Some of the most interesting and useful properties of optical materials are due to optical anisotropy or birefringence. Examples abound in optics. Polaroid films perform as polarizers in the visible spectral region, for example, but they act more as retarders at UV or near-IR wavelengths. Many crystals, such as calcite, quartz, magnesium fluoride, and lithium niobate, are naturally birefringent and can be used for polarizers, compensators, modulators, and other optical devices. Optical anisotropy can also be imposed externally using electric fields (the Pockels effect), magnetic fields (the Kerr effect), strain (the piezo-optic effect), and so on.

The measurement of optical anisotropy goes back to the very beginning of optics. The simplest experiment uses crossed polarizers: If we place an isotropic sample between the two crossed polarizers, then no light passes through the system, but an anisotropic sample will allow some light transmission. The literature is full of other, more complicated ways to measure optical anisotropy (involving polarimeters and ellipsometers, for example), usually using a combination of polarizers and retarders. Most of these experiments cannot measure all of the optical characteristics of anisotropic samples, however.

The interaction of materials with polarized light can best be understood using the Mueller-Stokes representation (another representation is due to R.C. Jones, and is summarized in reference 2). In the Mueller-Stokes representation, we represent the polarization state of a light beam by the Stokes vector $S = (I_0, I_{90} - I_0, I_{45} - I_{-45}, I_{rc} - I_{lc})^T$, where all elements are light intensities and the subscripts indicate the azimuthal angle of the polarization ($I_0$ is the total light intensity, and the $rc$ and $lc$ subscripts refer to right-circular and left-circular polarization states, respectively). We describe any intervening optical element by a $4 \times 4$ real matrix, called the Mueller matrix. In reflection (such as ellipsometry), an isotropic sample would have the Mueller matrix:

$$M = \begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix}$$

[1]
where $N = \cos(2\psi)$, $S = \sin(2\psi) \sin \Delta$, and $C = \sin(2\psi) \cos \Delta$, $\psi$ and $\Delta$ being the common ellipsometric angles. In transmission, we can use the same Mueller matrix to describe most materials. The meanings of the $N$, $S$, and $C$ parameters are considerably different, however: $-N$ now represents the diattenuation, and $S = (1 - N^2) \sin \delta$ and $C = (1 - N^2) \cos \delta$, where $\delta$ is the retardation. Equation 1 assumes that the fast axis of the material corresponds to the primary axis direction of the instrument (the plane of incidence in reflection); for random orientations, the Mueller matrix becomes:

$$
M = \begin{bmatrix}
1 & -N & C \\
-C & 1 & S \\
N & -S & 0 \\
0 & 0 & 0
\end{bmatrix}
$$

[2]

where $S_s = \sin(2\phi)$, $C_s = \cos(2\phi)$, and $\phi$ is the azimuthal angle of the fast axis.

Very few optical instruments measure enough parameters to totally characterize the Mueller matrix in equation 2 (typical ellipsometers measure only 2 to 4 parameters). Our group has developed the two-modulator generalized ellipsometer (2-MGE), which measures eight parameters and allows us to determine equation 2 completely.\(^3,4\)

Two Times the Ability

The 2-MGE consists of two polarizer/photoelastic-modulator (PEM) pairs in which each PEM operates at a distinct vibrational frequency; we use 50 and 60 kHz (see figure 1). In a PEM, a piece of crystalline quartz is electronically excited to resonate at a frequency $\omega$ determined by its shape and crystal orientation. The light beam passes through a bar of fused silica, which is also cut to resonate at $\omega$. The refractive index along the long axis of the fused silica bar will then oscillate at $\omega$, and the device acts as a dynamic retarder with retardation $\delta = A \sin(\omega t)$. If the incident light beam is polarized at 45° with respect to the long axis of the PEM, then the exiting light beam will be dynamically elliptically polarized, oscillating through the various polarization states at frequency $\omega$. The range of polarization states generated by the polarizer/PEM pair depends on the modulation amplitude $A$ of the PEM, and the wavelength of light. In the 2-MGE, each polarizer/PEM pair produces dynamically elliptically polarized light.

The configuration of the 2-MGE is: Polarizer-PEM$_0$ / Sample / PEM$_1$ -Polarizer, and can be operated either in transmission mode or in reflection mode as a traditional ellipsometer. The light intensity after the last polarizer is a function of time:

$$I(t) = I_0 + I_{00}X_0 + I_{00}X_0 + I_{01}Y_1 + I_{01}Y_1 + I_{10}X_0Y_1 + I_{10}X_0Y_1 + I_{11}X_1Y_1 + I_{11}X_1Y_1$$

[3]

where the basis functions are

$$X_i = \sin(A_i \sin(\omega t)), \quad Y_i = \cos(A_i \sin(\omega t)), \quad i = 0, 1$$

[4\text{a,b}]

The frequencies of the PEMs are $\omega_i$, and the PEM modulation amplitudes are proportional to $A_i$ (usually controlled by an external DC voltage). We can then describe the intensity waveform entirely by the nine $I_i$ coefficients, but most experiments normalize the intensity waveform to $I_{dc}$, leaving eight coefficients to be determined experimentally. Setting the azimuthal orientations of the two PEMs to (0°, 45°) or (45°, 0°) allows us to completely determine the sample Mueller matrix of equation 2.

The 2-MGE in Action

We have conducted a variety of experiments in our laboratory with the 2-MGE. We have used it in reflection mode to determine the optical functions of uniaxial crystals. In normal ellipsometric measurements of isotropic samples, the $S$- and $P$-polarization states are eigenmodes of the reflection; that is, pure $S$- (or $P$-) polarized input light results in pure $S$- (or $P$-) polarized output light. For anisotropic samples, this is no longer true and may result in cross polarization. As a consequence, the off-block elements of equation 1 are no longer 0, which requires more sophisticated ellipsometry measurements. This is easily done with the 2-MGE, where both the ordinary and the extraordinary complex refractive indices of uniaxial crystals can be determined with a single measurement.\(^5\)

With this method, we have determined the complex refractive index of zinc oxide. The direct band edge for light polarized perpendicular to the optic axis (ordinary axis) is about 40 mV below the direct band edge for light polarized along the optic axis (extraordinary axis). We can clearly observe the excitonic peaks for both polarizations.

In transmission, we can use the 2-MGE to completely characterize a general diattenuator and retarder, including the direction of the fast axis, as noted in equation 2. One such example of this type of optical element is a Polaroid film. From 500 to 700 nm, $N \sim -1$, and $S \sim C \sim 0$, which shows that the material acts as a good polarizer. Below about 450 nm and above about 700 nm, $|N| < 1$ and $|S|, |C| > 0$, indicating that the Polaroid becomes a combination of diattenuator and retarder. In the near-IR spectral region (above 800 nm), the Polaroid becomes more of a retarder than a diattenuator.

Figure 1 A photoelastic modulator (PEM) consists of an electronically excited piece of crystalline quartz resonating at $\omega$, optically contacted to a bar of fused silica, which is also cut to resonate at $\omega$. The PEM is usually paired with a polarizer oriented at ±45° with respect to the long axis of the PEM. The resultant Stokes vector of the transmitted light beam through a polarizer/PEM pair is given by $S$, where $\theta$ is the azimuthal angle of the PEM.
We can also use the system to make optical measurements of internal electric fields via the Pockels effect. Generally, when we apply a voltage to a material, the electric field remains uniform throughout the sample. In certain materials, however, an applied voltage triggers a refractive index change proportional to the local electric field (the Pockels effect). In some cases, for example lithium niobate (LiNbO₃), this interaction is large enough that the material can be used to make electrooptic devices such as modulators.

Our group has used the 2-MGE in transmission mode to examine several LiNbO₃ crystals under bias (see figure 2). In these experiments, the 2-MGE operates at a single wavelength, but with focusing optics to isolate a small part of the sample for examination. Translating the material using automatic x-y stages allows us to image different parts of the sample, thereby generating a retardation map of the sample. We have formed such a map for a LiNbO₃ crystal cut such that the c-axis is along the direction of the light. As can be seen, the application of the voltage alters the spatially dependent retardation, and the retardation and the direction of the fast axis (and therefore the magnitude and direction of the electric field) are clearly not uniform between the electrodes.

In summary, the 2-MGE is a particularly useful optical instrument for examining anisotropic samples. In reflection, the 2-MGE acts as a generalized ellipsometer, and can determine both the ordinary and extraordinary refractive indices for a properly oriented sample. In transmission, the 2-MGE can be used to determine the diattenuation, direction of the fast axis, and retardation as well as the circular diattenuation and polarization factor (not shown).

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