coupled with KTP's high damage threshold, wide optical bandwidth (>15GHz), thermal and mechanical stability, the combination makes it a unique material for modulator applications.

KTP's unique combination of these properties (high nonlinear coefficients, high damage threshold and non-hygroscopicity) make it well suited for laser system applications requiring high power, high efficiency and/or durability. One of the basic features of KTP is that the absorption loss is less than one percent [33]. This is why we are interested in discussing anisotropic properties of KTP crystal in this paper.

This paper is organized in five chapters. The first chapter generally deals with the interaction of electromagnetic wave with matter. The nature of light as well as general wave and Maxwell's equations are discussed. Also the propagation of electromagnetic wave through both isotropic and anisotropic crystals is included and in addition Fresnel's equations are elaborated. Chapter two entirely explains the polarization phenomenon and based on the Jones calculus and Stokes vectors its mathematical expressions are obtained. The third chapter briefly discusses the basic principles and experimental setup of RAS and TAS. Experimental results along with

their interpretations are explained in chapter four. The last chapter, chapter five, consists of brief summary and conclusion.

Chapter 1

THEORY OF LIGHT

1.1 Background of Light

If one were to ask the question "What is light, really?" there can be no simple answer. There is no familiar object or macroscopic model to employ as an analogy [2]. This question immutably remains while the answer subtly changes through time. The search for the explanation of the properties of light dates back to remote antiquity. The rectilinear propagation of light, as well as law of reflection, was explained by Euclid in 300B.C [1, 4].

The evolution in our understanding of the physical nature of light forms one of the most fascinating accounts in the history of science. Since the dawn of modern science in the sixteenth and seventeenth centuries light has been pictured either as particles or waves -incompatible model -each of which employed a period of prominence among the scientific community. In the twentieth century it became clear that somewhat light was both wave and particle, yet it was precisely neither. This concept refereed to as the wave-particle duality. The solution was achieved through the creation of quantum electrodynamics, one of the most successful theoretical structures in the

annals of physics [2, 3].

In the seventeenth century the most prominent advocate of a particle theory of light was Issac Newton [4], the same creative giant who had erected a complete science of mechanics and gravity. In his treatise Optics, Newton clearly regarded rays of light as streams of very small particles emitted from a source of light and traveling in straight lines. At the same time, Newton was aware of the phenomenon now referred to as Newton's fringes [3, 4].

Christian Huygens, a Dutch scientist contemporary with Newton, championed the view (in his Treatise of Light) that light is a wave motion; spreading out from a light source in all directions [2] and propagating through an all-pervasive elastic medium called the ether. Adopting the wave theory, Huygens was able to derive the laws of reflection and refraction and to explain double refraction [2, 3] in calcite as well.

Within two years of the century of the publication of Newton's Optics, the Englishman Thomas Young performed a decisive experiment that seemed to demand a wave theory of light. It was the double-slit experiment, in which an opaque screen with two small, closely spaced openings was illuminated by monochromatic light from a small source. The "shadows" observed formed a complex interference pattern like those produced with water waves [3].

Victories for the wave theory continued up to the twentieth century. In the mood of scientific confidence that characterized the latter part of nineteenth century, there was little doubt that light, like most other classical areas of physics, was well understood.

In 1821 Augustin Fresnel published results of his experiments and analysis, which required that light be a transverse wave. On this basis, double refraction in calcite could be understood as a phenomenon involving polarized light. It had been assumed

that light wave in an ether were necessary longitudinal, like sound weaves in a fluid, which can't support transverse vibrations. For each of two components of polarized light, Fresnel developed the Fresnel equations [24], which give the amplitude of reflected and transmitted at a plane of interface separating two optical media [3].

It was Luis de Broglie who saw the other side of picture, particle nature of light. In 1924 he published his speculations that sub atomic particles are endowed with wave particles. Thus the wave-particle duality came full circle. Light behaved like waves in its propagation and particle in the phenomena of interference and diffraction; it would, however, also behave as particles in its interaction with matter, as in the photoelectric effect [3].

1.2 Macroscopic Fields and Maxwell's Equations

1.2.1 Maxwell's Equations

When one applies Reflectance Anisotropy Spectroscopy (RAS) and Transmission Anisotropy Spectroscopy (TAS) as an optical techniques, it wouldn't give any sense without considering the Maxwell's equations. Propagation of light can best be understood in terms of electromagnetic waves [4], on the other hand, the behaviour of electric field $\overrightarrow{E}$ and magnetic field $\overrightarrow{H}$, which are independent of one another can be described by a total of four equations. Two of which relate charge density (ρ) and current density ($\overrightarrow{J} = \sigma \overrightarrow{E}$) [4, 5, 6], where σ is the conductivity of the material.

Macroscopic regions of matter can contain a net charge or current density [25]. For a neutral dielectric medium (no free charges), the Maxwell's equations are

$$\overrightarrow{\nabla} \cdot \overrightarrow{D} = 0 \tag{1.2.1}$$

$$\overrightarrow{\nabla} \cdot \overrightarrow{B} = 0 \tag{1.2.2}$$

$$\overrightarrow{\nabla} \times \overrightarrow{E} = -\frac{\partial \overrightarrow{B}}{\partial t} \tag{1.2.3}$$

$$\overrightarrow{\nabla} \times \overrightarrow{H} = \frac{\partial \overrightarrow{D}}{\partial t} + \overrightarrow{J_c} \tag{1.2.4}$$

where J_c is the conduction current density. For macroscopically average fields, we define the magnetic induction, $\overrightarrow{B}$ as

$$\overrightarrow{B} = \mu(\overrightarrow{H} + \overrightarrow{M}) \tag{1.2.5}$$

and the electric displacement vector,$\overrightarrow{D}$, as [25]

$$\overrightarrow{D} = \epsilon \overrightarrow{E} + \overrightarrow{P} \tag{1.2.6}$$

where the polarization $\overrightarrow{P}$ is the electric dipole moment peer unit volume of the medium and $\overrightarrow{M}$ is the volume density of magnetic dipole, called magnetization. Polarization is the only term in the Maxwell's equations relating directly to the medium.

Up on substitution of equations (1.2.4) and (1.2.5) in to equations (1.2.1) to (1.2.3), the Maxwell's equations can be rewritten as

$$\overrightarrow{\nabla} \cdot \overrightarrow{E} = -\frac{1}{\epsilon}\overrightarrow{\nabla} \cdot \overrightarrow{P} + \frac{1}{\epsilon}\rho \tag{1.2.7}$$

$$\overrightarrow{\nabla} \cdot \overrightarrow{H} = -\overrightarrow{\nabla} \cdot \overrightarrow{M} \tag{1.2.8}$$

$$\overrightarrow{\nabla} \times \overrightarrow{E} = -\mu\frac{\partial \overrightarrow{H}}{\partial t} - \mu\frac{\partial \overrightarrow{M}}{\partial t} \tag{1.2.9}$$

$$\overrightarrow{\nabla} \times \overrightarrow{H} = \epsilon\frac{\partial \overrightarrow{E}}{\partial t} + \frac{\partial \overrightarrow{P}}{\partial t} + \overrightarrow{J_c} \tag{1.2.10}$$

where ϵ is the primitivity and μ is the permeability of the material.

1.2.2 The General Wave Equation

For nonmagnetic, electrical neutral medium, $\overrightarrow{M}$ and ρ are both zero [25]. Therefore,in free space Maxwell's equations reduced to the following.

$$\overrightarrow{\nabla} \times \overrightarrow{E} = \mu_0 \frac{\partial \overrightarrow{H}}{\partial t} \tag{1.2.11}$$

$$\overrightarrow{\nabla} \times \overrightarrow{H} = \epsilon_0 \frac{\partial \overrightarrow{E}}{\partial t} + \frac{\partial \overrightarrow{P}}{\partial t} + \overrightarrow{J_c} \tag{1.2.12}$$

$$\overrightarrow{\nabla} \cdot \overrightarrow{E} = -\frac{1}{\epsilon_0} \overrightarrow{\nabla} \cdot \overrightarrow{P} \tag{1.2.13}$$

$$\overrightarrow{\nabla} \cdot \overrightarrow{H} = 0 \tag{1.2.14}$$

The general wave equation for $\overrightarrow{E}$ field [4, 7] is obtained by taking the curl of equation (1.2.11) and the time derivative of the equation (1.2.12) and eliminating $\overrightarrow{H}$. The result is

$$\overrightarrow{\nabla} \times (\overrightarrow{\nabla} \times \overrightarrow{E}) + \frac{1}{c^2} \frac{\partial^2 \overrightarrow{E}}{\partial t^2} = -\mu_0 \frac{\partial^2 \overrightarrow{P}}{\partial t^2} - \mu_0 \frac{\partial \overrightarrow{J_c}}{\partial t} \tag{1.2.15}$$

The two terms on the right-hand side of the above equation are called source terms. They stem from the presence of polarization changes and conduction charges, respectively, within the medium. The way in which the propagation of light is affected by the source is revealed by the solution of the wave equation when the source terms are included. In the case of non-conducting media the polarization term $-\mu_0 \frac{\partial^2 \overrightarrow{P}}{\partial t^2}$ is of importance. It turns out that this term leads to an explanation of many optical effects, including, dispersion, absorption, double refraction, and optical activity to mention only few [7].

1.3 Interaction of Light with Matter

1.3.1 Propagation of Light in an Isotropic Dielectric Materials

In this section, we shall consider the propagation of light waves in homogeneous (isotropic) dielectric materials. The starting point of the treatment of light in manner from electromagnetic prospective is contained with in the four Maxwell's equations. In a nonconducting, isotropic medium [25], the electrons are permanently bound to the atoms comprising the medium and there is no preferential direction. This is what is meant by a simple isotropic dielectric such as glass. Suppose that each electron, of charge $-e$, in a dielectric is displaced a distance $\overrightarrow{r}$ from its equilibrium position. The resulting macroscopic polarization of the medium is given by

$$\overrightarrow{P} = -Ne\overrightarrow{r} \tag{1.3.1}$$

where N is the number of electron per unit volume. If the displacement of the

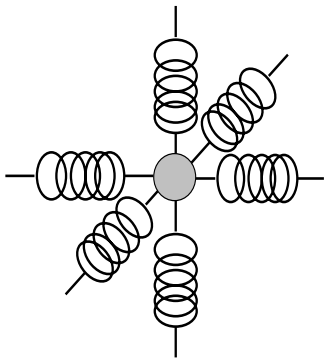

Figure 1.1: The classical oscillation model for an isotropic medium - all the springs are the same, and the oscillator can vibrate equally in all directions.

electron is the result of an application of a static electric field $\overrightarrow{E}$, and if the electron

is elastically bounded to its equilibrium position with a force constant K, then the force equation is

$$-e\overrightarrow{E} = K\overrightarrow{r} \tag{1.3.2}$$

The static polarization is therefore given by

$$\overrightarrow{P} = \frac{Ne^2}{K}\overrightarrow{E} \tag{1.3.3}$$

However, if the impressed field $\overrightarrow{E}$ varies with time, the above equation doesn't hold. In order to find the true polarization in this case, we must take the actual motion of the electrons into account. To do this we consider the bound electrons as classical damping harmonic oscillators. The differential equation of motion is

$$m\frac{d^2\overrightarrow{r}}{dt^2} + m\gamma\frac{d\overrightarrow{r}}{dt} + K\overrightarrow{r} = -e\overrightarrow{E} \tag{1.3.4}$$

The term $\mathrm{m}\gamma\frac{d\overrightarrow{r}}{dt}$ represents a frictional damping force that is proportional to the velocity of the electron, the proportionality constant being $m\gamma$.

Now suppose that the applied electric field $\overrightarrow{E}$ varies harmonically with time according to the usual factor $\exp(-i\omega t)$ and consequently displacement also varies harmonically with this factor. Assuming that the motion of the electron has the same harmonic time dependence, we find equation (1.3.4) becomes

$$(-m\omega^2 - i\omega m\gamma + K)\overrightarrow{r} = -e\overrightarrow{E} \tag{1.3.5}$$

consequently, the polarization, equation (1.3.1) is given by

$$\overrightarrow{P} = \frac{Ne^2}{-m\omega^2 - i\omega m\gamma + K}\overrightarrow{E} \tag{1.3.6}$$

It reduces to static value, eqn. (1.3.3), when $\omega = 0$. Thus for a given amplitude of the impressed electric field, the amount of polarization varies with frequency. The

phase of $\overrightarrow{P}$ relative to that of electric field, also depends on the frequency. This is shown by the presence of the imaginary term in the denominator.

A more significant representation of eqn. (1.3.6) is

$$\overrightarrow{P} = \frac{\frac{Ne^2}{m}}{\omega_0^2 - \omega^2 - i\omega\gamma}\overrightarrow{E} \tag{1.3.7}$$

in which we have used the natural resonance frequency of the bound electrons ω_0 [25] given by

$$\omega_0 = \sqrt{\frac{K}{m}} \tag{1.3.8}$$

The polarization formula eqn. (1.3.7) is similar to the amplitude formula for a derived harmonic oscillators as indeed it should be, since it is the displacement of the elastically bound electrons that actually constitute the polarization. We should therefore expect to find an optical resonance phenomena of some kind occurring for light frequency in the neighbourhood of the resonance frequency ω_0.

To show how the polarization affects the ppropagation of light we return to the general wave equation (1.2.15). For an isotropic dielectric there is no conduction term. Hence we have

$$\overrightarrow{\nabla} \times (\overrightarrow{\nabla} \times \overrightarrow{E}) + \frac{1}{c^2}\frac{\partial^2 \overrightarrow{E}}{\partial t^2} = -\mu_0 \frac{Ne^2}{m}\Big(\frac{1}{\omega_o^2 - \omega^2 - i\gamma\omega}\Big)\frac{^2\overrightarrow{E}}{\partial t^2} \tag{1.3.9}$$

Also, from the linear relationship between $\overrightarrow{P}$ and $\overrightarrow{E}$, it follows from eqn. (1.2.13) that $\overrightarrow{\nabla} \cdot \overrightarrow{E} = 0$. Consequently $\overrightarrow{\nabla} \times \big(\overrightarrow{\nabla} \times \overrightarrow{E}\big) = -\nabla^2 \overrightarrow{E}$, and the above wave equation reduces to somewhat simpler one

$$\nabla^2 \overrightarrow{E} = \frac{1}{c^2}\big(1 + \frac{Ne^2}{m}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}\big)\frac{\partial^2 \overrightarrow{E}}{\partial t^2} \tag{1.3.10}$$

after rearranging terms and using the relation $\frac{1}{c^2} = \mu_0\epsilon_0$ [7, 8].

1.3.2 Propagation of Light in Anisotropic Crystals

Until now we have confined our attention to the propagation of light through isotropic medium, i.e, substances whose optical properties are the same in all directions. In many crystals, however, the optical as well as other physical properties are different along different directions. Here it is now the point of the discussion of the propagation of light through such type of crystals - anisotropic crystals. The distinguishing basic feature of crystalline state as far as optical properties are concerned, is the fact that the crystals are generally electrically anisotropic. This means that the polarization produced in the crystal for a given electric field is not just a simpler scalar constant times the field, but varies in a manner that depends on the direction of applied field in relation to crystal lattice. One of the consequence is that the speed of propagation of a light wave in a crystal is a function of direction of propagation and polarization of light.

It turns out that there are generally two possible values of phase velocity for a given direction of propagation. These two values are associated with mutually orthogonal polarizations of the light wave. Such type of crystals are said to be doubly refracting or birefringent. Actually not all crystals exhibit double refraction. Crystals of the cubic class of symmetry, such as sodium chloride, never exhibit double refraction, but are optically isotropic. All crystals, other than cubic crystals, do show double refraction,however [7].

A model to illustrate the anisotropic polarizations of a crystal is show in figure 1.2. A bound electron is pictured here as attached to a set of fictitious elastic springs. The springs have different stiffness for different directions of the electrons displacement

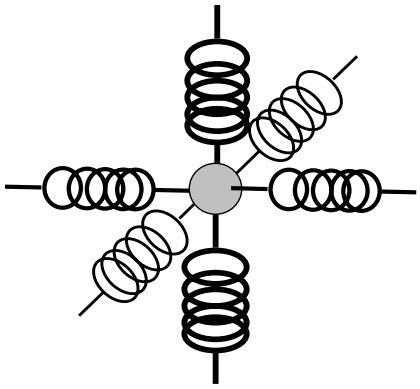

Figure 1.2: Model to show anisotropic binding of an electron in a crystal.

from its equilibrium position within the crystal lattice. Consequently, the displacement of the electron under the action of the external field $\overrightarrow{E}$ depends on the direction of the field as well as its magnitude. This is also true for the resulting polarization $\overrightarrow{P}$.

The dependence of $\overrightarrow{P}$ on $\overrightarrow{E}$ [25] is expressible as a tensor relation in the form

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \epsilon_0 \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & \chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & \chi_{33} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} \tag{1.3.11}$$

This can be represented as

$$\overrightarrow{P} = \epsilon_0 \chi \overrightarrow{E} \tag{1.3.12}$$

where χ is the electric susceptibility tensor and has the form

$$\chi = \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & \chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & \chi_{33} \end{pmatrix} \tag{1.3.13}$$

The corresponding displacement vector $\overrightarrow{D}$ is given by $\overrightarrow{D} = \epsilon_0(1+\chi)\overrightarrow{E} = \epsilon\overrightarrow{E}$, where

$$\epsilon = \epsilon_0(1+\chi) \tag{1.3.14}$$

which is known as the dielectric tensor [25].

For ordinary non-absorbing crystals the χ tensor is symmetric so there always exist a set of coordinate axes, called principal axes, such that the χ tensor assumes the diagonal form

$$\chi = \begin{pmatrix} \chi_{11} & 0 & 0 \\ 0 & \chi_{22} & 0 \\ 0 & 0 & \chi_{33} \end{pmatrix} \tag{1.3.15}$$

The three χs are known as principal susceptibilities. Corresponding to these, the quantities $K_{11} = 1+\chi_{11}$ ... , and so forth, are called the principal dielectric constants [22].

In view of eqn. (1.3.12), the general wave equation can take the form

$$\overrightarrow{K} \times (\overrightarrow{K} \times \overrightarrow{E}) + \frac{\omega^2}{c^2}\overrightarrow{E} = -\frac{\omega^2}{c^2}\chi\overrightarrow{E} \tag{1.3.16}$$

in which case we used $\exp(\overrightarrow{k} \cdot \overrightarrow{r} - \omega t)$ for the electric field. Writing in terms of components, the above equation is equivalent to the following three equations:

$$\begin{cases} (-k_y^2 - k_x^2 + \frac{\omega^2}{c^2})E_x + k_x k_y E_y + k_x k_z E_z = -\frac{\omega^2}{c^2}\chi_{11}E_x \\ k_x k_y E_x + (-k_x^2 - k_z^2 + \frac{\omega^2}{c^2})E_y + + k_y k_z E_z = -\frac{\omega^2}{c^2}\chi_{22}E_y \\ k_x k_z E_x + k_y k_z E_y + (-k_x^2 - k_y^2 + \frac{\omega^2}{c^2})E_z = -\frac{\omega^2}{c^2}\chi_{33}E_z \end{cases} \tag{1.3.17}$$

Assume that the wave is propagating in a particular direction of one of the principal axes, say the x-axis in which case $K_x = K$, $K_y = K_z = 0$ to interpret the physical meaning of the above equation and the three equations reduce to

$$\begin{cases} \frac{\omega^2}{c^2}E_x = -\frac{\omega^2}{c^2}\chi_{11}E_x \\ (-k^2 + \frac{\omega^2}{c^2})E_y = -\frac{\omega^2}{c^2}\chi_{22}E_y \\ (-k^2 + \frac{\omega^2}{c^2})E_z = -\frac{\omega^2}{c^2}\chi_{33}E_z \end{cases} \tag{1.3.18}$$

The first equation implies that $E_x = 0$, because neither ω or χ_{11} is zero. This means that the field $\overrightarrow{E}$ is transverse to the x-axis, which is the direction of propagation. If $E_y \neq o$, the second equation results in

$$k = \frac{\omega}{c}\sqrt{1+\chi_{22}} = \frac{\omega}{c}\sqrt{K_{22}} \tag{1.3.19}$$

The third equation, likewise, implies that if $E_z \neq 0$, then

$$k = \frac{\omega}{c}\sqrt{1+\chi_{33}} = \frac{\omega}{c}\sqrt{K_{33}} \tag{1.3.20}$$

Now $\frac{\omega}{c}$ is the phase velocity of the wave. Thus we have two possible phase velocities as explained earlier, namely, $\frac{c}{\sqrt{K_{22}}}$ if the $\overrightarrow{E}$ vector points in the y-direction, and $\frac{c}{\sqrt{K_{33}}}$ if the $\overrightarrow{E}$ vector is in the z-direction.

We can deduce that for any direction of the propagation vector $\overrightarrow{K}$, there are two possible values of the magnitude k and hence also for phase velocity [7, 10]. The three principal indices of refraction n_1, n_1, and n_1 are related as follows

$$\begin{cases} n_1 = \sqrt{1+\chi_{11}} = \sqrt{K_{11}} \\ n_2 = \sqrt{1+\chi_{22}} = \sqrt{K_{22}} \\ n_3 = \sqrt{1+\chi_{33}} = \sqrt{K_{33}} \end{cases} \tag{1.3.21}$$

In view of the fact that the principal indices are related with components of the χ tensor by eqn. (1.3.17), crystals can be classified according to the χ tensor as follows. In a uniaxial crystal the index of refraction that corresponds to the two equal elements, $\chi_{11} = \chi_{22}$, is called the ordinary index n_0 which does obey the Snell's law for electric fields normal to the optic axis [31], and the other index, corresponding to χ_{33}, is called the extraordinary index n_E for electric fields parallel to the optic axis [31], and doesn't obey the Snell's law [27].

An extraordinary wave, in negative crystals ($n_0 > n_E$), polarized in the z-direction [21], moves with maximum velocity in a direction perpendicular to the optical axis

Isotropic	cubic	$\chi = \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & a \end{pmatrix}$	$\chi_{11} = \chi_{22} = \chi_{33} = a$ $n = \sqrt{1+a}$
Uniaxial	*trigonal* *tetragonal* *hexagonal*	$\chi = \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix}$	$\chi_{11} = \chi_{22} = a, \chi_{33} = b$ $n_o = \sqrt{1+a}$ $n_E = \sqrt{1+b}$
Biaxial	*triclinic* *monoclinic* *orthorhombic*	$\chi = \begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & c \end{pmatrix}$	$\chi_{11} = a, \chi_{22} = b, \chi_{33} = c$ $n_1 = \sqrt{1+a}$ $n_2 = \sqrt{1+b}$ $n_3 = \sqrt{1+c}$

Table 1.1: Classification of crystals depending upon the susceptibility tensor.

(velocity proportional to $\frac{1}{n_E}$) [26] while in positive crystals ($n_0 < n_E$), polarized in the x or y direction [21], moves with minimum velocity in a direction perpendicular to optical axis. On the other hand, an ordinary wave, in the positive crystals, moves with maximum velocity in the optical axis (velocity proportional to $\frac{1}{n_0}$) and in negative crystals moves with minimum velocity in the optical axis [7]. See figure 1.3 below.

The optic axis is axis of symmetry with respect to both the crystal forms and the arrangement of atoms and if any property is measured for different directions, it is found to be the same along any line perpendicular to this axis [29].

1.4 Reflection and Refraction at a Plane Boundary

The basic laws of reflection and refraction in geometrical optics were derived earlier on the basis of Huygens and Fermat's principles [4]. Regarding light as an electromagnetic wave we show that the law of reflection and refraction can also be deduced from application of boundary conditions for electromagnetic waves [25]. More importantly, this approach also leads to the Fresnel equations [24], which describe the fraction of incident energy transmitted or reflected at the plane surface (relates also

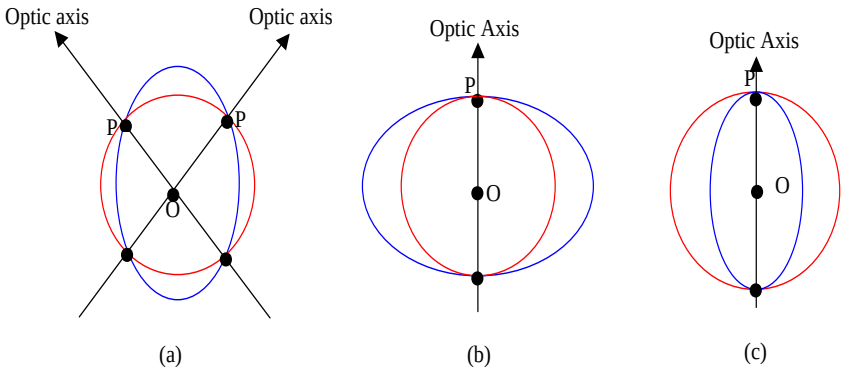

Figure 1.3: Wave fronts in: (a) biaxial crystals (b) negative uniaxial crystal (c) positive uniaxial crystals

amplitude). This quantities will be seen to depend not only on the change in refractive index and the angle of incident at the surface, but also the polarization of the incident light [11].

Consider a plane harmonic wave incident upon a plane boundary separating two different optical media (fig. 1.4) expressed by

$$\overrightarrow{E} = \overrightarrow{E}_0 \exp\left(i(\overrightarrow{k} \cdot \overrightarrow{r} - \omega t)\right) \tag{1.4.1}$$

The reflected and transmitted waves can be expressed respectively, in forms like that of the incident wave of equation (1.4.1)

$$\overrightarrow{E}_r = \overrightarrow{E}_{0r} \exp\left(i(\overrightarrow{k}_r \cdot \overrightarrow{r} - \omega_r t)\right) \tag{1.4.2}$$

$$\overrightarrow{E}_r = \overrightarrow{E}_{0r} \exp\left(i(\overrightarrow{k}_r \cdot \overrightarrow{r} - \omega_r t)\right) \tag{1.4.3}$$

In the boundary plane, where all three waves exist simultaneously, there must be a fixed relationship between the three wave amplitude (and this irradiance) that has

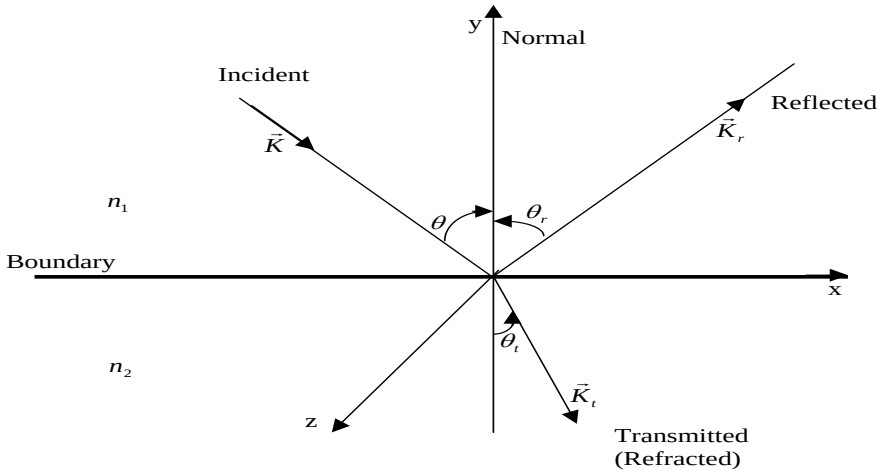

Figure 1.4: Wave vectors for light incident on a boundary separating two different optical media.

yet to be determined, since such a relationship can not depend on the arbitrary choice of a boundary point $\overrightarrow{r}$ nor a time t, it follow that the phases of the three waves, which depend on r and t, must themselves be equal:

$$(\overrightarrow{k} \cdot \overrightarrow{r} - \omega t) = (\overrightarrow{k}_r \cdot \overrightarrow{r} - \omega_r t) = (\overrightarrow{k}_t \cdot \overrightarrow{r} - \omega_t t \tag{1.4.4}$$

In particular, at the boundary point $r = 0$,

$$-\omega t = -\omega_r t = -\omega_t t$$

or

$$\omega = \omega_r = \omega_t \tag{1.4.5}$$

so that all frequencies are equal. On the other hand, at $t = 0$ within the boundary plane, eqn. (1.4.4) yields:

$$\overrightarrow{k} \cdot \overrightarrow{r} = \overrightarrow{k}_r \cdot \overrightarrow{r} = \overrightarrow{k}_t \cdot \overrightarrow{r} \qquad (at\ boundary) \tag{1.4.6}$$

Several condition can be drown from the relations of eqn. (1.4.6). First notice that by subtracting any two members, these relations are equivalent to

$$(\overrightarrow{k} - \overrightarrow{k}_r) \cdot \overrightarrow{r} = (\overrightarrow{k} - \overrightarrow{k}_t) \cdot \overrightarrow{r} = (\overrightarrow{k}_r - \overrightarrow{k}_t) \cdot \overrightarrow{r} = 0 \tag{1.4.7}$$

Equation (1.4.6) requires that the vectors $\overrightarrow{k}_r$ and $\overrightarrow{k}_t$ lie in the plane determined by the vector $\overrightarrow{k}$ and $\overrightarrow{r}$. Thus all three propagation vectors are coplanar in the xz-plane, and we conclude that the reflected and incident waves lie on the plane of incidence. Next, consider the first two members of eqn. (1.4.6), which govern the relationship between the incident and reflected waves. In terms of the angle designated fig. 1.4, they are equivalent to

$$kr \sin\theta = k_r r \sin\theta_r$$

Since both waves travel in the same medium, their wavelength are identical and so $k = k_r$. Therefore, we have the

$$law\ of\ reflection : \theta = \theta_r \tag{1.4.8}$$

Finally, the last two members of eqn. (1.4.6) are equivalent to

$$k_r r \sin\theta_r = k_t r \sin\theta_t \tag{1.4.9}$$

writing $k_r = \frac{n_r \omega}{c}$ and $k_t = \frac{n_t \omega}{c}$, eqn. (1.4.9) becomes snell's [11, 12, 13]

$$law\ of\ refraction : n_r \sin\theta_r = n_t \sin\theta_t \tag{1.4.10}$$

1.4.1 Amplitude Coefficients

It is convenient at this point to consider two different cases. The first case is that in which the electric field vector of the incident wave is parallel to the boundary

plane, that is, perpendicular to the plane of incidence. This case is called transverse electric (TE) polarization. The second case is that in which the magnetic vector of the incident wave is parallel to the boundary plane. This is called transverse magnetic (TM) polarization. The general case is handled by using appropriate linear combinations. The direction of the electric and magnetic vectors for the two cases are shown in the figure 1.5 below. As shown in the figure below the boundary is taken to

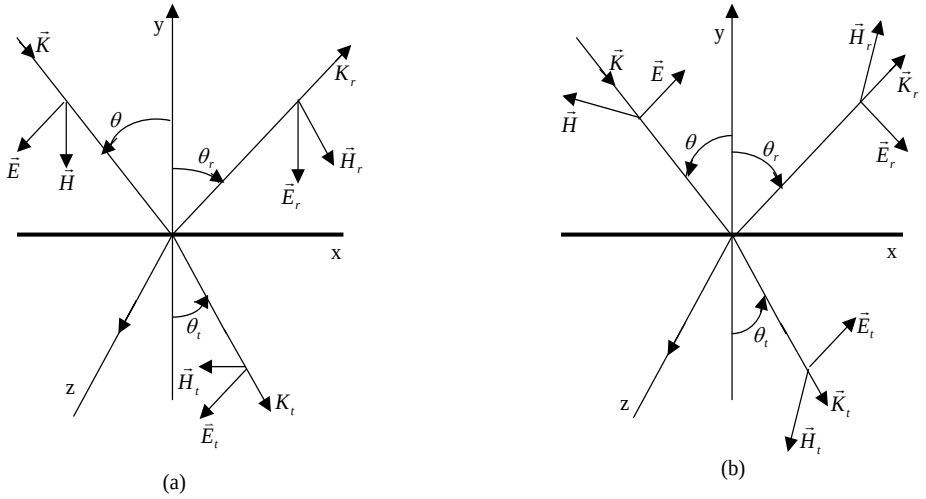

Figure 1.5: Wave vectors and associated fields for: (a) TE and (b) TM polarization.

be xz-plane so that the y-axis is normal to the boundary. The xy-plane is the plane of incidence.

We now apply the well known boundary conditions [25] which requires that the tangential components of the electric and magnetic fields are continuous as the boundary is crossed [12]. For the polarization, the requirements are:

$$E + E_r = E_t \tag{1.4.11}$$

$$B\cos\theta - B_r\cos\theta = B_t\cos\theta_t \tag{1.4.12}$$

where we have made the use of eqn. (1.4.10). The negative sign indicates the B_r-component is along the negative x-direction in the reflected beam. Similarly for TM polarization, we have

$$B + B_r = B_t \tag{1.4.13}$$

$$-E\cos\theta + E_r\cos\theta = -E_t\cos\theta_t \tag{1.4.14}$$

Magnetic fields can be expressed in terms of electric fields through the relation

$$\overrightarrow{E} = \upsilon\overrightarrow{B} = (\frac{c}{n})\overrightarrow{B}$$

Writing the index of refraction for incident reflecting media as n_1 and n_2, respectively, eqn. (1.4.11) through (1.4.14) can be recast as follows:

$$TE: \quad \begin{cases} E + E_r = E_t \\ n_1 E\cos\theta - n_1 E_r\cos\theta = n_2 E_t\cos\theta_t \end{cases} \tag{1.4.15}$$

$$TM: \quad \begin{cases} n_1 E + n_1 E_r = E_t \\ -E\cos\theta + n_1 E_r\cos\theta = -E_t\cos\theta_t \end{cases} \tag{1.4.16}$$

Next, eliminating E_t from each points of equation and solving for the reflection coefficient $r = \frac{\overrightarrow{E}_r}{\overrightarrow{E}}$,

$$TE: \qquad r = \frac{\overrightarrow{E}_r}{\overrightarrow{E}} = \frac{\cos\theta - n\cos\theta_t}{\cos\theta + n\cos\theta_t} \tag{1.4.17}$$

$$TM: \qquad r = \frac{\overrightarrow{E}_r}{\overrightarrow{E}} = \frac{n\cos\theta - \cos\theta_t}{n\cos\theta + \cos\theta_t} \tag{1.4.18}$$

where we have introduced a relative refractive index $n = \frac{n_2}{n_1}$.

1.4.2 Fresnel equation

The Fresnel equations that relate the reflection and transmission coefficients to the amplitudes can be easily obtained from the above equations. Since n and θ_t

are related to θ through Snell's law [25], $\sin\theta = n\sin\theta_t$, θ_t may be eliminated using

$$n\cos\theta_t = n\sqrt{1-\sin^2\theta_t} = \sqrt{n^2-\sin^2\theta} \tag{1.4.19}$$

The results are then

$$TE: \qquad r = \frac{\overrightarrow{E}_r}{\overrightarrow{E}} = \frac{\cos\theta - \sqrt{n^2-\sin^2\theta}}{\cos\theta + \sqrt{n^2-\sin^2\theta}} \tag{1.4.20}$$

$$TM: \qquad r = \frac{\overrightarrow{E}_r}{\overrightarrow{E}} = \frac{n^2\cos\theta - \sqrt{n^2-\sin^2\theta}}{n^2\cos\theta + \sqrt{n^2-\sin^2\theta}} \tag{1.4.21}$$

The reflectance $R = |r|^2$, for both modes. Returning to the eqs. (1.4.15) and (1.4.16), if E_r is eliminated instead of E_t, similar steps lead to the following equations describing the transmission coefficient $t = \frac{E_t}{E}$:

$$TE: \qquad t = \frac{\overrightarrow{E}_t}{\overrightarrow{E}} = \frac{2\cos\theta}{\cos\theta + \sqrt{n^2-\sin^2\theta}} \tag{1.4.22}$$

$$TM: \qquad t = \frac{\overrightarrow{E}_t}{\overrightarrow{E}} = \frac{2n\cos\theta}{n^2\cos\theta + \sqrt{n^2-\sin^2\theta}} \tag{1.4.23}$$

The transmittance $T = |t|^2$ and hence equations (1.4.20) through (1.4.23) are the Fresnel equations, giving reflection and transmission coefficients, the ratio of both reflected and transmitted electric field amplitudes to the incident electric field amplitude [11, 12].

Chapter 2

Polarization of Light Waves

2.1 Concept of Polarization

In the preceding chapter we have seen the nature of light in details. In this chapter we are going to see the basic properties of polarization of electromagnetic waves. Polarization is the process of transforming unpolarized light in to polarized light [23]. Polarization is the property that is common to all types of vector waves. Electromagnetic waves possess this property and so do elastic and spin weaves in solids, for example [14]. The vibration of the polarization of light occur in a single plane [23].

2.2 Unpolarized Light

Unpolarized light is not an elementary state of polarization and is in a sense more complicated than the polarized light [15]. It is a light wave which is vibrating in more than one plane [23]. In general, an individual microscopic source of light (such as atoms or molecules) emits light that in a given direction has a well-defined polarization state. It is the superposition of many individual contributions to form

the total macroscopic electric field that can produce unpolarized light [15].

2.3 Polarized Light

Because the field vectors are perpendicular to the propagation direction, there is a degree of freedom involving their orientation not available in the case of scalar waves. The polarization properties of light are manifestations of this degree of freedom [15]. Light is a transverse EMW and consider only the cases where the electric field vector is resided in a fixed plane. This plane is referred to as the plane of vibration and the light is said to be plane polarized. The amplitudes and relative phase of the interacting waves will determine the state of polarization of the composite disturbance [16, 17]. The monochromatic wave can take the form

$$\overrightarrow{E}(x, y, z) = E_x\hat{x} + E_y\hat{y} \tag{2.3.1}$$

where

$$E_x(x, y, z) = A_x \cos(\omega t - kz - \phi_x) \tag{2.3.2}$$

$$E_y(x, y, z) = A_y \cos(\omega t - kz - \phi_y) \tag{2.3.3}$$

with positive amplitude A_x, A_y and phase angles ϕ_x and ϕ_y. The classification of the types of polarized light will depend on the relative phase

$$\delta = \phi_y - \phi_x \tag{2.3.4}$$

and the relative sizes of the amplitudes.

2.3.1 Elliptical polarization

Before the manipulation of elliptical polarization,we need to see the resultant electric field which is the superposition of the two components as stated above [31]. Using the trigonometric properties and letting $\phi_x = 0$, the resultant will be

$$E = E_x + A_y \frac{E_x}{A_x} \cos\delta + A_y \sqrt{1 - \cos^2(\omega t - kz)} \sin\delta \tag{2.3.5}$$

From eqn. (2.3.3), one obtains

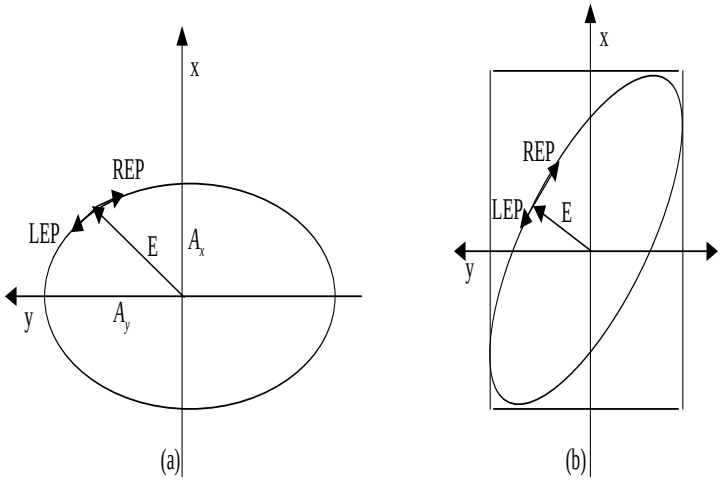

Figure 2.1: Representation of elliptically polarized light for: (a) $\phi = \frac{\pi}{2}$; (b) $\phi \neq \frac{\pi}{2}$

$$\frac{E_y}{A_y} = \frac{E_x}{A_x} \cos\delta + \sqrt{1 - \left(\frac{E_x}{A_x}\right)^2} \tag{2.3.6}$$

which results in the following equation.

$$\left(\frac{E_y}{A_y}\right)^2 + \left(\frac{E_x}{A_x}\right)^2 - \frac{2E_x E_y}{A_x A_y} \cos\delta = \sin^2\delta \tag{2.3.7}$$

This equation is the combination of two electric fields mutually perpendicular to each other and differing in phase by an angle δ and represents an elliptical polarization of light. The other types of polarization can be obtained in special cases.

2.3.2 Circular polarization

The circular polarization will be obtained when the amplitudes are equal to some constant A and $\delta = \pm\frac{\pi}{2}$ that is eqn. (2.3.7) becomes the equation of circle as

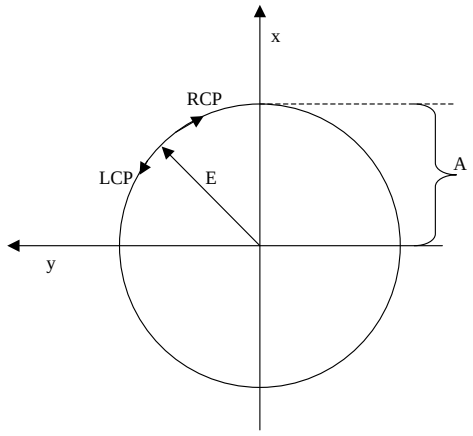

Figure 2.2: Representation of circularly polarized light as seen in a plane perpendicular to the direction of propagation . The electric field vector sweeps out a circle as time evolves.

$$E_x^2 + E_y^2 = A^2 \tag{2.3.8}$$

When $\delta = +\frac{\pi}{2}$ E_y leads E_x by $\frac{\pi}{2}$rad; that is, E_y reaches its maximum value a quarter of a cycle before E_x does. This represents a clockwise circle - right circularly polarized (RCP) which can be designated as R-state. The other case is left circularly polarized

(LCP) - stated as L-state when $\delta = -\frac{\pi}{2}$. In this case all the observations are reversed [31].

2.3.3 Linear polarization

This type of polarization sometimes called plane polarized light. Linear polarized light results when A_x and A_y are constat real vectors and $\phi = 0$ or $\phi = \pi$ [31] for then E_y proportional to E_x

$$E_y = \frac{A_y}{A_x} E_x \qquad for \qquad \phi = 0 \tag{2.3.9}$$

$$E_y = -\frac{A_y}{A_x} E_x \qquad for \qquad \phi = \pi \tag{2.3.10}$$

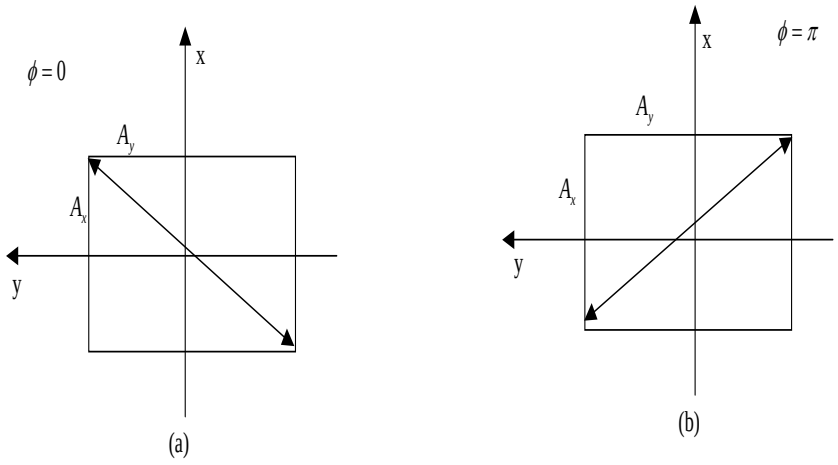

Figure 2.3: Representation of linearly polarized light.

2.4 Jones Calculus

Over fifty years ago R.Clark Jones described a very useful technique for describing the change in polarization state of the light as it passed through an optical system containing various interfaces and polarizing elements [28]. The Jones Calculus treats the optical system as a linear system describable by an appropriate Jones matrix that transforms vectors describing the polarization state of the wave. In this sense the Jones Calculus is analogous to paraxial ray analysis.

2.4.1 Jones Vectors

A more general way of expressing the complex amplitude of plane harmonic wave is

$$E_0 = \hat{i}E_{0x} + \hat{j}E_{0y} \tag{2.4.1}$$

where E_{0x} and E_{0y} can both be complex. Accordingly, they can be written in the exponential form as

$$E_{0x} = |E_{0x}| \exp(i\phi_x) \tag{2.4.2}$$

$$E_{0y} = |E_{0y}| \exp(i\phi_y) \tag{2.4.3}$$

A convenient notation for the above pair of complex amplitudes is the following matrix known as the Jones vector

$$\begin{pmatrix} E_{0x} \\ E_{0y} \end{pmatrix} = \begin{pmatrix} |E_{0x}| \exp(i\phi_x) \\ |E_{0y}| \exp(i\phi_y) \end{pmatrix} \tag{2.4.4}$$

where ϕ_x and ϕ_y are the appropriate phases. This Jones vector represents the state of polarization (Table 2.1)

One of the applications of the Jones notation is calculating the result of adding two or more waves of given polarizations whose result is obtained simply by adding the Jones vectors. For instance one can easily find the sum of right and left circular polarization of equal amplitude gives linearly polarized light in x-direction and its amplitude is twice that of either of the circular components.

State of polarization	**Jones vector**
Linearly polarized in $x-$*direction*	$\begin{pmatrix} 1 \\ 0 \end{pmatrix}$
Linearly polarized in $y-$*direction*	$\begin{pmatrix} 0 \\ 1 \end{pmatrix}$
Linearly polarized at 45^0 *relative to* $x-$*axis*	$\begin{pmatrix} 1 \\ 1 \end{pmatrix}$
Left circular polarization	$\begin{pmatrix} 1 \\ i \end{pmatrix}$
Right circular polarization	$\begin{pmatrix} 1 \\ -i \end{pmatrix}$
Elliptically left polarization	$\begin{pmatrix} 2 \\ i \end{pmatrix}$
Elliptically right polarization	$\begin{pmatrix} 1 \\ 2i \end{pmatrix}$

Table 2.1: Jones vectors for different states of polarization.

Another use of the matrix notation is computing the effect of inserting a linear optical element, on the train of such elements, in to a beam of light of given polarization. The optical elements are represented by 2×2 matrix called Jones matrices. We will see this in the next subsection.

2.4.2 Jones Matrix

In a linear system description the output Jones vector of a light wave after it has interacted with an optical system has components that are linearly related to

its input components. The types of optical devices that can be so represented include linear polarizers, circular polarizers, phase retarders (quarter-wave plates and so forth), isotropic phase changers, and isotropic observers.

Let the vector of the incident light be $\begin{pmatrix} A \\ B \end{pmatrix}$ and the vector of emerging light be $\begin{pmatrix} A' \\ B' \end{pmatrix}$. The relation of the incident and the emerging light represented as

$$\begin{pmatrix} a & b \\ c & d \end{pmatrix} \begin{pmatrix} A \\ B \end{pmatrix} = \begin{pmatrix} A' \\ B' \end{pmatrix} \tag{2.4.5}$$

where $\begin{pmatrix} a & b \\ c & d \end{pmatrix}$ is the Jones matrix of the optical element. If light is sent through a train of optical elements, then the result is given by the matrix multiplication

$$\begin{pmatrix} A' \\ B' \end{pmatrix} = \begin{pmatrix} a_n & b_n \\ c_n & d_n \end{pmatrix} ... \begin{pmatrix} a_2 & b_2 \\ c_2 & d_2 \end{pmatrix} \begin{pmatrix} a_1 & b_1 \\ c_1 & d_1 \end{pmatrix} \begin{pmatrix} A \\ B \end{pmatrix} \tag{2.4.6}$$

It should be noticed that the Jones calculus (vector) is of use only for computing results with light that is initially polarized in some way, that means, there is no Jones vector representation for unpolarized light [12, 18]

2.4.3 Orthogonal and Orthonormal Polarization

Two waves whose state of polarization are represented by the complex vector amplitudes E_1 and E_2 are said to be orthogonally polarized if

$$E_1 \cdot E_2^* = 0 \tag{2.4.7}$$

where the asterisk denotes the complex conjugate.

Optical Device	**Jones Matrix**
Linearly polarizer oriented along $x-$*direction*	$\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$
Linearly polarizer oriented along $y-$*axis*	$\begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}$
Linearly polarizer oriented at an angle θ *to the* $x-$*axis*	$\begin{pmatrix} \cos^2\theta & \sin\theta\cos\theta \\ \sin\theta\cos\theta & \sin^2\theta \end{pmatrix}$
Right circular polarizer	$\frac{1}{2}\begin{pmatrix} 1 & i \\ -i & 1 \end{pmatrix}$
$Left$ *circular polarizer*	$\frac{1}{2}\begin{pmatrix} 1 & -i \\ i & 1 \end{pmatrix}$
$Quarter-$*wave plate,* $fast$ *axis horizontal*	$\exp(\frac{i\pi}{4})\begin{pmatrix} 1 & 0 \\ 0 & i \end{pmatrix}$
$Quarter-$*wave plat,* $fast$ *axis vertical*	$\exp(\frac{i\pi}{4})\begin{pmatrix} 1 & 0 \\ 0 & -i \end{pmatrix}$

Table 2.2: Jones matrix for common optical devices.

On the other hand such two Jones vectors E_1 and E_2 are said to be orthonormal to each other when they satisfy the condition [14]

$$E_1^* \cdot E_2 = E_1 E_2^* = 1$$

For linearly polarized light, orthogonality merely means that the fields are polarized at right angles to one another. In the case of circular polarization it is readily seen the right and left circular polarizations are mutually orthogonal states. But, there is a corresponding orthogonal polarization for any type of polarization. In terms of Jones vectors it is easy to verify that $\begin{pmatrix} A_1 \\ B_1 \end{pmatrix}$ and $\begin{pmatrix} A_2 \\ B_2 \end{pmatrix}$ are orthogonal if

$$A_1 A_2^* + B_1 B_2^* = 0 \tag{2.4.8}$$

2.5 Poincare Sphere

In the previous section we have seen the matrix representation of polarization. Now in this section we seek for an other method interpreted in the following manner. Poincare (1892) used a spherical representation to identify the various states of polarization of light [14, 19]. The Poincare sphere is a unit radius S_0 spherical surface each point of which signifies a different polarization state. It is the polarization space in the form of spherical surface whose points are one by one correspondence with different state of polarization of light. The state of polarization of light can be described by using different kinds of polarization space. The complex plane representation of polarized light gives another space whose points are in one to one correspondence with different possibilities of polarization.

In terms of the cartesian components of the transverse electric field, the four Stokes parameters are defined as follows [14]:

$$S_0^2 = S_1^2 + S_2^2 + S_3^2 = E_x^2 + E_y^2 \tag{2.5.1}$$

$$S_1 = E_x^2 - E_y^2 \tag{2.5.2}$$

$$S_2 = 2E_xE_y\cos\delta \tag{2.5.3}$$

$$S_3 = 2E_xE_y\sin\delta \tag{2.5.4}$$

where δ is the phase shift between the horizontal and the vertical components of the electric field $\overrightarrow{E}$ propagating in z-direction. To see the physical interpretation of the Poincare sphere about the polarization consider the points on the sphere represented. For point M, $S_1 = S_2 = 0$. This implies from the eqs. (2.5.2) and (2.5.3) we get $E_x = E_y$ and $\delta = \pm\frac{\pi}{2}$ in which $\delta = -\frac{\pi}{2}$ represents the point M' and $\delta = \frac{\pi}{2}$ represents

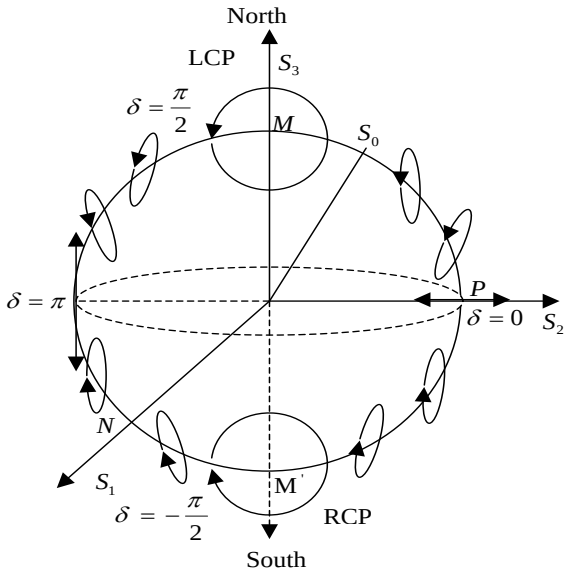

Figure 2.4: Representation of polarization of light using Poincare sphere.

the point M which are the circular polarization conditions as discussed in section 2.3. For point P, $S_1 = S_3 = 0$ and this implies from the eqs. (2.5.2) and (2.5.4) that $\sin\delta = 0$ and we get $\delta = 0$ or $\delta = \pi$ which is the condition of linearly polarized light. For point N, we have $S_2 = S_3 = 0$ which is the superposition of linear and circular polarization. but we know that the superposition of circular polarization gives the linear polarization and hence at point N we get linear polarization.

As one moves on the Poincare sphere from point M to N, the state of polarization changes from the LCP at M to elliptically polarized light of different eccentricity and finally at N we get linearly polarized light along S_1 direction and then from N to

P we will have elliptically polarized and finally RCP light and so on. The following points can be taken as the summary of this section.

1. The north and the south pole stands for the left and right circular polarization respectively.

2. Every point on the equator represents a linear polarization state; each point implies different oscillation direction.

3. The northern hemisphere excluding the north pole represents right handed elliptical and the southern hemisphere excluding the south pole represents left handed elliptical polarization respectively [19].

2.6 The Stokes Parameters and Stokes Vector

Here now different possible states of polarization of TE wave of light can also be represented by a set of four real quantities, called the Stokes parameters, each of which has the dimension of intensity [14]. In terms of cartesian components of the TE, the four Stokes parameters denoted by eqs. (2.5.1) through (2.5.4) where S_0 gives the total intensity of the light wave and therefore is always positive. S_1 can be either positive, negative, or zero depending on whether the wave has strong preference to the x linear polarization, to the y linear polarization, or to neither one of these two states, respectively. S_2, as defined above, represents the preference of the wave to either $+\frac{\pi}{4}$ or the -$\frac{\pi}{4}$ linearly polarized component. If S_2 is positive (negative) the wave has strong preference to the $+\frac{\pi}{4}(-\frac{\pi}{4})$ linear polarization. If S_2 is zero the wave has no preference to either of these two polarizations. The Stokes parameter S_3, as defined, represents the preference of the wave to either the right-handed or

to the left-handed circularly polarized component. S_3 is positive, negative, or zero dependent on the wave possessing stronger preference to the right-circular state, the left-circular state or to one of these two states respectively.

2.6.1 The Stokes Vector

The Stokes parameters of monochromatic light wave can be grouped in a 4×1 column vector [14]:

$$S = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} \tag{2.6.1}$$

called the Stokes vector of the wave. This can also be written horizontally between curly brackets and the elements separated by commas as follows.

$$S = \{S_0, S_1, S_2, S_3\} \tag{2.6.2}$$

We see that for unpolarized light, there is no preference to any particular polarizations so that $S_1 = S_2 = S_3 = 0$ and the Stokes vector assumes the form

$$S_{un} = \{S_0, 0, 0, 0\} \tag{2.6.3}$$

The totally (in view of figure 4.2) polarized light satisfies the following condition

$$S_0^2 = S_1^2 + S_2^2 + S_3^2 \tag{2.6.4}$$

and partially polarized and unpolarized light satisfies the inequality

$$S_0^2 > S_1^2 + S_2^2 + S_3^2 \tag{2.6.5}$$

An important quantity in the description of partially polarized light is the degree of polarization which is defined as the ratio of the intensity of the totally polarized component to the total intensity of the wave [14]

$$\mathcal{P} = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} \tag{2.6.6}$$

The degree of polarization $\mathcal{P}$ varies from zero for unpolarized light to unity in the case of totally polarized light and assumes intermediate (fraction) values for partially polarized light. The following properties of this Stokes polarization subspace we can readily arrive at

1. The origin, $\mathcal{P}$ =0 represents the unpolarized state.

2. Each point on the surface of the unit sphere, $\mathcal{P}$, represents a distinct totally polarized state. It is clear that the unit sphere in the Stokes subspace is the Poincare.

3. Excluding the origin, any point inside the unit sphere, $0 < \mathcal{P} < 1$, represents a partially polarized light. Points outside the unit sphere, $\mathcal{P} > 1$, don't represent any physical state of polarization.

In the next chapter we need to concentrate on the experimental part. It includes the basic principles of both RAS and TAS techniques and also consists the setup of these techniques which takes us to the main core part of this thesis.

Chapter 3

Theory and Experimental Setup of RAS and TAS

3.1 Introduction

In this chapter the basic principles of the optical techniques RAS and TAS and their application in the study of the optical properties of surfaces described. To pass through these we begin with the basic concepts. Spectroscopy is a powerful tool used in science to study a variety of materials and phenomena. Some processes involve the emission of light after a material is excited electrically or optically; these include atomic emission, fluorescence, photoluminescence and the Raman effect. In other cases, the light that is transmitted through, reflected from or absorbed by a material is measured. Absorption, Transmission, and Reflection are three types of spectroscopy from which we are interested in only two of them -reflection and transmission.

Reflectance anisotropy spectroscopy (RAS), also referred to as reflectance difference spectroscopy (RDS), is a powerful tool for characterizing the surfaces of crystalline materials [29]. It is a non-destructive optical probe of surfaces that is capable of operating within a wide range of environments [10]. Knowledge of the dynamics of the state of polarization of light propagating in an anisotropic media has become an important issue in the development of sensing techniques (like RAS) based on polarization properties of light [30]. RAS has been applied to various semiconductor surfaces [10, 29].

RAS may be used to measure anisotropy in many systems. For example, anisotropic shaped entities of an isotropic material that are aligned in a common direction on an isotropic substrate may generate an RAS signal [10]. RAS is sensitive to the surface electronic structure in the region of the Fermi level (E_F), and features in RA spectra of clean semiconductor and metal surfaces can be identified with electronic transitions between surface states. RAS measures the difference in reflectance (Δr) of normal incidence plane-polarized light between two orthogonal directions in the surface plane (x, y) normalized to the mean reflectance (r) [10, 29]:

$$\frac{\Delta r}{r} = 2\frac{(r_x - r_y)}{r_x + r_y} \tag{3.1.1}$$

where the reflectance r are complex Fresnel reflection amplitudes.

The advantage of this purely photonic probe of surfaces is the ability to operate in environments consisting of any optically transparent medium [10]. This capability opens up the study of surfaces in catalytic and corrosive environments at high pressure and at the solid/liquid interface.

Transmission anisotropy spectroscopy (TAS) also termed as birefringence is also

a powerful tool for characterizing an optical anisotropy of transparent samples. Experiments show that although most of the incident light passes through transparent samples, the reflected beam can be more sensitive to the optical anisotropy of thin films than the transmitted beam [10]. The same equation is used for the determination of the optical anisotropy but it measures the difference in transmittance Δt instead of reflectance of normal incidence plane-polarized light between two orthogonal directions in the surface plane (x, y) normalized to the mean reflectance (t).

3.2 Principles of RAS

The measurement of reflection anisotropy requires an accurate method of determining the polarization state of the light reflected from the surface. Thus, RAS is a form of 'polarimetry' and shears a common heritage with techniques such as spectroscopic ellipsometry (SE), circular dichroism (CD), optical rotary dispersion (ORD), and linear birefringence (LB) [10].

3.3 Principles of TAS

For transparent samples the possibility of measuring optical anisotropy in transmission arises. To achieve this one simply needs to place the reflection section of an RA spectrometer behind the sample (although historically experimental studies of this type is termed as birefringence rather than transmission anisotropy studies). Likewise the RAS technique, this technique also requires an accurate method of determining the polarization state of the light transmitted through the sample. Transmission anisotropy instrumentation has been developed independently of RAS

and in some cases predates it [10]. The simple schema is as shown in the fig. 3.3 below.

Modulation spectroscopy can be conveniently and advantageously implemented-both intensity and phase by modulating the external parameters and using the lock-in amplifier to detect the resulting signals in optical response. But from these two we have only used the intensity modulation. However, let's see both one by one below.

3.3.1 Intensity Modulation

If linearly polarized light is incident on a sample rotating with angular frequency ω, the intensity of the reflected by the sample is therefore [10]

$$I \propto R - \frac{\Delta R}{2}\cos(2\omega t) \tag{3.3.1}$$

where R and ΔR represent, respectively, the average and the anisotropy of the reflected intensities:

$$R = \frac{|r_x|^2 + |r_y|^2}{2}\ , \qquad \Delta R = |r_x|^2 - |r_y|^2 \tag{3.3.2}$$

The ratio of the oscillatory ($I_{2\omega}$) and time-independent (I_0) contributions to the transmitted or reflected intensity is a direct measure of $\frac{\Delta R}{R}$:

$$\frac{I_{2\omega}}{I_o} = \frac{1}{2\sqrt{2}}\frac{\Delta R}{R} \tag{3.3.3}$$

In the same way we can use the equations for the transmitted light in such a way that instead of R we use Tas discussed earlier.

The main disadvantage of rotating sample spectrometer is simply that it isn't always convenient or even possible to rotate the sample for in situ studies. Therefore it is worth mentioning that it is convenient to rotate the polarizer instead of the sample. This is why we are dealing with the rotating polarizer RAS and TAS techniques.

3.3.2 Phase Modulation

The photoelastic modulation (PEM) is essentially a dynamic wave plate whose effect is that the light polarized perpendicular to the modulation axis is unaffected by PEM while light polarized along the modulation axis undergoes a retardation, Γ [10]. Thus a sinusoidal electric field introduces an oscillatory retardation:

$$\Gamma = \Gamma_0 \sin(\omega t) \tag{3.3.4}$$

Here the significant retardation occurs when the crystal is driven at its resonance frequency.

The RA is most fully expressed as $\frac{\Delta r}{r}$, a complex quantity, while $\frac{\Delta R}{R}$ is real and, for small anisotropies, approximately equal to $2Re\{\frac{\Delta r}{r}\}$ [10]. Thus by modulating phase rather than intensity, measurement of both the real and imaginary part of $\frac{\Delta r}{r}$ becomes possible (don't forget that these concepts also hold true for transmission). With the aid of these modulations, surface induced RAS/TAS signals have magnitude typically of the order of less than or equal to 10^{-3} [10], and hence the AC signals are 100 times smaller than the corresponding DC level.

3.4 The Rotating Polarizer RAS and TAS Setup

The overall setup (shown in the fig. 3.1 below) consist of different parts. The HeNe laser (PHYWE) source; Chopper (Stanford Research Systems, Model: SR540) to reduce the noise; Half-wave plate (Melles Griot, Model: 02WRM021) to rotate the polarization; Sample; Analyzer to analyze the light reflected from/or transmitted through the sample; Photo detector to detect the light flux impinging on it; Lock-in-Amplifier (Stanford Research Systems, Model: SR830) to amplify the signal and the

Stepper motor controller (Melles Griot, Model: 17BSC002) to control the half-wave plate and the analyzer.

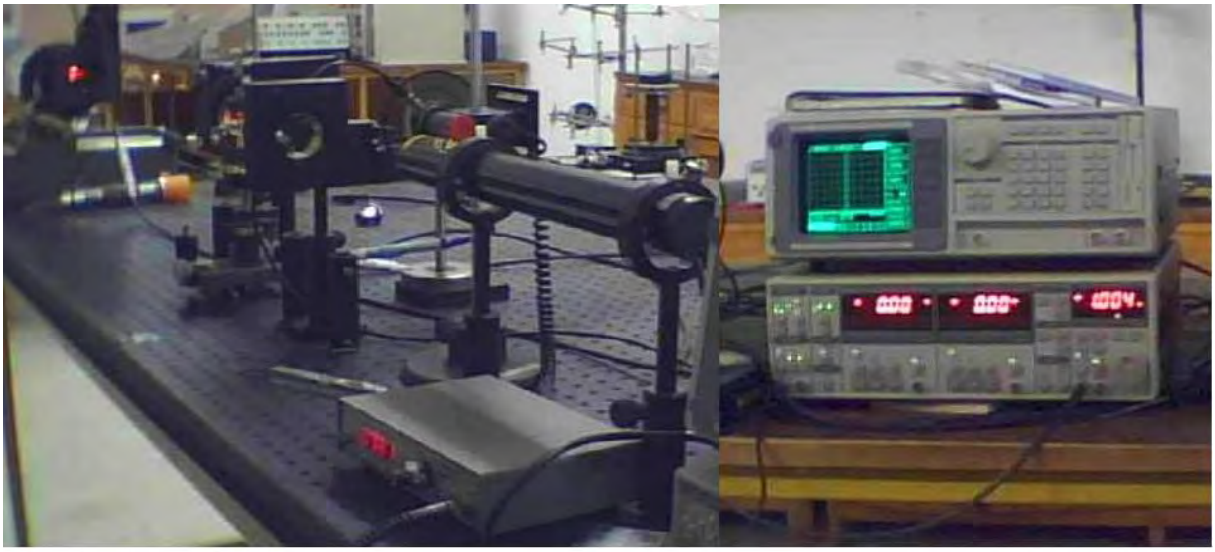

Figure 3.1: RAS setup with lock-in amplifier and spectrum analyzer.

This is the overview of the setup but the simple schema can be shown as follows specifically for both techniquees. It is necessary here to give a brief description of the optical devices used in this setup.

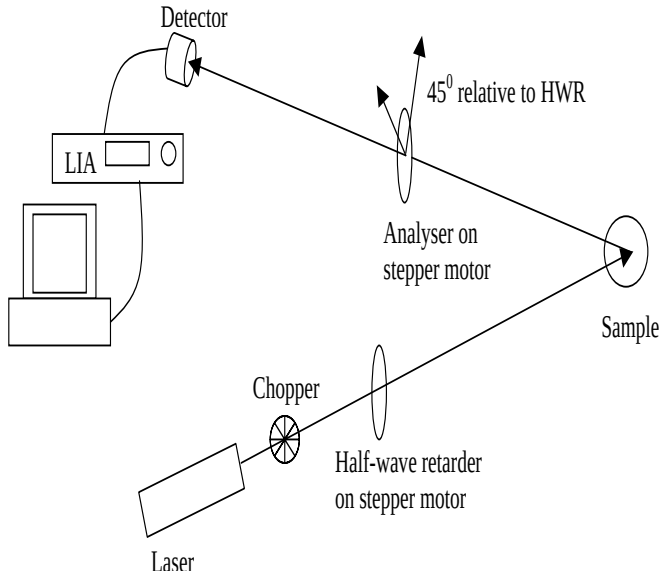

Figure 3.2: Rotating analyzer-polarizer setup of RAS.

3.5 Methodology

The schematic set-up of both RAS and TAS spectrometry is shown in the figures (3.2) and (3.3) above. A well collimated beam of monochromatic linearly polarized light from a 5 mv HeNe laser source (PHYWE), is passed through half-wave plate (HWP) ($\frac{\lambda}{2}$)(Melles Griot, Model: 02WRM021) and chopper (Stanford Research Systems, Model: SR540). Light emergent from the HWP is reflected at a near normal incidence ($\approx 5^0$ off normal) from the fixed sample and some part is transmitted through the specular surface of a fixed sample. In this experiment, since the interest is only on the light coming along the perpendicular to the optical axis, the reflected as well as the transmitted light must pass through analyzing section of the instrument that consists analyzer, its axis is making 45^0 from the HWP, followed

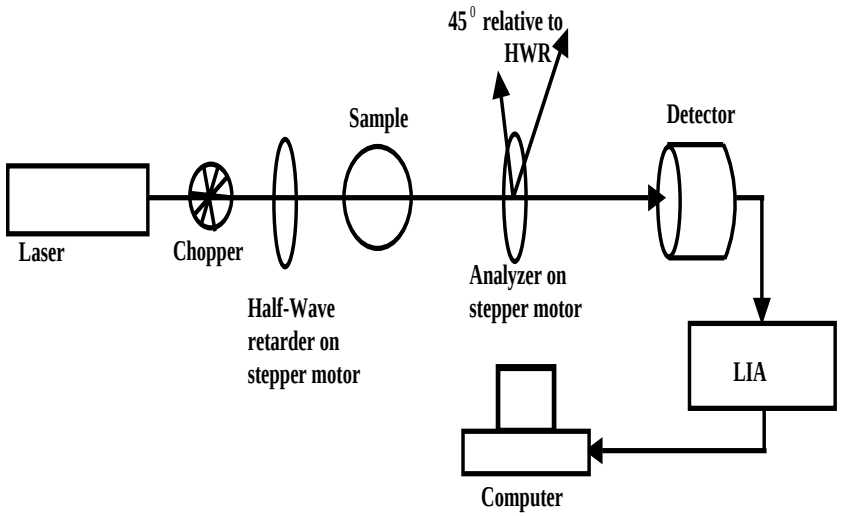

Figure 3.3: Rotating analyzer-polarizer setup of TAS.

by a photodetector. Thus, the light received by the photodetector is amplified for measurement by a lock-in amplifier (LIA) (Stanford research systems, Model SR830). Therefore, both RAS and TAS measurment have been taken simultaneously at the same time with the help of stepper motor (Melles Groit, Model: 17BSC002). Since LIA is interfaced with the computer having a LabView software, the data will be easily accessed.

Chapter 4

Data Analysis and Interpretation

As discussed in the previous chapters, to apply both the RAS and TAS techniques it is mandatory first to determine the optical axis of the sample under the study. When light of arbitrary polarization propagates through anisotropic crystal, it can be considered to consist two independent waves that are polarized orthogonally with one another and traveling with different phase velocities. But those two phase velocities may have the same value, when they are propagating in the direction of optical axis of the crystal. To see special nature of the transmitted as well as reflected light we first have a look at some properties of KTP crystal. It is known that KTP crystal is a biaxial crystal and has three principal refractive indices n_1, n_2 and n_3 which are all different in magnitude. When light enters the optical axis of anisotropic crystals, it behaves in a manner similar to the interaction with isotropic crystals, and passing through at a single velocity. However, when light enters a non-equivalent axis, it is transmitted or reflected in to two rays each polarized with vibration directions oriented at right angles to one another, and traveling at different velocities. From this phenomenon one can easily realize that, the intensity of transmitted or reflected light along the optical axis is some what intense than the reflected or transmitted light

from other axis. To start with let's see the previous results [35] in order to compare it with our results (see fig. 4.1)

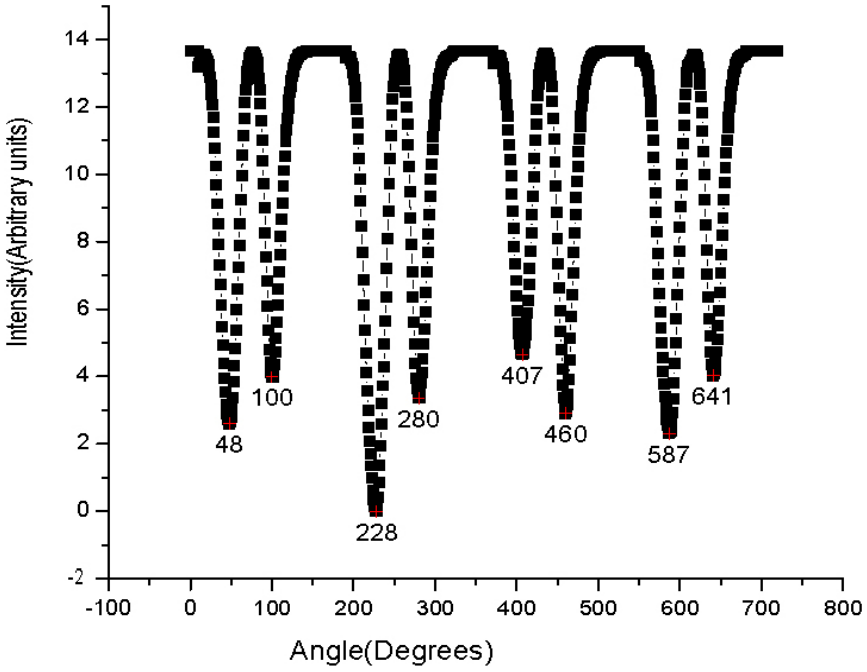

Figure 4.1: Reflection of light as the function of angle of rotation.

When we compare fig. 4.1 with fig. 4.2 (our present work result), there is a sharp drop in intensity and the width between the dropping intensities is very small in fig. 4.2 as compared to that of the fig. 4.1. This decease in gap between the intensities has important application in band width. based on this background, we extended our knowledge to further application which is the determination of degree of anisotropy with TAS technique and also our new research part. The data collected for TAS is shown in the fig. 4.3.

From the figures 4.2 and 4.3, the intensities represents minimum values for reflection and maximum values for transmission and it is possible to realize that the

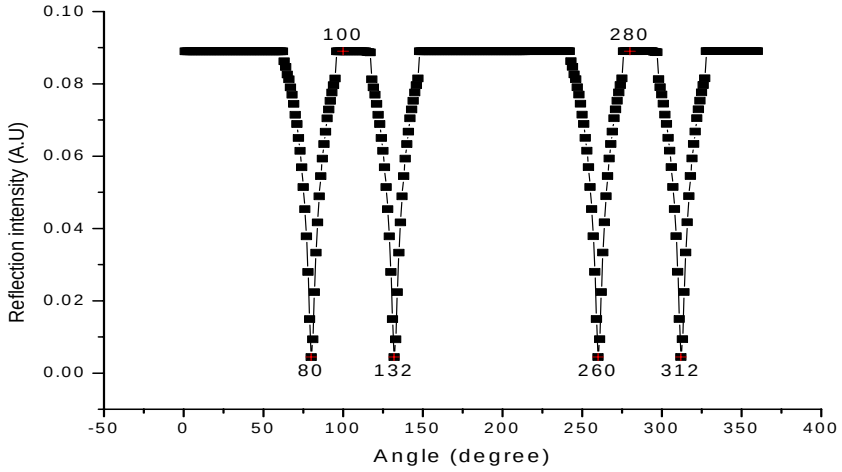

Figure 4.2: Reflected light as a function of angle of rotation using fixed sample RAS.

crystal has C_2 symmetry. Furthermore, it is expected that the light reflected from as well as transmitted through the optical axis may has minimum (for reflection) and maximum (for transmission) value comparing to the intensity reflected from or transmitted through the other part of the material.

From the figures 4.2 and 4.3 the minima and maxima respectively show that there is a two-fold C_2 symmetries (the difference between 80^0 and 260^0, and 132^0 and 312^0 is 180^0). By considering the above argument the two optical axes of the KTP crystal can be found along 80^0 and 260^0 and along 132^0 and 312^0.

4.1 Data Analysis and Interpretation

Since the optical axes of the KTP crystal have been determined, it is now possible to calculate the degree of anisotropy by using the intensity modulation method as we have mentioned in the preceding chapter. This is done by the principle that

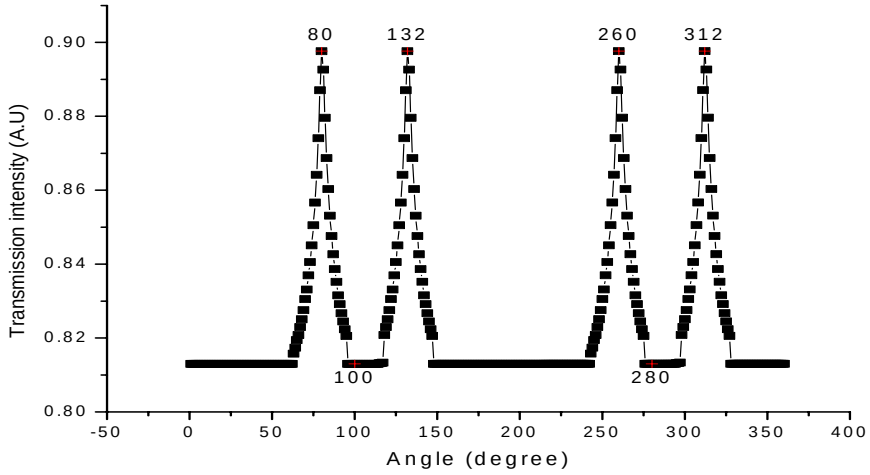

Figure 4.3: Transmitted light as a function of angle of rotation using fixed sample TAS.

the polarization of the incident light was adjusted in such a way that it is parallel to thee selected optical axis of the crystal. Then the analyzer was rotated 360^0 to take those two important parameters, i.e, the transmitted light along the optical axis (t_x) and the reflected light along the optical axis (r_x) which are found when the optical axis makes $+45^0$ from the incident light and the transmitted or reflected light perpendicular to the optical axis (t_y) or (r_y) which are found when the optical axis makes -45^0 from the incident light. The values are summarized in the tables (4.1) and (4.2) below.

Degree	$\lvert r_x\rvert^2 \times 10^{-3}$	Degree	$\lvert r_y\rvert^2 \times 10^{-3}$
55	7.921	25	7.893
56	7.921	24	7.878
57	7.921	23	7.864

Table 4.1: Values of r_x and r_y.

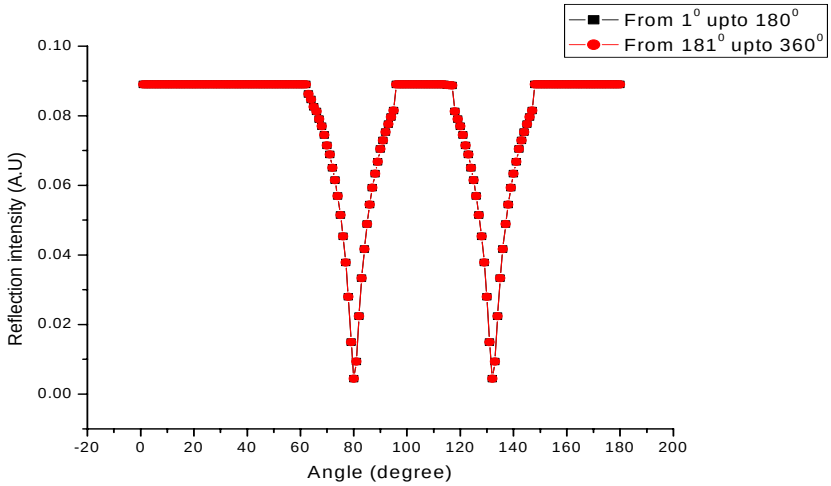

Figure 4.4: Symmetry along 180^0 which shows C_2 symmetry for RAS.

Degree	$\|t_x\|^2$	Degree	$\|t_y\|^2$
55	0.660969	25	0.661229
56	0.660969	24	0.661359
57	0.609690	23	0.661489

Table 4.2: Values of t_x and t_y.

Using eqn. (3.1.1) the magnitude of the degree of anisotropy of the KTP crystal is computed. Here the average value obtained from the data is 3.597×10^{-3} and 3.9×10^{-4} for reflection and transmission respectively. It can also realized that, how much the selected optical axes is symmetry by considering fig. 4.4 and fig. 4.5, which shows that fitting of the values of intensity from 0^0 to 180^0 and the values of intensity from 181^0 to 360^0. In addition, from the figures 4.2 and 4.3 we can find that the difference in angle between the optical axes. As can be seen, there are two optical axes and thus $\Delta\theta_3$ should be equal to $\Delta\theta_1$ and also $\Delta\theta_4$ should be equal to $\Delta\theta_2$. Fig.

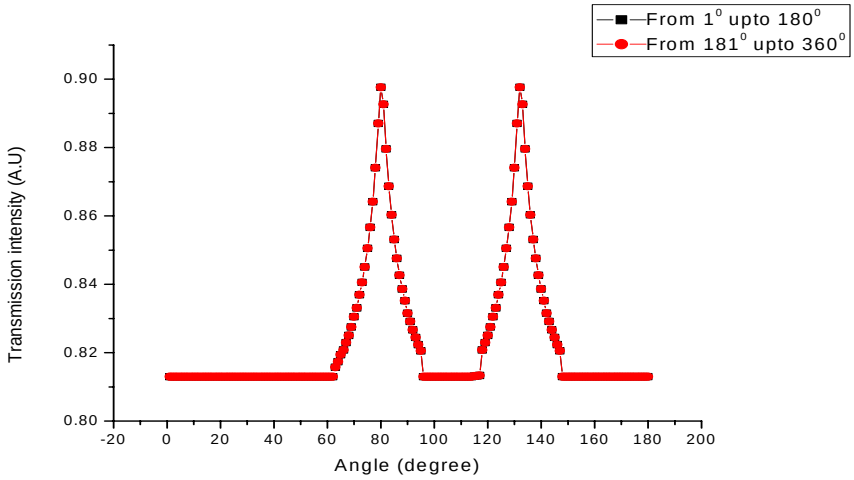

Figure 4.5: Symmetry along 180^0 which shows C_2 symmetry for TAS.

4.6 explains this concept. Thus

$$\Delta\theta_3 = \Delta\theta_1 = 312^0 - 260^0 = 52^0, \Delta\theta_4 = \Delta\theta_2 = 260^0 - 132^0 = 128^0 \qquad (4.1.1)$$

It is also important to see that if one undergoes the coupled task of TAS and RAS spectrometry, it is worth mentioning to consider the absorption correction. That is once both reflectance as well as transmittance are known it is easy to see how much the sample under the study absorbs light. In our case as can be seen from the fig. 4.7, the absorption is very small as compared to the quantities measured. On the other hand transmission is high and we can say that its reflection is in between the absorption and transmission for the wavelength (632.8 nm) we used in our experiment.

We know that for absorbing media the refractive index is no longer real, instead, it is complex. The imaginary part is responsible for the loss of energy due to the absorption of the medium. In the same way the amplitude of the light passing through

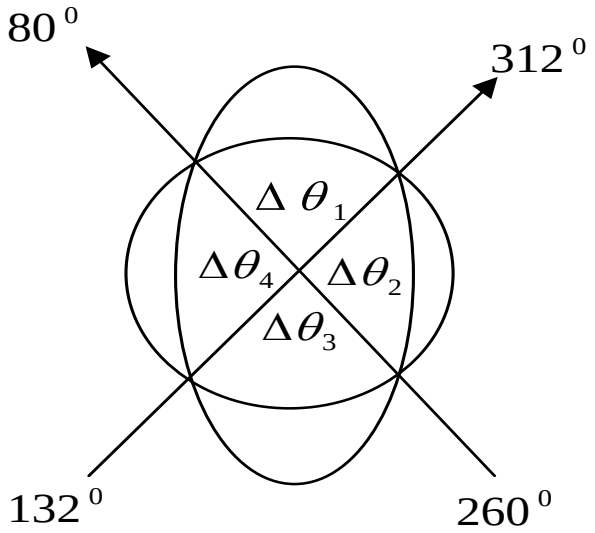

Figure 4.6: Optical axes of KTP crystal.

the medium isn't constant but it decreases. Therefore, from the fig. 4.7 we see that the absorption of KTP crystal is very small which also leads to fact that the imaginary part of this crystal is very small. When the imaginary part of a medium is large the sum of the reflectance and the transmittance is lowered because the loss of energy becomes large.

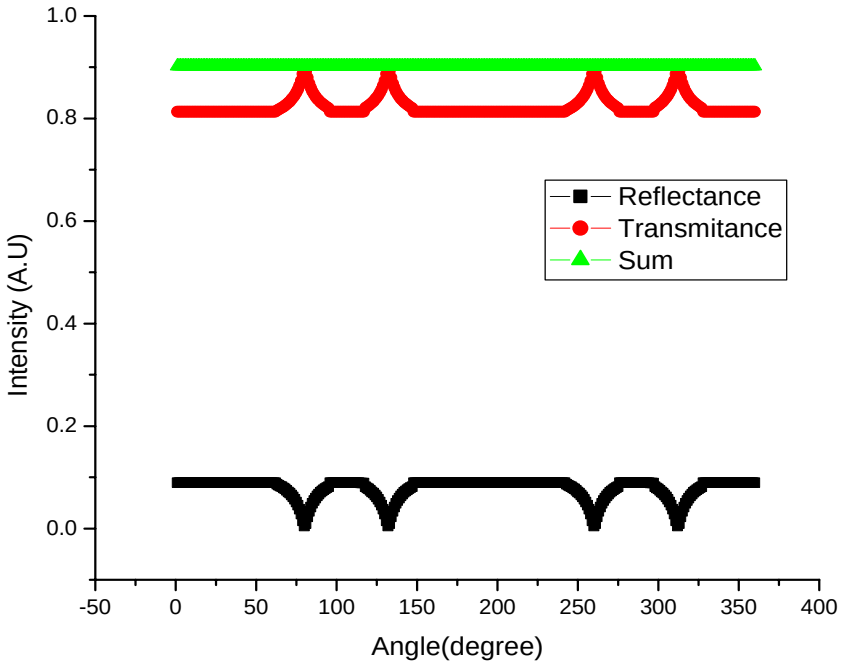

Figure 4.7: Combination of reflectance, transmittance, and their sum intensities as a function of angle of rotation for the fixed sample technique.

Chapter 5

Conclusion

The transmitted and reflected light results from the interaction of light with matter may contain a number of information about the material, especially when the transmitted and the reflected light is analyzed by using sensitive optical techniques like TAS and RAS. In this work the non-destructive and sensitive optical techniques TAS and RAS are applied to calculate the degree of anisotropy of KTP crystal. Since KTP crystal has C_2 symmetry, by considering only one optical axis along 80^0 the magnitude of the degree of anisotropy of the KTP crystal is obtained to be 3.597×10^{-3} for reflection and 3.9×10^{-4} for transmission respectively. In this work, the magnitude of the degree of anisotropy is of the order of 10^{-3} for reflection and 10^{-4} for transmission which is in agreement with the results found from the experiments [10]. It can also be said that not only the RAS technique but also the TAS technique is capable for the determination of anisotropy of crystals. It can also be said that this coupled work helps the determination of absorption correction. Thus, the absorption loss of KTP crystal is very small. But transmission is large and the reflection is in between the transmission and the absorption.

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