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Infrared Material Property Measurements with Polarimetry and Spectropolarimetry (Paper)

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Polarimetry; Spectropolarimetry; Material properties 15

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Infrared material property measurements with polarimetry and spectropolarimetry

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ABSTRACT

Polarimetry and spectropolarimetry are optical measurement techniques which use polarized light to obtain electrooptical material property information. These techniques are being used to make measurements on infrared electrooptical materials. Infrared materials of interest are those which find use in two dimensional modulators, i.e. optical processing applications and infrared scene projectors. Polarimetry measurements are made at one infrared wavelength at a time using laser sources. Spectropolarimetry measurements are made over an entire infrared spectral region, e.g. 2 to 14 μm. A Fourier transform infrared spectrometer is the source of radiation in this case. The Mueller matrix formulation can be used to describe the polarized light and its interaction with the sample and measurement system optics. A Mueller matrix of the sample can then be obtained from a series of measurements with different input polarization states. The sample Mueller matrix contains information on the polarization properties. Electric fields are imposed on the sample as optical measurements are made. Fundamental constants associated with the sample material can be derived from the measured Mueller matrix elements.

1. INTRODUCTION

Bulk properties of transmissive materials can be measured using the experimental methods of polarimetry and spectropolarimetry. We show in this paper how these techniques are implemented and how fundamental properties may be derived from the measurements.

The Mueller matrix formalism is used to process the polarimetric information. A mathematical background on the Mueller formalism and crystal physics are presented. We then describe details of an infrared laser polarimeter and an infrared spectropolarimeter. Derivation of electrooptic tensor coefficients directly from the measured Mueller matrix is discussed.

2. MATHEMATICAL BACKGROUND

The mathematics for describing the polarization modulation when a beam of optical radiation is passed through a material is presented here. The mathematical formalism for polarized light is discussed first, followed by the mathematics necessary to describe crystal physics.

Two algebraic systems have been developed for the solution of polarization problems in optics, the Jones formalism and the Mueller formalism. The Jones formalism is a natural consequence of the mathematical phase and amplitude description of light. The Mueller formalism comes from experimental considerations of the intensity measurements of polarized light. The Mueller formalism is used for the present work and is described below. Other treatments of the Mueller and Jones formalisms can be found in Gerrard and Burch[1], Azzam and Bashara[2], Clarke and Grainger[3], and Theocaris and Gdoutos[4].

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2.1 The Mueller formalism

The Mueller formalism owes its name to Hans Mueller, who built upon the work of Stokes[5], Soleillet[6], and Perrin[7] to formalize polarization calculations based on intensity. This work was also done during the 1940's but originally appeared in a now declassified report (Mueller[8]) and in a course of lectures at MIT in 1945-1946.

The Mueller formalism uses the Stokes vector, a four element real vector, to represent the polarization state. This vector is often represented as

$$S = \begin{pmatrix} s_0 \\ s_1 \\ s_2 \\ s_3 \end{pmatrix} \quad (1)$$

where s_0 is related to intensity, s_1 to preference for horizontal polarization, s_2 to preference for 45° polarization, and s_3 to preference for right circular polarization. The inequality

$$s_0^2 \geq s_1^2 + s_2^2 + s_3^2 \quad (2)$$

holds, indicating that some part of the light intensity may be in a depolarized state.

The Mueller matrix is a 4x4 matrix representing optical elements or an optical system, i.e.

$$M = \begin{pmatrix} m_{11} & m_{12} & m_{13} & m_{14} \\ m_{21} & m_{22} & m_{23} & m_{24} \\ m_{31} & m_{32} & m_{33} & m_{34} \\ m_{41} & m_{42} & m_{43} & m_{44} \end{pmatrix} \quad (3)$$

where the m_{ij} are real numbers.

The Stokes vector describes completely the polarization state of the light in terms of intensity. The Mueller matrix is an empirical entity for a given ray path through an optical element or system, taking the form required to produce the correct output polarization state. There is redundancy built into the Mueller matrix, since only seven of its elements are independent[9] if there is no depolarization in the optical system. In the most general case, the Mueller matrix can have sixteen independent elements[10], however not every 4x4 Mueller matrix is a physically realizable polarizing element. The latter aspect of the Mueller formalism is discussed by Cloude[11,12].

Matrix products represent the action of sequential optical elements on a light beam. In the Mueller formalism this is represented by

$$M_S = M_K \ldots M_2 M_1 = \prod_{k=K}^{1} M_k \quad (4)$$

where M_S is the system Mueller matrix and the M_k are the element matrices. The element matrices are multiplied on the left in the order that the light encounters the elements. The Mueller matrices are in general functions of wavelength and their own orientation θ with respect to some arbitrary reference, i.e. $M = M(\lambda, \theta)$.

The Mueller matrix can, unlike the Jones matrix, represent a depolarizer or scatterer, and van de Hulst[10] describes this property. The Mueller matrix formalism has two advantages for experimental work over the Jones formalism. The intensity is represented explicitly in the Mueller formalism, and scattering can be included in the calculations.

2.2 Crystal physics

In general, crystals are anisotropic with respect to the propagation of electromagnetic waves in the presence of static electric fields. The relationship between the displacement and the field is (MKS units)

$$D_i = \epsilon_{ij} E_j \tag{5}$$

where ϵ_{ij} is the dielectric or permittivity tensor[13]. The impermeability tensor η_{ij} is defined as

$$\eta_{ij} = \epsilon_o (\epsilon^{-1})_{ij} \tag{6}$$

where ϵ^{-1} is the inverse of the dielectric tensor and ϵ_o is the permittivity of free space. The principal indices of refraction n_x, n_y, and n_z are related to the principal values of the impermeability tensor and the principal values of the permittivity tensor by

$$\frac{1}{n_x^2} = \eta_{ii} = \frac{\epsilon_o}{\epsilon_{ii}}, \qquad \frac{1}{n_y^2} = \eta_{jj} = \frac{\epsilon_o}{\epsilon_{jj}}, \qquad \frac{1}{n_z^2} = \eta_{kk} = \frac{\epsilon_o}{\epsilon_{kk}}. \tag{7}$$

The electrooptic coefficients are defined by the equation

$$\eta_{ij}(E) - \eta_{ij}(0) \equiv \Delta\eta_{ij} = r_{ijk} E_k + s_{ijkl} E_k E_l + O(E^n) \tag{8}$$

where η_{ij} is a function of the applied field E, r_{ijk} are the linear electrooptic tensor or Pockels electrooptic coefficients, and the s_{ijkl} are the quadratic or Kerr electrooptic coefficients. Terms higher than quadratic are small and are neglected.

The linear electrooptic tensor is of third rank with 3^3 elements; however, symmetry enables a contraction. If the medium is lossless and optically inactive, ϵ_{ij} is a symmetric tensor, η_{ij} is a symmetric tensor, r_{ijk} has symmetry where coefficients with permuted first and second indices are equal, and s_{ijkl} has symmetry where coefficients with permuted first and second indices are equal and coefficients with third and fourth coefficients are equal. The linear electrooptic coefficients are assigned two indices so that they are r_{lk} where l runs from 1 to 6 and k runs from 1 to 3. The quadratic coefficients are assigned two indices so that they become s_{ij} where i runs from 1 to 6 and j runs from 1 to 6. For a given crystal symmetry class, the form of the electrooptic tensor is available from standard tables[13].

Light propagating in anisotropic materials experiences refractive index and phase velocity dependent upon the propagation direction and polarization state. The refractive index for propagation in some direction $\vec{k} = x\hat{i} + y\hat{j} + z\hat{k}$ can be obtained from the index ellipsoid. In the principal coordinate system the index ellipsoid is given by

$$\frac{x^2}{n_x^2}+\frac{y^2}{n_y^2}+\frac{z^2}{n_z^2}=1 \tag{9}$$

in the absence of an applied electric field. The length of the semimajor and semiminor axes of the ellipse formed by the intersection of this index ellipsoid and a plane normal to the propagation direction (both geometric objects being centered at the origin) are the two principal indices of refraction for that propagation direction. If two of the three indices of the index ellipsoid are equal, the crystal is defined to be uniaxial and the equation for the index ellipsoid is

$$\frac{x^2}{n_o^2}+\frac{y^2}{n_o^2}+\frac{z^2}{n_e^2}=1\,. \tag{10}$$

Where there are three distinct principal indices, the crystal is defined as biaxial.

The alteration of the index ellipsoid of a crystal with application of an electric and/or magnetic field is useful for the modulation of polarization state. The equation for the index ellipsoid of a crystal in an electric field is

$$\eta_{ij}(E)x_i x_j = 1 \tag{11}$$

or

$$(\eta_{ij}(0)+\Delta\eta_{ij})x_i x_j = 1\,. \tag{12}$$

This equation can be written as

$$\begin{aligned}&x^2\left(\frac{1}{n_x^2}+\Delta\left(\frac{1}{n}\right)_i^2\right)+y^2\left(\frac{1}{n_y^2}+\Delta\left(\frac{1}{n}\right)_i^2\right)+z^2\left(\frac{1}{n_z^2}+\Delta\left(\frac{1}{n}\right)_i^2\right)\\&+2yz\left(\Delta\left(\frac{1}{n}\right)_i^2\right)+2xz\left(\Delta\left(\frac{1}{n}\right)_i^2\right)+2xy\left(\Delta\left(\frac{1}{n}\right)_i^2\right)=1\end{aligned} \tag{13}$$

or

$$\begin{aligned}&x^2\left(\frac{1}{n_x^2}+r_{1k}E_k+s_{1k}E_k^2+2s_{14}E_2E_3+2s_{15}E_3E_1+2s_{16}E_1E_2\right)\\&+y^2\left(\frac{1}{n_y^2}+r_{2k}E_k+s_{2k}E_k^2+2s_{24}E_2E_3+2s_{25}E_3E_1+2s_{26}E_1E_2\right)\\&+z^2\left(\frac{1}{n_z^2}+r_{3k}E_k+s_{3k}E_k^2+2s_{34}E_2E_3+2s_{35}E_3E_1+2s_{36}E_1E_2\right)\\&+2yz(r_{4k}E_k s_{4k}E_k^2+2s_{44}E_2E_3+2s_{45}E_3E_1+2s_{46}E_1E_2)\\&+2zx(r_{5k}E_k+s_{5k}E_k^2+2s_{55}E_3E_1+2s_{56}E_1E_2)\\&+2xy(r_{6k}E_k+s_{6k}E_k^2+2s_{64}E_2E_3+2s_{65}E_3E_1+2s_{66}E_1E_2)=1\end{aligned} \tag{14}$$

where the E_k are components of the electric field along the principal axes and repeated indices are summed.

If the quadratic coefficients are assumed to be small and only the linear coefficients are retained, then

$$\Delta\left(\frac{1}{n}\right)^2_i = \sum_{k=1}^{3} r_{ik} E_k \tag{15}$$

and k = 1,2,3 corresponds to the principal axes x, y, and z. For a uniaxial crystal in a field along z, the index ellipsoid becomes

$$\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} + 2r_{63}E_z xy = 1. \tag{16}$$

A coordinate rotation can be done to obtain the major axes of the new ellipsoid. In the present example, this yields the new ellipsoid

$$\left(\frac{1}{n_o^2} + r_{63}E_z\right)x'^2 + \left(\frac{1}{n_o^2} - r_{63}E_z\right)y'^2 + \frac{z^2}{n_e^2} = 1 \tag{17}$$

The new and old z axes are the same, but the x' and y' axes have rotated around the z axis.

The refractive indices along the new x and y axes are:

$$\begin{aligned} n_x' &= n_o - \frac{1}{2}n_o^3 r_{63} E_z \\ n_y' &= n_o + \frac{1}{2}n_o^3 r_{63} E_z. \end{aligned} \tag{18}$$

For light linearly polarized at 45 degrees, the x and y components experience the different refractive indices n_x' and n_y'. The birefringence is defined as the index difference $n_y' - n_x'$. Since the phase velocities of the x and y are different, there is a phase retardation Γ (in radians) between the x and y components of E given by

$$\Gamma = \frac{\omega}{c}(n_y' - n_x')l = \frac{2\pi}{\lambda}n_o^3 r_{63} E_z l \tag{19}$$

where l is the path length of light in the crystal. The electric field of the incident light beam is:

$$\vec{E} = \frac{1}{\sqrt{2}}E(\hat{x} + \hat{y}) \tag{20}$$

After transmission through the crystal, the electric field is:

$$\frac{1}{\sqrt{2}}E(e^{i\Gamma/2}\hat{x}' + e^{-i\Gamma/2}\hat{y}') \tag{21}$$

If the path length and birefringence are selected such that $\Gamma = \pi$, the modulated crystal acts as a half wave linear retarder and the transmitted light has field components

$$\frac{1}{\sqrt{2}}E(e^{i\pi/2}\hat{x}' + e^{-i\pi/2}\hat{y}') = \frac{1}{\sqrt{2}}E(e^{i\pi/2}\hat{x}' - e^{i\pi/2}\hat{y}')$$
$$= E\frac{e^{i\pi/2}}{\sqrt{2}}(\hat{x}' - \hat{y}'). \quad (22)$$

The axis of linear polarization of the incident beam has been rotated by 90 degrees by the phase retardation of π radians or one-half wavelength. The incident linear polarization state has been rotated into the orthogonal polarization state. An analyzer at the output end of the crystal aligned with the incident (or unmodulated) plane of polarization will block the modulated beam. For an arbitrary applied voltage producing a phase retardation of Γ the analyzer transmits a fractional intensity $\cos^2\Gamma$. This is the principle of the Pockels cell.

3. POLARIMETRY

Polarimetry, or transmission ellipsometry, is the branch of ellipsometry concerned with measuring the polarization state of a light beam and deducing the polarizing properties of bulk materials. A polarimeter is an optical instrument used for the determination of the polarization state of a light beam. Given the knowledge of how the polarimeter itself acts on the light, the polarization change produced by inserting a bulk sample into the polarimeter may be determined. Implementation of polarimetry can be identical to ellipsometry except that the optical path goes completely through the sample.

The infrared laser polarimeter described here is designed to be operated over the 3 to 14 μm spectral region using laser sources at several wavelengths. This spectral region is of great interest for evaluation of materials used as elements of optical processing systems or thermal imaging systems.

The optical configuration used is called the dual rotating compensator ellipsometer when it is used in ellipsometry. The data processing prescription which accompanies these optics has been described by Azzam[14] and by Hauge[15]. This polarimeter uses two fixed polarizers and two rotating waveplates in a configuration discussed below. Mueller matrix elements and Stokes vectors are used here to represent the polarization elements and polarized light, respectively. The Mueller matrix formulation is used because it is preferable for experimental work where scattering and depolarization measurements are required. Two waveplates are rotated at different but harmonic rates and a modulation of the detected intensity results. The Mueller matrix of the sample is found through a relationship between the Fourier coefficients of a series representing the modulation and the elements of the sample matrix.

Figure 1 shows a block diagram of the polarimeter. The polarimeter has five sections, the laser source, the polarizing optics, the sample, the analyzing optics, and the detector.

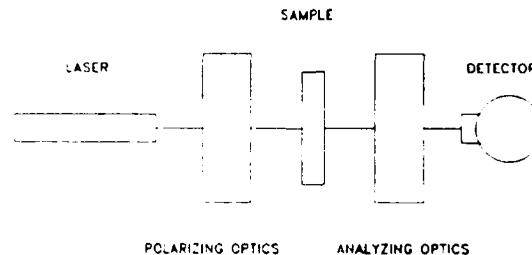

Figure 1: Polarimeter block diagram

The polarizing optics consist of a polarizer and a quarter wave plate both mounted in computer controlled rotating stages. The sample region is followed by the analyzing optics which consist of a quarter wave plate in a computer controlled rotating stage followed by a polarizer. Polarization sensitivity of the detector is not important since the orientation of the final polarizer does not change.

Figure 2 shows the instrument in more detail. This initial work has been done with a CO_2 laser. The beam is chopped at about 1 kHz. A beamsplitter is used to divert a fraction of the beam to monitor the power output of the laser. The remaining beam then passes through the polarimeter elements including the sample, and is focused with a ZnSe lens onto a HgCdTe detector.

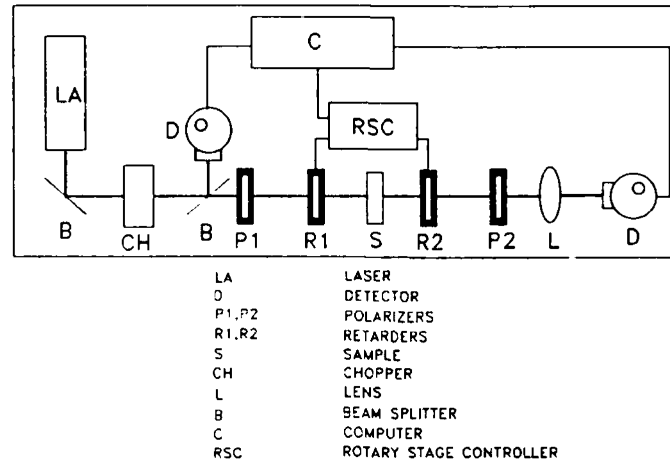

Figure 2: Polarimeter optics diagram

This polarimeter measures a chopped signal which is modulated by rotating the polarizing optical elements. The elements of the Mueller matrix can be encoded on the modulated signal. The output signal is then Fourier analyzed to determine the Mueller matrix elements. This particular implementation, due to Azzam[14], uses two aligned and fixed linear polarizers and two rotating quarter wave retarders as shown in Figure 3.

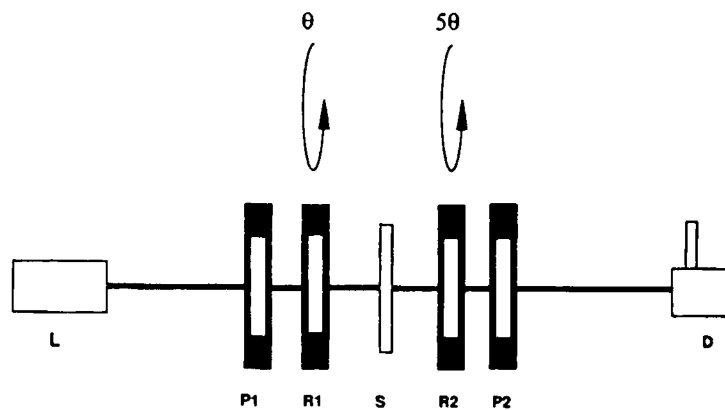

Figure 3: Polarizing elements and rotation rates

The second retarder is rotated at a rate of five times that of the first. This generates twelve harmonic frequencies in the Fourier spectrum as shown below.

The Mueller matrix for the system is

$$\mathbf{L}_2\, \mathbf{R}_2(\Theta)\, \mathbf{M}\, \mathbf{R}_1(\Theta)\, \mathbf{L}_1 \tag{23}$$

where **L** indicates a linear polarizer, **R** indicates a linear retarder, and **M** is the sample. Mueller matrices are then substituted for: a linear retarder with quarter wave retardation and a fast axis at 0° for $\mathbf{R}_1$ and $\mathbf{R}_2$; a horizontal linear polarizer $\mathbf{L}_2$; a linear horizontal polarizer for $\mathbf{L}_1$; and, a sample for **M**. The detected intensity is given by

$$I = c\,\mathbf{A}\,\mathbf{M}\,\mathbf{P} \tag{24}$$

where $\mathbf{P} = \mathbf{R}_1\, \mathbf{L}_1\, \mathbf{S}$ is the Stokes vector of light leaving the polarizing optics with **S** being the Stokes vector of the light from the source, $\mathbf{A} = \mathbf{L}_2\, \mathbf{R}_2$ is the Mueller matrix of the analyzing optics, **M** is the Mueller matrix of the sample, and c is a proportionality constant obtained from the absolute intensity. Explicitly,

$$I = c \sum_{i,j=1}^{4} a_i p_j m_{ij} \tag{25}$$

or

$$I = c \sum_{i,j=1}^{4} \mu_{ij} m_{ij} \tag{26}$$

where the a_i are the elements of **A**, the p_j are the elements of **P**, the m_{ij} are the elements of the Mueller matrix **M**, and where

$$\mu_{ij} = a_i p_j . \tag{27}$$

The order of the multiplication can be changed as above because we are only measuring the intensity, i.e. the first element of the Stokes vector. Only the first row of the matrix **A** is involved in the calculation, i.e.

$$\begin{pmatrix} a_1 & a_2 & a_3 & a_4 \\ - & - & - & - \\ - & - & - & - \\ - & - & - & - \end{pmatrix} \begin{pmatrix} m_{11} & m_{12} & m_{13} & m_{14} \\ m_{21} & m_{22} & m_{23} & m_{24} \\ m_{31} & m_{32} & m_{33} & m_{34} \\ m_{41} & m_{42} & m_{43} & m_{44} \end{pmatrix} \begin{pmatrix} p_1 \\ p_2 \\ p_3 \\ p_4 \end{pmatrix} = \begin{pmatrix} I \\ - \\ - \\ - \end{pmatrix} \tag{28}$$

and multiplying through

$$\begin{aligned} I = {} & a_1(m_{11}p_1 + m_{12}p_2 + m_{13}p_3 + m_{14}p_4) \\ & + a_2(m_{21}p_1 + m_{22}p_2 + m_{23}p_3 + m_{24}p_4) \\ & + a_3(m_{31}p_1 + m_{32}p_2 + m_{33}p_3 + m_{34}p_4) \\ & + a_4(m_{41}p_1 + m_{42}p_2 + m_{43}p_3 + m_{44}p_4). \end{aligned} \tag{29}$$

The μ_{ij} are then given by

$$\mu_{11} = 1,$$
$$\mu_{12} = \cos^2 2\theta,$$
$$\mu_{13} = \sin 2\theta \cos 2\theta,$$
$$\mu_{14} = \sin 2\theta,$$
$$\mu_{21} = \cos^2 10\theta,$$
$$\mu_{22} = \cos^2 2\theta \cos^2 10\theta,$$
$$\mu_{23} = \sin 2\theta \cos 2\theta \cos^2 10\theta,$$
$$\mu_{24} = \sin 2\theta \cos^2 10\theta,$$
$$\mu_{31} = \sin 10\theta \cos 10\theta,$$
$$\mu_{32} = \cos^2 2\theta \sin 10\theta \cos 10\theta,$$
$$\mu_{33} = \sin 2\theta \cos 2\theta \sin 10\theta \cos 10\theta,$$
$$\mu_{34} = \sin 2\theta \sin 10\theta \cos 10\theta,$$
$$\mu_{41} = -\sin 10\theta,$$
$$\mu_{42} = -\cos^2 2\theta \sin 10\theta,$$
$$\mu_{43} = -\sin 2\theta \cos 2\theta \sin 10\theta,$$
$$\mu_{44} = -\sin 2\theta \sin 10\theta, \tag{30}$$

when the rotation ratio is 5:1. These equations can be expanded in a Fourier series to yield the Fourier coefficients which are functions of the Mueller matrix elements.

The inversion of these relations gives the Mueller matrix elements in terms of the Fourier coefficients:

$$m_{11} = a_0 - a_2 + a_8 - a_{10} + a_{12},$$
$$m_{12} = 2a_2 - 2a_8 - 2a_{12},$$
$$m_{13} = 2b_2 + 2b_8 - 2b_{12},$$
$$m_{14} = b_1 - 2b_{11} = b_1 + 2b_9 = b_1 + b_9 - b_{11},$$
$$m_{21} = -2a_8 + 2a_{10} - 2a_{12},$$
$$m_{22} = 4a_8 + 4a_{12},$$
$$m_{23} = -4b_8 + 4b_{12},$$
$$m_{24} = -4b_9 = 4b_{11} = 2(-b_9 + b_{11}),$$
$$m_{31} = -2b_8 + 2b_{10} - 2b_{12},$$
$$m_{32} = 4b_8 + 4b_{12},$$
$$m_{33} = 4a_8 - 4a_{12},$$
$$m_{34} = 4a_9 = -4a_{11} = 2(a_9 - a_{11}),$$

$$m_{41} = 2b_3 - b_5 = -b_5 + 2b_7 = (b_3 - b_5 + b_7),$$
$$m_{42} = -4b_3 = -4b_7 = -2(b_3 + b_7),$$
$$m_{43} = -4a_3 = 4a_7 = 2(-a_3 + a_7),$$
$$m_{44} = -2a_4 = 2a_6 = (a_6 - a_4). \quad (31)$$

The 5:1 rotation ratio is not the only ratio which can be used to determine Mueller matrix elements, but it is the lowest ratio where the expressions for the Fourier coefficients may be inverted.

4. SPECTROPOLARIMETRY

Spectropolarimetry is the spectroscopic study of the polarization properties of materials. This measurement technique is a generalization of conventional optical spectroscopy and of conventional polarimetry. Commercial spectropolarimeters and spectropolarimeters in the literature are usually concerned with instruments which pass one wavelength at a time through a sample. These instruments typically use a scanning monochrometer source. Because of the polarization effects that arise from diffraction gratings, and the length of time that would be necessary to obtain sample Mueller matrices at each wavelength, these spectropolarimeters are unsatisfactory for acquisition of precision polarimetric data. A spectropolarimeter based on a Fourier transform infrared (FTIR) spectrometer has been designed and built[16,17]. This instrument retains the advantages of a FTIR spectrometer over conventional spectrometers and is uniquely suited to collect polarimetric data quickly over large wavelength ranges. An optical diagram of the FTIR spectrometer as modified to be a spectropolarimeter is shown in Figure 4. This spectropolarimeter contains the same polarization element configuration used in the laser polarimeter described above. Spectrometer data at each orientation of the polarization elements as described for the polarimeter is collected and integrated to yield the Mueller matrix of a sample as a function of wavelength.

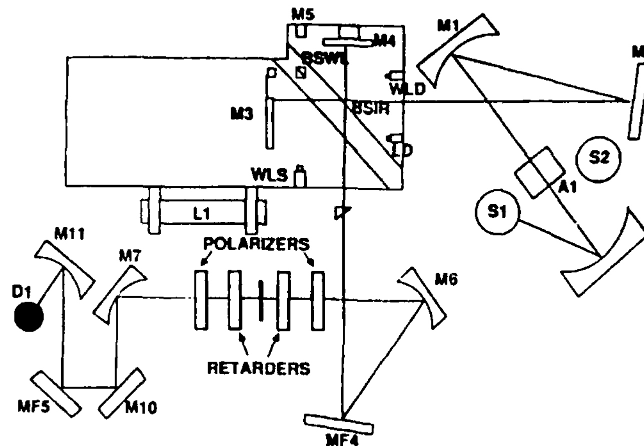

Figure 4: Spectropolarimeter optics

5. MEASUREMENT OF ELECTROOPTIC COEFFICIENTS

The goal of this work is to obtain electrooptic coefficients from Mueller matrices. The method by which this is accomplished in described in this section.

The application of an electric field across a crystal produces an index change as described. Specific index changes for a sample material is best presented in tabular form. Table 1 gives the index ellipsoid and the indices along the new principal axes. The new principal indices are obtained by solving an eigenvalue problem. For example, for a cubic material with a field perpendicular to the (1 1 1) plane, the index ellipsoid is

$$\frac{x^2+y^2+z^2}{n_o^2}+\frac{2}{\sqrt{3}}r_{41}E(yz+zx+xy)=1 \tag{32}$$

and the eigenvalue problem is

$$\begin{pmatrix} \frac{1}{n_o^2} & \frac{2r_{41}E}{\sqrt{3}} & \frac{2r_{41}E}{\sqrt{3}} \\ \frac{2r_{41}E}{\sqrt{3}} & \frac{1}{n_o^2} & \frac{2r_{41}E}{\sqrt{3}} \\ \frac{2r_{41}E}{\sqrt{3}} & \frac{2r_{41}E}{\sqrt{3}} & \frac{1}{n_o^2} \end{pmatrix} V=\frac{1}{n'^2}V. \tag{33}$$

The secular equation is then

$$\begin{pmatrix} \frac{1}{n_o^2}-\frac{1}{n'^2} & \frac{2r_{41}E}{\sqrt{3}} & \frac{2r_{41}E}{\sqrt{3}} \\ \frac{2r_{41}E}{\sqrt{3}} & \frac{1}{n_o^2}-\frac{1}{n'^2} & \frac{2r_{41}E}{\sqrt{3}} \\ \frac{2r_{41}E}{\sqrt{3}} & \frac{2r_{41}E}{\sqrt{3}} & \frac{1}{n_o^2}-\frac{1}{n'^2} \end{pmatrix}=0 \tag{34}$$

and the roots of this equation are the new principal indices.

The phase retardation accumulated by polarized light in traversing a medium with anisotropic properties is given by

$$\Gamma=2\pi(n_a-n_b)L/\lambda \tag{35}$$

where L is the medium thickness in the direction of propagation, λ is the wavelength of light, and n_a, n_b are the indices experienced in two orthogonal directions perpendicular to the direction of propagation. In the longitudinal mode of operation, the electric field and propagation direction are both along the z axis. The refractive indices experienced by the light are in the plane containing the x and y principal axes. If the light polarization and crystal are aligned so that the polarization is 45° from either principal axis, the phase retardation will be

$$\Gamma=2\pi(n_y'-n_x')L/\lambda \tag{36}$$

where n_y', n_x' are the (new) principal indices with the field applied. (For crystals with natural birefringence and with no electric field, these indices may just be the principal indices.)

The phase delays for light polarized at 45° to the principal axes of the material of Table 1 can now be calculated. The principal indices of the materials for an electric field parallel to the z and optical axes (longitudinal mode) are given in the first section of the table. The phase retardation for the $\bar{4}3m$ cubic material is

$$\Gamma_{cubic} = 2\pi n_o^3 r_{41} E L / \lambda . \tag{37}$$

If the electric field is expressed in terms of electric potential and charge separation, i.e. E = V/d, then the phase retardation is

$$\Gamma_{cubic}^{long} = 2\pi n_o^3 r_{41} V / \lambda \tag{38}$$

since the charge separation d is equal to the optical path through the crystal L. The phase retardation is linear with applied voltage. A retardation of π in the longitudinal mode will require a voltage

$$V_{hw}^{long} = \frac{\lambda}{2 n_o^3 r_{41}} . \tag{39}$$

The half wave voltage for CdTe is about 44 kV and for GaAs it is about 98 kV for 10.6 μm radiation. Since the voltage is directly proportional to wavelength, the half wave voltage would be considerably smaller at visible wavelengths.

The principal indices for the transverse mode are given in the second section of the table. The phase retardation for $\bar{4}3m$ cubic material is

$$\Gamma_{cubic} = 2\pi n_o^3 r_{41} E L / \lambda . \tag{40}$$

This is the same expression as for the longitudinal mode. In this mode, the charge separation is not the same as the optical path, so that when E is given as V/d, the phase delay is given as

$$\Gamma_{cubic}^{trans} = 2\pi n_o^3 r_{41} V L / d\lambda \tag{41}$$

The half wave voltage is given by

$$V_{hw}^{trans} = \frac{d\lambda}{2 n_o^3 r_{41} L} . \tag{42}$$

The half wave voltages for CdTe and GaAs for an optical path of 50mm and electrode separation of 5mm are 4.4kV and 9.8kV respectively.

The Mueller matrix formalism representation of a retarder with fast axis at arbitrary orientation angle θ is

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos^2 2\theta + \sin^2 2\theta \cos\delta & (1-\cos\delta)\sin 2\theta \cos 2\theta & -\sin 2\theta \sin\delta \\ 0 & (1-\cos\delta)\sin 2\theta \cos 2\theta & \sin^2 2\theta + \cos^2 2\theta \cos\delta & \cos 2\theta \sin\delta \\ 0 & \sin 2\theta \sin\delta & -\cos 2\theta \sin\delta & \cos\delta \end{pmatrix} \tag{43}$$

where the retardance is δ. If the retarder fast axis is assumed to be at 0°, the matrix becomes, substituting for δ the retardance of the crystal,

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & \cos\frac{2\pi}{\lambda}n^3 r_{41} V\frac{L}{d} & \sin\frac{2\pi}{\lambda}n^3 r_{41} V\frac{L}{d} \\ 0 & 0 & -\sin\frac{2\pi}{\lambda}n^3 r_{41} V\frac{L}{d} & \cos\frac{2\pi}{\lambda}n^3 r_{41} V\frac{L}{d} \end{pmatrix}. \qquad (44)$$

It is now clear that the electrooptic coefficient r_{41} can be obtained from the measured Mueller matrix.

Note that for purposes of obtaining the electrooptic coefficient experimentally, the fast axis of a electrooptic crystal acting as an ideal retarder can be at any orientation. The (4,4) matrix element of the matrix for a retarder with fast axis at angle Θ is independent of fast axis orientation, and the fast axis orientation can be eliminated elsewhere by adding the (2,2) and (3,3) matrix elements or squaring and adding elements in the fourth row and column. Given a measured Mueller matrix of a crystal, a known applied voltage, and a known refractive index, the electrooptic coefficient r_{41} can easily be obtained.

6. SUMMARY AND CONCLUSIONS

Two instruments, an infrared laser polarimeter and an infrared spectropolarimeter have been described. Using the Mueller formalism for the treatment of polarization, it has been shown that fundamental constants of sample electrooptic materials may be derived from measurement procedures described in this paper. Experimental procedures must be accomplished with careful preparation because of the many difficulties associated with these sensitive polarization measurements. The difficulty of obtaining data is commensurate with the difficulty of obtaining crystalline samples, and the equipment investment is orders of magnitude larger than the already high cost of the crystalline samples.

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Table 1: Electrooptical properties of cubic $\bar{4}3m$ crystals

E perpendicular to <001> plane $E_x = E_y = 0,\ E_z = E$

Index Ellipsoid: $$\frac{x^2 + y^2 + z^2}{n_o^2} + 2r_{41}Exy = 1$$

$$n_x' = n_o + \frac{1}{2}n_o^3 r_{41} E$$

$$n_y' = n_o - \frac{1}{2}n_o^3 r_{41} E$$

$$n_z' = n_o$$

E perpendicular to <110> plane $E_x = E_y = E/\sqrt{2},\ E_z = 0$

Index Ellipsoid: $$\frac{x^2 + y^2 + z^2}{n_o^2} + \sqrt{2}r_{41}E(yz + zx) = 1$$

$$n_x' = n_o + \frac{1}{2}n_o^3 r_{41} E$$

$$n_y' = n_o - \frac{1}{2}n_o^3 r_{41} E$$

$$n_z' = n_o$$

E perpendicular to <111> plane $E_x = E_y = E_z = E/\sqrt{3}$

Index Ellipsoid: $$\frac{x^2 + y^2 + z^2}{n_o^2} + \frac{2}{\sqrt{3}}r_{41}E(yz + zx + xy) = 1$$

$$n_x' = n_o + \frac{1}{2\sqrt{3}}n_o^3 r_{41} E$$

$$n_y' = n_o + \frac{1}{2\sqrt{3}}n_o^3 r_{41} E$$

$$n_z' = n_o - \frac{1}{\sqrt{3}}n_o^3 r_{41} E$$

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