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Phase-conjugate interferometry for thin film analysis

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The University of Arizona, 1990
PHASE-CONJUGATE INTERFEROMETRY FOR
THIN FILM ANALYSIS

by
Elaine Ruth Parshall

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1990
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ABSTRACT

A phase-conjugate interferometric method of thin film analysis obtains three independent parameters with which to determine a film's refractive index $n$, absorption coefficient $\kappa$, and thickness $d$. Because dimensionless intensity ratios are used, this method is self-calibrating except for light source polarization and incident angle. The use of self-pumped phase-conjugate reflectors makes the interferometer self-aligning and results in infinite spacing of fringes of equal thickness. A single layer thin film sample was analyzed by this technique, and the results compared to those of ellipsometry.
INTRODUCTION

Increased application of thin film structures to such widely diverse technologies as solar energy and magneto-optics, high reflectance and anti-reflectance coatings, and spectral filters requires suitable techniques for characterizing the optical properties of an absorbing thin film. Preferably, the technique should be easy to use, permit multi-wavelength measurements at varying angles of incidence, provide unambiguous data, and be relatively inexpensive. Ellipsometry, probably the most common method of thin film analysis, can become very complicated and expensive in providing multi-wavelength, multi-incident angle measurements.

An alternative method of thin film analysis based on two-beam interferometry has existed since the early 1970's that is simpler and less expensive than ellipsometry. At the same time, a single experimental set-up based on interferometric methods provides more independent parameters than ellipsometry does with which to characterize the thin film.

A phase-conjugate interferometer (PCI) can perform the thin film analysis more easily and with greater accuracy than a conventional interferometer. An interferometer becomes a PCI when the conventional mirrors are replaced with photorefractive crystals that produce a phase-conjugate reflection by self-pumped degenerate four-wave mixing. These devices are commonly known as phase-conjugate mirrors (PCM's).

This thesis discusses interferometric analysis of thin films using a PCI. It is shown that such an interferometer greatly
simplifies optical alignment, improves the temporal stability of the interfering beams, and lessens the effect of distorted wavefronts due to nonhomogeneities in the beam paths as compared to a regular interferometer.

The theory of thin film interferometry is described in section I. In section II, the use of phase-conjugate mirrors is explained, followed by a discussion on their application to thin film interferometry. Section III describes an experimental set-up for characterization of a single layer film and presents some results. A discussion section follows. Improvements to this technique with extensions to analyzing different types of multilayer films are discussed in the conclusion.
SECTION I: THIN FILM INTERFEROMETRY

Absorption in the beamsplitter of a two-beam interferometer causes a relative phase change between the two exiting beams. A comparison of the beam intensities (hereafter defined as optical power per unit area) at ports 1 and 2 of Fig. 1 provides information regarding this phase change from which the beamsplitter's optical properties can be determined. Likewise, a thin film's optical properties can be found if the beamsplitter (BS 2) is replaced with the film of interest deposited on a transparent substrate. It is shown below that the output intensities of the interferometer are functions of the film's reflection and transmission coefficients.

It is assumed that the source light incident on the thin film has unity amplitude. The resulting intensities at ports 1 and 2 are

\[ D_1 = |\rho_1 t^t + \rho_2 r^{r'}|^2 \]
\[ D_2 = |\rho_1 t^r + \rho_2 r^{r'}|^2, \]

where \( r, r', t, \) and \( t' \) are the reflection and transmission coefficients of the film, primed and unprimed quantities denote light incident on the film from the left and right respectively, and \( \rho_1 \) is the reflectivity of each interferometer mirror. For the present, it is assumed that these mirrors are not PCM's. Expanding equations (1) and (2) gives

\[ D_1 = |\rho_1 t^t|^2 + |\rho_2|^2 |r^{r'}|^4 + (\rho_1^* \rho_2 t^t r^{r'} + c.c.) \]
\[ D_2 = |\rho_1 t^r|^2 + |\rho_2 t^{r'}|^2 + (\rho_1^* \rho_2^* t^t r^{r'} + c.c.), \]
where (\*) and c.c. denote complex conjugate. It is seen that the outputs of the interferometer can be expressed in terms of the film's reflection and transmission coefficients $r$, $r'$, $t'$ and $t$.

Furthermore, the reflection and transmission coefficients are functions of the film's optical parameters: the index-of-refraction $n$, the absorption coefficient $\kappa$, and the thickness $d$. The most
commonly used expressions for $r$ and $t$ for light polarized in the plane-of-incidence (p-polarized) are given by:

$$
\begin{align*}
\begin{vmatrix}
A & B \\
C & D \\
\end{vmatrix}
= \begin{vmatrix}
A_p & B_p \\
C_p & D_p \\
\end{vmatrix} + \begin{vmatrix}
A & B \\
C & D \\
\end{vmatrix}
\end{align*}
$$

$$
\begin{align*}
& r_p = \frac{A_p + B_p - C - D_p}{A_p + B_p + C + D_p} \\
& r_p = \frac{2A_p}{A_p + B_p + C + D_p} \\
& t_p = \frac{2A_p}{A_p + B_p + C + D_p}
\end{align*}
$$

The subscript $i$ denotes the $i$th medium, $p_i = n_i/cos\theta_i$, $\theta_i$ is the propagation angle, and $A$, $B$, $C$, and $D$ are the elements of the 2x2 thin film characteristic matrix:

$$
\begin{align*}
M_i =
\begin{vmatrix}
A & B \\
C & D \\
\end{vmatrix}
= \begin{vmatrix}
\cos\beta_i & (i/p_i)\sin\beta_i \\
-i^p\sin\beta_i & \cos\beta_i \\
\end{vmatrix}
\end{align*}
$$

where $\beta_i = (2\pi/\lambda)n_i d_i \cos\theta_i$, $d_i$ being the $i$th-layer film thickness, and $\lambda$ is the incident wavelength in vacuum.

However, for this analysis, it was found to be more convenient to use a method formalized by Yeh in which $r$, $r'$, $t$ and $t'$ can be expressed as

$$
\begin{align*}
M = \begin{vmatrix}
M_{11} & M_{12} \\
M_{21} & M_{22} \\
\end{vmatrix}
\end{align*}
$$

$$
\begin{align*}
& r = M_{21}/M_{11} \\
& r' = -M_{12}/M_{11} \\
& t = 1/|M| \\
& t' = |M|/M_{11}
\end{align*}
$$

where $M_{i,j}$ is an element of a matrix that characterizes both the film and the bounding media. This matrix differs from the one in equation (7) which describes only the film's optical properties. The electromagnetic derivation of these relationships follows.
Boundary conditions for electromagnetic waves require that the tangential components of the electric field \( E \) and the magnetic field \( H \) be continuous at an interface between two dissimilar media. The media are assumed to be homogeneous, isotropic, linear and stationary, and the incident fields linearly polarized. Referring to Fig. 2, for \( p \)-polarized waves at \( z = 0 \),

\[
(E_1 + E_1')\cos\theta_1 = (E_2 + E_2')\cos\theta_2 \tag{9a}
\]

\[
(E_1 - E_1')n_1 = (E_2 - E_2')n_2 \tag{9b}
\]

where \( \theta_1 \) is the angle of propagation in medium \( i \), primed and unprimed quantities denote fields travelling to the left and right, respectively, and \( H_i = n_i E_i \). These equations for \( E \) and \( H \) can be written in matrix form as

\[
\begin{pmatrix}
D_p(1) & E_1 \\
E_1' & D_p(2)
\end{pmatrix} = \begin{pmatrix}
D_p(1) & E_2 \\
E_2' & D_p(2)
\end{pmatrix}, \tag{10}
\]

where

\[
D_p(i) = \begin{pmatrix}
\cos\theta_i & \cos\theta_i \\
n_i & n_i
\end{pmatrix} \tag{11}
\]

Yeh refers to \( D_p(i) \) as the dynamical matrices for the \( p \)-wave. At \( z = L \), the tangential components of \( E \) and \( H \) become
Figure 2: Thin film boundary conditions for \( p \)-polarized fields.

\[
\begin{align*}
(E_2 \exp[-i\beta_1] & + E_2' \exp[i\beta_1]) \cos \theta_2 - (E_3 + E_3') \cos \theta_3 \quad (12a) \\
(E_2 \exp[-i\beta_1] & - E_2' \exp[i\beta_1]) n_2 - (E_3 - E_3') n_3 \quad (12b)
\end{align*}
\]

where \( \beta_1 \) is as defined for equation (7). These equations can be written as

\[
P^{-1}(2)D_p(2)\Sigma_2 = D_p(3)\Sigma_3, \quad (13)
\]
where $\Sigma_1 = \frac{E_1}{E_1'}$, $P(i)$ is defined as

$$P(i) = \begin{bmatrix} \exp[i\beta_i] & 0 \\ 0 & \exp[-i\beta_i] \end{bmatrix},$$

(14)

and is called the propagation matrix, and $P^{-1}$ denotes the inverse of $P$.

The reflection and transmission coefficients can be expressed as

$$r_p = \frac{E_1'}{E_1} \quad \text{and} \quad t_p = \frac{E_3}{E_1},$$

(15)

for $E_2' = E_3' = 0$, respectively. Therefore, equations (10) and (13) need to be rearranged to obtain an expression for $E_1$ and $E_1'$ in terms of $E_3$ and $E_3'$:

$$\Sigma_1 = D_p^{-1}(1)D_p(2)\Sigma_2,$$

(16)

$$\Sigma_2 = P(2)D_p^{-1}(2)D_p(3)\Sigma_3.$$

(17)

Combining these two equations and dropping the $p$-subscript for $D$ yields

$$\Sigma_1 = D_1^{-1}D_2P_2D_2^{-1}D_3\Sigma_3.$$  

(18)

Equation (18) describes the $E$ fields for a single layer. For $N$ layers, this equation can be written as

$$\Sigma_1 = D_1^{-1}(D_1P_1D_1^{-1}D_2P_2D_2^{-1}...D_NP_ND_N^{-1})D_S\Sigma_S,$$

(19)

or

$$\Sigma_1 = D_1^{-1}(\prod_{n=1}^{N} D_nP_nD_n^{-1})D_S\Sigma_S,$$

(20)
where the subscripts i and s denote the incident and substrate mediums, respectively. The resulting product of the dynamical and propagation matrices can be regarded as a single matrix $M$ for the system.

To derive the Fresnel coefficients for a single layer film, this matrix is written in the form

$$D_1^{-1}D_2P_2D_2^{-1}D_3 = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix}. \quad (21)$$

Equation (20) may then be rewritten as

$$\Sigma_1 = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \Sigma_3 . \quad (22)$$

Expanding this equation gives

$$E_1 = M_{11}E_3 + M_{12}E_3', \quad (23a)$$

$$E_1' = M_{21}E_3 + M_{22}E_3', \quad (23b)$$

and for $E_3' = 0,$

$$E_1 = M_{11}E_3 \quad (24a)$$

$$E_1' = M_{21}E_3. \quad (24b)$$

Referring then to equation (15), $r$ and $t$ become

$$r = M_{21}/M_{11} \quad t = 1/M_{11} . \quad (25)$$
Likewise, for the reverse propagating beam,

\[ r' = \frac{E_3}{E_3'} \quad t' = \frac{E_1'}{E_3'} \] (26)

for \( E_1 = 0 \), and rearranging equation (23) gives

\[ E_3 = -E_3' \left( \frac{M_{12}}{M_{11}} \right) \] (27a)

\[ E_1' = E_3' \left( \frac{M_{22}M_{11} - M_{21}M_{12}}{M_{11}} \right), \] (27b)

so that \( r' \) and \( t' \) become

\[ r' = -\frac{M_{12}}{M_{11}} \quad t' = \frac{|M|}{M_{11}}, \] (28)

where \( |M| = M_{22}M_{11} - M_{21}M_{12} \).

Referring to equation (21), these matrix elements have the form

\[
\begin{align*}
2M_{11} &= \frac{\cos \theta_3}{\cos \theta_1} + \frac{n_3}{n_1} \cos \beta_2 + \frac{n_2 \cos \theta_3}{n_1 \cos \theta_2} + \frac{n_3 \cos \theta_2}{n_2 \cos \theta_1} \quad (29a) \\
2M_{12} &= \frac{\cos \theta_3}{\cos \theta_1} - \frac{n_3}{n_1} \cos \beta_2 + \frac{n_2 \cos \theta_3}{n_1 \cos \theta_2} - \frac{n_3 \cos \theta_2}{n_2 \cos \theta_1} \quad (29b) \\
2M_{21} &= \frac{\cos \theta_3}{\cos \theta_1} - \frac{n_3}{n_1} \cos \beta_2 - \frac{n_2 \cos \theta_3}{n_1 \cos \theta_2} - \frac{n_3 \cos \theta_2}{n_2 \cos \theta_1} \quad (29c) \\
2M_{22} &= \frac{\cos \theta_3}{\cos \theta_1} + \frac{n_3}{n_1} \cos \beta_2 - \frac{n_2 \cos \theta_3}{n_1 \cos \theta_2} + \frac{n_3 \cos \theta_2}{n_2 \cos \theta_1} \quad (29d)
\end{align*}
\]

Yeh’s reflection coefficient of equation (25) is equivalent to Born and Wolf’s of equation (5) if the numerator and denominator of (25)
are multiplied by $n_1/cos\theta_3$. The transmittances $T$ are equal if $T$ for Yeh is written as

$$T = (n_3cos\theta_3/n_1cos\theta_1)|1/M_{11}|^2, \quad (30)$$

whereas for Born and Wolf,

$$T = (n_3cos\theta_1/n_1cos\theta_3)|t_p|^2, \quad (31)$$

where $t_p$ is defined in equation (6). Furthermore, the classical thin film characteristic matrix in equation (7) equals Yeh's $D_1P_1D_1^{-1}$.

By substituting equations (25) and (28) into equations (3) and (4), the output intensities are expressed in terms of the matrix elements:

$$D_1 = \frac{R_1^2|M|^2 + R_2^2|M_{12}|^4 + (R_1R_2|M|(M_{12}^*|2exp[-i\psi] + c.c)}{|M_{11}|^4}, \quad (32a)$$

$$D_2 = \frac{R_1^2|M_{21}|^2 + R_2^2|M_{12}|^2 - (R_1R_2|M|2M_{21}^*M_{12}|exp[-i\psi] + c.c.}}{|M_{11}|^4}, \quad (32b)$$

where $\rho_1\rho_2^* = R_1R_2exp[-i\psi]$ and $R_1 = |\rho_1|$. Letting $M_{ij} = m_{ij}exp[i\phi_{ij}]$, these equations become

$$D_1 = \frac{R_1^2|M|^2 + R_2^2|m_{12}|^4 + 2R_1R_2|M|m_{12}^2cos[\Psi]}{m_{11}^4}, \quad (33a)$$

$$D_2 = \frac{R_1^2|M|^2|m_{21}|^2 + R_2^2|M|^2|m_{12}|^2 - 2R_1R_2|M|2m_{21}m_{12}cos[\Psi - \Delta]}{m_{11}^4}, \quad (33b)$$
where $\psi = \psi + 2\phi_{12}$ and $\Delta = \phi_{21} + \phi_{12}$. These equations relate the signals at ports 1 and 2 ($D_1$ and $D_2$) to the thin film matrix elements and correspondingly $n$, $\kappa$, and $d$. The signal analysis will be explained in Section III. The mechanism of the phase-conjugate mirrors which are used in this experiment is discussed in the next section.
SECTION II: PHASE-CONJUGATE MIRRORS

This section is divided into three parts. The first part explains optical phase-conjugation (OPC), the second part contains a description of degenerate four-wave mixing and the photorefractive effect, and the third part examines self-pumped PCM's and their applicability to thin film interferometry.

Optical phase conjugation

In order to describe the operation of a PCM, the concept of OPC must first be explained. OPC occurs when an optically nonlinear material, having optical fields incident upon it, produces an electromagnetic field that propagates in the reverse direction with the reverse overall phase of one of the incident optical fields. In other words, the propagation vector changes direction by 180 degrees, and the electric field amplitude is complex conjugated. This effect can be demonstrated by a plane wave travelling in the +z direction

\[ E_1(r,t) = A_1(r,t) e^{i(wt - k\cdot r)} + c.c., \]  

where \( w \) is the oscillation frequency and \( k \) the propagation vector of the field, and \( A_1(r,t) \) is the slowly-varying complex amplitude envelope of the wave. The reason for separating out this component of the wave is explained later. Equation (36) is substituted into Maxwell's wave equation

\[ \nabla^2 E - \epsilon(r) \mu_0 (\partial^2 E / \partial t^2) = 0, \]

where \( \epsilon(r) \) is the nonlinear medium's dielectric tensor and \( \mu_0 \) the magnetic permeability of the medium, yielding
\[ \nabla^2 A_1 + \left[ w^2 \varepsilon(r) \mu_0 - k^2 \right] A_1 - 2ik(\partial A_1 / \partial z) = 0. \tag{38} \]

The time-varying terms \(|\partial A_1 / \partial t|\) and \(|\partial^2 A_1 / \partial^2 t|\) are assumed to be much smaller than \(|w^2 A_1|\). If equation (38) is then complex conjugated, where the spatial complex conjugate of \(E_1\) is given by

\[ E_1^*(r,t) = A_1^*(r,t)e^{i(\omega t + kz)} + c.c., \tag{39} \]

the wave equation becomes\(^1\)

\[ \nabla^2 A_1^* + \left[ w^2 \varepsilon(r) \mu_0 - k^2 \right] A_1^* + 2ik(\partial A_1 / \partial z) = 0, \tag{40} \]

where \(\varepsilon(r)\) is assumed to be real-valued. This equation also describes a wave

\[ E_2(r,t) = A_2(r,t)e^{i(\omega t + kz)} + c.c., \tag{41} \]

which is travelling in the \(-z\) direction with a complex amplitude \(A_2 = A_1^*\). \(E_1\) is said to be phase-conjugated. As shown in Fig. 3, a wave that is phase-conjugated has its wavefront distortions removed when the phase-conjugate wave propagates back through the distorting medium. If this medium were a converging lens with collimated light incident on it, the phase-conjugate wave would become recollimated upon travelling back through the lens.

Now how does OPC occur? The optical nonlinearities of the phase-conjugating medium arise when incident optical fields cause the polarization of the material to change, as manifested by a change in its index-of-refraction. The polarization can be expressed as
Figure 3: Complex conjugate of an input field with distorting medium. From reference 11.

\[ P = \varepsilon_0 \chi E, \quad (42) \]

where \( \chi \) is the material's electric susceptibility, and \( E \) represents the sum of the fields incident on the medium. Expanding \( \chi \) in a power series with respect to \( E \),
\[ \chi = \chi^{(1)} + \chi^{(2)}E + \chi^{(3)}E^2 + \ldots, \quad (43) \]

and substituting this expression into equation (42) yields

\[ P = \varepsilon_0 E(\chi^{(1)} + \chi^{(2)}E + \chi^{(3)}E^2 + \ldots). \quad (44) \]

\( \chi^{(1)} \) corresponds to first-order optical effects that are independent of \( E \) such as a medium's index-of-refraction or absorption properties. \( \chi^{(2)} \), a second-order property, is responsible for second harmonic generation, the electro-optic effect, and other three-wave mixing effects that are linearly dependent on \( E \). The third-order property, characterized by \( \chi^{(3)} \), is intensity-dependent \( (I \propto |E|^2) \). Stimulated Brillouin and Raman scattering, self-focusing, and non-degenerate and degenerate four-wave mixing result from this property.

**Degenerate four-wave mixing and the photorefractive effect**

In degenerate four-wave mixing (FWM), the effect of interest in this thesis, it is assumed that each incident field has the same frequency and polarization. The resulting nonlinear polarization can be written as

\[ P_l = 4\chi_{ijkl}E_jE_kE_1, \quad (45) \]

where the subscripts refer to the four optical fields, and \( \chi_{ijkl} \), a fourth rank tensor, is proportional to \( \chi^{(3)} \). This expression for \( P \) contains terms with oscillation frequencies \( [+/-w_1 +/ -w_2 +/ -w_3] \) and propagation vectors \( [+/-k_1 +/ -k_2 +/ -k_3] \).

The interaction of these fields can be described using Maxwell's equations. If the medium's polarization is expressed as a sum of linear and nonlinear components, of
\[ P = P_L + P_{NL} \text{,} \quad (46) \]

then the wave equation becomes

\[ \nabla^2 E - \mu_0 \epsilon (\partial E^2 / \partial t^2) = \mu_0 (\partial^2 P_{NL} / \partial t^2) \text{,} \quad (47) \]

where the relationship \( \epsilon E = \epsilon_0 E + P_L \) is used. \( P_{NL} \) is seen as a driving, or source term that produces the phase-conjugate field \( E \). To conserve energy and momentum, \( P \) must be phase-matched to \( E \). This requirement is satisfied by terms in equation (45) with frequencies \( \omega_1 = \omega_2 = \omega_3 = -\omega_4 \), and propagation vectors \( k_1 = -k_2, k_4 = -k_3 \) as in the FWM scheme shown in Fig. 4. Beams \( A_1 \) and \( A_2 \), the pump beams, have wave vectors \( k_1 \) and \( k_2 \), the signal beam \( A_4 \) has wave vector \( k_4 \), and the phase conjugate beam \( A_3 \) has wave vector \( k_3 \).

To solve the wave equation for each field, the equations are reduced to first-order partial differential equations by making the slowly-varying envelope approximation (SVEA). This approximation requires that the complex amplitude envelope, \( A_i(r,t) \), vary only a small amount over one period of the wave, or

\[ |\partial^2 A_i / \partial z^2| \ll |k \partial A_i / \partial z|, |k^2 A_i|, \quad (48a) \]

\[ |\partial^2 A_i / \partial t^2| \ll |w \partial A_i / \partial t|, |w^2 A_i|. \quad (48b) \]

The terms remaining for \( \nabla^2 E \) and \( \partial^2 E / \partial t^2 \) are then

\[ \nabla^2 E_1 = [-2ik \partial A_1 / \partial z - k^2 A_1] e^{i(\omega t - kz)} + \text{c.c.}, \quad (49a) \]

\[ \partial^2 E_1 / \partial t^2 = [2i w \partial A_1 / \partial t - w^2 A_1] e^{i(\omega t - kz)} + \text{c.c.} \quad (49b) \]
If $P_{NL}$ is expressed as

$$P_{NL} = P(r,t)e^{i(\omega t - kz)} + \text{c.c.},$$

(50)

Figure 4: Four-wave mixing arrangement for OPC.

where $P(r,t)$ is the slowly-varying envelope component of $P_{NL}$, and the same assumptions are made for $P_{NL}$ that were made for $A_1$, then the second-order wave equation is reduced to

$$[\partial/\partial z + \mu_0 \varepsilon \partial/\partial t]A_1 = iw/2\mu_0/\varepsilon P_{NL}.$$  

(51)
The following coupled wave equations result:\(^{12}\)

\[
\frac{dA_1}{dz} = -\kappa (9 |A_1|^2 + 18 |A_2|^2)A_1 \\
\frac{dA_2}{dz} = \kappa (9 |A_2|^2 + 18 |A_1|^2)A_2 \\
\frac{dA_3}{dz} = -\kappa A_1A_2A_4^* \\
\frac{dA_4}{dz} = \kappa A_1A_2A_3^*,
\]

where \(\kappa = (i\omega/2)\mu_0/\epsilon_0\chi^{(3)}\). The coupling between fields is governed by the third-order susceptibility \(\chi^{(3)}\), which, as was mentioned earlier, is an intensity-dependent effect. Consequently, intense optical fields are usually required for OPC to occur in media that depend on this type of nonlinearity to produce a phase-conjugate beam.

However, low intensity fields can be used for OPC if the nonlinearity arises from the photorefractive effect in crystals such as barium titanate (BaTiO_3) or BSO (Bi_{12}SiO_{20}). Changes in the index-of-refraction in these materials are a result of the electro-optic effect, a \(\chi^{(2)}\) property, that is only linearly dependent on \(E\). The photorefractive effect occurs when two beams form an interference pattern with spatial distribution \(I(x)\) within a photorefractive crystal. Electrical charges in the crystal migrate to regions of low intensity, thereby creating a space charge \(\rho_{sc}\). An electric field is set up in the crystal according to Poisson's equation\(^{11}\)

\[
\nabla E_{sc} = \rho_{sc}/\epsilon_0.
\]
Through the electro-optic effect, the index-of-refraction changes in the crystal,
\[ \Delta n = -(1/2) n_0^3 r_{\text{eff}} E_{\text{sc}}, \]  
where \( r_{\text{eff}} \) is the effective electro-optic coefficient whose strength depends on the crystalline composition and the orientation of the crystal axis with respect to the space charge field. The value of the electro-optic coefficient is also dependent upon the polarization of the incident fields, being largest in perovskites such as BaTiO₃ for extraordinary polarization. These effects are summarized in Fig. 5.

Due to the gradient of \( E_{\text{sc}} \) in equation (53), the index-of-refraction grating is spatially shifted with respect to \( \rho_{\text{sc}} \) and \( I(x) \). For a photorefractive crystal that has no externally applied field other than optical fields, the phase shift equals \( \pi/2 \). This phase shift is responsible for the coupling which occurs between two beams incident on a crystal, and for the subsequent amplification of one beam at the expense of the other.¹³

The phase grating formed from the interference of the two beams can be written as¹⁴
\[ n = n_0 + (n_I \exp[i\phi_I]/2I_0)(A_1A_4^* + A_2A_3^*)\exp[ikz] + \text{c.c}, \]  
where \( n_0 \) is the crystal's first-order index-of-refraction, \( n_I \) is proportional to \( E_{\text{sc}} \), \( \phi_I \) is the spatial phase shift of \( n_I \) with respect to the interference pattern, \( k_I \) is the phase-grating vector due to the interference of beams 1 and 4, and 2 and 3, and \( I_0 \) is the sum of the four beam's intensities. Because of normalization by \( I_0 \), the change
the space charge $\rho^{SC}$, and consequently $n$, are approximately independent of total intensity.\textsuperscript{15}

![Diagram of photorefractive index grating]

Figure 5: Formation of a photorefractive index grating. From reference 13.

When equation (55) is substituted into the scalar wave equation for each of the four fields incident upon the crystal as shown in Fig. 6, the coupled wave equations become

$$\frac{dA_1}{dz} = (-\gamma/I_0)(A_1A_4^* + A_2^*A_3)A_4$$

(56a)
where $\gamma = (iwn_1 \exp[-i\phi_1])/(2c\cos\alpha)$, $c$ is the velocity of light, and $\alpha$ is the angle that beams $A_1$ and $A_2$ make with respect to the normal to the crystal surface.
So for photorefractive FWM, the coupling is seen to be intensity-independent. Only the time response, or time of formation of the phase grating is intensity-dependent. The number of charges that migrate per unit time depends upon the incident optical power per unit area whereas the total number of charges that are free to migrate is fixed, regardless of incident intensity.

Both photorefractive and non-photorefractive FWM media have been used as PCM's for such purposes as distortion-correction devices in resonator cavities and as mirrors in interferometers. For these particular applications, the PCM corrects for both uniform and relative phase shifts of the incident wavefronts. But for thin-film interferometry, if the uniform phase shift due to the thickness and absorption of the thin film is cancelled by a phase-conjugate reflection, the information for analysis of the optical parameters is lost.

On the other hand, it is desirable to cancel out relative wavefront distortions due to nonuniformities in the thin film and its substrate. A self-pumped PCM accommodates both of these situations: uniform phase shifts in the incident wavefront are not cancelled by the phase-conjugate reflection, while relative phase shifts are. A description of this phenomena and how the pump beams are generated follows.

**Self-pumped phase-conjugation**

The properties of a phase-conjugate reflection for a self-pumped PCM differ compared to that of an externally-pumped PCM because the
pump beams of the former are generated internally from the incident beam \( A_4 \), and therefore have approximately the same phase as \( A_4 \). The internal pump beams are generated as amplified scattering from scattering centers that causes the incident beam \( A_4 \) to fan out within the crystal. Photorefractive gain generates counterpropagating oscillation beams with feedback via total internal reflection at the crystal surfaces. These oscillation beams act as the pump beams for phase conjugation.

The phase-conjugate reflection \( A_3 \) is given by\(^{23}\)

\[
A_3 = \rho' \exp[i(\psi_1 + \psi_2)]A_4^*, \quad (57)
\]

where \( \psi_1 \) and \( \psi_2 \) are the phases of the pump beams, and \( \rho' \), the complex reflection coefficient of the PCM, is independent of the phases of the interacting beams. In a self-pumped phase conjugator, the pump beams are derived directly from the signal so that

\[
\begin{align*}
\psi_1 &= \psi_4 + \delta_1 \\
\psi_2 &= \psi_4 + \delta_2,
\end{align*}
\]

where \( \psi_4 \) is the phase of the signal beam, and \( \delta_1 \) and \( \delta_2 \) depend on the optical path travelled by beam 4 in the PCM while becoming pumps 1 and 2, respectively. Thus

\[
A_3 = \rho \exp[2i\psi_4]A_4^*, \quad (59)
\]

where \( \rho = \rho' \exp[\delta_1 + \delta_2] \), which simply represents an overall phase shift in the signal beam. If \( A_4^* = |A_4| \exp[-i\psi_4] \), then equation (59) becomes
\[ A_3 = \rho |A_4| \exp[i\psi_4], \]  

(60)

and the phase of \( A_3 \) is proportional to \( \psi_4 \), not \(-\psi_4\). Uniform phase shifts in \( A_4 \) will not cancelled by \( A_3 \). On the other hand, for an externally-pumped phase-conjugate reflection,

\[ A_3 = \rho' \exp[i(\psi_1 + \psi_2)] A_4^*, \]  

(61a)

or

\[ A_3 = \rho |A_4| \exp[-i\psi_4], \]  

(61b)

where \( \rho' \) is the complex reflection coefficient for the externally-pumped PCM, and \( \rho = \rho' \exp[i(\psi_1 + \psi_2)] \). The phase of \( A_3 \) is proportional to \(-\psi_4\), so that both uniform and relative phase shifts of \( A_4 \) will be cancelled for this PCM.

The concepts of equation (60) were demonstrated using a two-beam interferometer that had a self-pumped PCM in one arm and a regular mirror in the other arm. When a uniform phase shift produced by a Babinet-Soleil compensator was introduced into either arm of the interferometer, the resulting fringe pattern shifted but always remained visible. When a nonuniform phase shift, such as produced by a frosted plastic plate, was placed into the PCM arm, the fringes at first disappeared and then reappeared, slightly shifted in space because of the finite thickness of the plate. When the same plate was placed in front of the regular mirror, the fringes disappeared completely. The reappearance of the fringes indicates that the relative wavefront distortions were cancelled by the PCM, while the fringe shift indicates that the uniform phase shift was not cancelled.
by either mirror. If the self-pumped PCM was replaced with an externally-pumped PCM, the fringe pattern did not shift when either the Babinet-soleil compensator or the frosted plastic plate was placed in the PCM arm. The uniform phase shift due to the finite thickness of the compensator or glass plate was cancelled by the phase-conjugate reflection.

Using these self-pumped PCM's as mirrors in a two-beam interferometer greatly simplifies alignment procedures compared to regular mirrors. The two beams from the separate arms of the interferometer must overlap at the beamsplitter/thin film so that the spacing of the fringes of equal thickness is maximized. This overlap can be difficult to attain in a regular interferometer. In a PCI, this overlap condition is guaranteed regardless of the relative positions of the two PCM's because a phase-conjugate reflection travels exactly back along its incident path. This is equivalent to saying that phase differences between the two arms caused by tilt in the wavefronts or nonuniform air turbulence in the beam paths are cancelled by the phase-conjugate reflection because both effects are relative phase shifts. On the other hand, phase differences between the two arms due to path length differences are not cancelled because these are uniform phase shifts. However, as long as the path length difference is less than the coherence length of the light source, and all the fringes of equal path length are incident on the detectors, alignment is not a problem. Further alignment simplification results from a self-pumped PCM being self-aligning because the pump beams are generated internally. The PCM also has a wide field-of-view so that
the angle between the crystal axis and input beam is not critical as long as the crystal axis points along the same direction as the input beam.

All of these qualities - relative phase shift cancellation, uniform phase shift preservation, and ease of alignment make the use of self-pumped PCM's very desirable for thin film interferometry. An experiment using two self-pumped PCM's in a thin film interferometer is described in the next section.
SECTION III: EXPERIMENT AND RESULTS

The two-beam interferometer is set up in the manner suggested in Section I, Fig. 1. As shown in Fig. 7, two self-pumped BaTiO$_3$ photorefractive crystals replace the conventional end mirrors, and a single-layer film deposited on a glass substrate is placed at the BS 2 position. A piezomirror, driven by a 4 - 10 Hz sinusoidal signal of 500V from a Burleigh high-voltage source and Hewlett-Packard function generator, is inserted into one arm of the interferometer in order to produce a time-varying phase difference between the two arms. A 2 mW helium neon laser, polarized parallel to the plane-of-incidence of the film, is isolated from optical feedback by an acousto-optic modulator driven by a 40 MHz signal. A 5 mm aperture allows only the first diffracted order from the modulator to enter the interferometer. The input beam's amplitude is then divided by the thin film sample, and the resulting two beams go to each PCM. After being reflected from the PCM's, the two beams recombine at the thin film sample and are detected at ports 1 and 2 by silicon photodetectors. BS 1 samples the phase-conjugate beams for port 1. The detector signals are amplified by a transimpedance circuit based on an OP-27 operational amplifier and then input to either an oscilloscope or to a voltmeter.

Detectors at ports 1 and 2 respond to the time-varying intensities according to equation (33)

\[
D_1 = (\alpha^2 + \beta^2 + 2\alpha\beta\cos[\Psi + \phi(t)])
\]  \hspace{1cm} (62)

\[
D_2 = \delta^2 + \gamma^2 - 2\delta\gamma\cos[\Psi - \Delta + \phi(t)]
\]  \hspace{1cm} (63)
Figure 7: Phase-conjugate interferometer for thin film analysis.

where $\alpha$, $\beta$, $\delta$, and $\gamma$ are defined as

$$\begin{align*}
\alpha &= \eta R_1 |M|/m_{11}^2 \\
\beta &= \eta R_2 |m_{12}|^2/m_{11}^2 \\
\delta &= R_1 |M|m_{21}/m_{11}^2 \\
\gamma &= R_2 |M|m_{12}/m_{11}^2,
\end{align*}$$

(64) (65)

and $\phi(t)$ is the phase change introduced by the piezomirror. The factor of $\eta$ in equation (64) represents the relative efficiency of the
detectors and the reflectivity of BS 1. The five parameters \( \alpha, \beta, \delta, \gamma, \) and \( \Delta \) remain to be determined so that the film's optical properties can be found.

Accordingly, if each PCM is blocked in turn and the outputs of each detector are measured, the following voltages are obtained from each detector:

<table>
<thead>
<tr>
<th>Block PCM 1 (( R_1 = 0 ))</th>
<th>Block PCM 2 (( R_2 = 0 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Detector 1</td>
<td>( \beta^2 )</td>
</tr>
<tr>
<td>Detector 2</td>
<td>( \gamma^2 )</td>
</tr>
<tr>
<td></td>
<td>( \alpha^2 )</td>
</tr>
<tr>
<td></td>
<td>( \delta^2 )</td>
</tr>
</tbody>
</table>

Different ratios of these measurements can then be analyzed to extract the various matrix elements. However, the only combination of \( \alpha, \beta, \delta \) and \( \gamma \) that requires no calibration of the PCM reflectivities \( R_1 \) and \( R_2 \), or the output ports' relative efficiency \( \eta \) is

\[
\frac{\beta\delta}{\alpha\gamma} = \frac{m_{12}m_{21}}{|M|} \tag{66}
\]

This ratio provides a single independent parameter. Two more are required to determine the three unknowns of the film, \( n, \kappa, \) and \( d \).

The second measurement is \( \Delta \). There are several ways to obtain this quantity, as discussed in Section IV, but the method chosen for this work consisted of measuring the phase angle of the ellipse formed by projecting the detector signals onto the x-y inputs of an oscilloscope. This phase angle equals the phase difference between the two signals. In Fig. 8 it is seen that for two sinusoidal signals, \( \gamma \)
This measurement requires no calibration of the detector’s signals because both A and B are measured on the y-axis, or by detector 1.

Finally, a third measurement is needed. A measure of the film’s transmission, given by

\[ T = \frac{(1/m_{12}^2)n_3\cos\theta_3/n_1\cos\theta_1}{}, \]  

(68)
is sufficient, where \( n_1 \) and \( n_3 \) are the refractive indices of the incident medium and film substrate, and \( \theta_1 \) and \( \theta_3 \) are the incident angle and the substrate angle, respectively. However, it has been assumed until now that the beam intensities are being measured in the substrate material. Transmission from the substrate back into air must be accounted for according to the Fresnel transmission coefficient for \( p \)-polarized light:

\[
    t_p = \frac{(2n_3 \cos \theta_3)}{(n_3 \cos \theta_4 + n_4 \cos \theta_3)},
\]

where the subscript 4 denotes air. The actual transmission through the film/substrate system is then

\[
    T = \frac{1}{m_1^2} |t_p|^2
\]

if media 1 and 4 are identical. The existing set-up was easily modified by moving one of the detectors to a position following the film with respect to the input beam. A ratio of the detector's signal with the sample first in place and then removed yields \( T \).

Three independent measurements which require no knowledge of absolute beam intensities or mirror reflectivities now exist. A multidimensional minimization routine based on Powell's method fit the measured values of \( \phi_{12} + \phi_{21} \), \( m_{12}m_{21}/|M| \), and \( m_{11}^2 \) to \( n, \kappa, \) and \( d \) of the film. It is assumed that \( n_1, n_3, n_4, \theta_1, \theta_3, \) and \( \theta_4 \) are known.

Using this phase-conjugate interferometry technique, a thin film sample of vanadium oxide, \( V_2O_5 \), was analyzed. By blocking each PCM in
turn, the following values for $a^2$, $\beta^2$, $\delta^2$, and $\gamma^2$ were obtained from the voltmeter:

\[
\begin{align*}
\alpha^2 &= 4.60 \text{ V} \\
\delta^2 &= 2.49 \text{ V} \\
\beta^2 &= 0.049 \text{ V} \\
\gamma^2 &= 2.13 \text{ V}
\end{align*}
\]

which gives $\beta\delta/\alpha\gamma = 0.108$. The phase difference between the two ports was measured from the ellipse shown in Fig. 9, yielding $A = 0.185 \text{ V}$, $B = 1.0 \text{ V}$, and $\Delta = +/- 0.185$. A transmission measurement gave $T = 0.797$, or $m_{11}^2 = 2.35$. With these values as inputs, the minimization program found $n = 2.1 +/- 0.06$, $\kappa = 0.068 +/- 0.004$, and $d = 79 +/- 12 \text{ nm}$. Initial estimates for $n$, $\kappa$, and $d$ were based on ellipsometry and profilometry measurements which resulted in $n = 2.0$, $\kappa = 0.1$, and $d = 82 +/- 30 \text{ nm}$. Inaccuracy in the interferometry measurements are attributed to the difficulty in reading the exact voltage of the oscilloscope traces. The measurements were repeated several times with less than a 5% variation in recorded values. These results are discussed in more detail in the following section.
Figure 9: Ellipse formed from the signals at Ports 1 and 2, displayed on the x-y inputs of an oscilloscope. The thin film sample was V\textsubscript{2}O\textsubscript{5}. A = 0.185V, B = 1.0V.
SECTION IV: DISCUSSION

The claim was made in the introduction that phase-conjugate interferometric thin film analysis is superior to conventional interferometric techniques. This claim was based on improved temporal stability, simplification of optical alignment, and reduction in wavefront distortions with a phase-conjugate interferometer. A further claim was made that this technique has many advantages over common ellipsometry. The justification for these claims is given below.

Temporal stability improves for a PCI for the same reason that optical alignment is easier. The interfering beams will overlap at BS 2 with zero phase difference for nonuniform phase shifts, or in other words, fringes of equal thickness will always have infinite spatial periodicity. As mentioned earlier, these phase shifts are caused by such phenomena as small-scale nonuniform air turbulence and nonparallel mirrors (both a problem in conventional interferometry). It was noted in the laboratory that the output intensity of an interferometer constructed with regular mirrors fluctuated much more with table vibrations or movements in the room than did the output intensity of a PCI.

The cancellation of relative wavefront distortions due to the thin film sample occurs for beams reflected and transmitted back to port 1. Examination of beam propagation through the interferometer of Fig. 7 reveals that the beam initially reflected by the sample to PCM 2 travels back along the same path to port 1, as does the beam that is
initially transmitted through the sample to PCM 1 and then transmitted back through to port 1. As was pointed out in Section II, a phase-conjugate beam from a self-pumped PCM will cancel any relative wavefront distortions after retracing its path through the distorting medium. On the other hand, the outputs at port 2 do not have wavefront distortions completely cancelled when phase-conjugated from each PCM because the beams do not follow the same path back to port 2. The beam that is initially reflected from the film to PCM 2 is then transmitted through the film to port 2, and the beam that is initially transmitted through the film to PCM 1 is reflected from the film to port 2.

Having demonstrated the advantages of a PCI compared to a regular interferometer, comparisons must now be made of its performance relative to an ellipsometer. First, the ellipsometer has several calibration requirements including determination of the angle-of-incidence, sample plane tilt, waveplate transmission and retardation, and location of the transmission axis of two polarizers relative to the sample plane-of-incidence. Second, an ellipsometer measures only two parameters, \( \delta \) and \( \psi \), that characterize the effect that a film has on an incident beam's polarization upon reflection from the film\(^{28} \). Only two unknowns of the film, for example, \( n \) and \( k \), can be determined from these two parameters. The thickness \( d \) of the film must be measured separately using a method such as profilometry, unless the ellipsometer has the capability of varying the angle-of-incidence or the incident wavelength, thereby providing two more
independent parameters with which to determine \( n, \kappa, \) and \( d \) of the film.

But these additional measurements can cause problems of their own: \( n \) and \( \kappa \) are wavelength dependent, the ellipsometer uses a half-waveplate which is also wavelength dependent, and the values estimated for \( n, \kappa, \) and \( d \) in an ellipsometer program such as McCrackin’s can be very sensitive to small changes in incident angle. One would potentially have to recalibrate the ellipsometer every time the incident angle or incident wavelength was changed. And complicating this calibration procedure is the ellipsometer’s sensitivity to tilt in the sample plane which causes a corresponding tilt in the film’s plane-of-incidence. The transmission axes of the waveplate and polarizers are defined with respect to this plane, so that if the sample plane is not perfectly horizontal, the change in polarization of the reflected beam will not be measured correctly and errors in \( \delta \) and \( \psi \) result.

One can see the problem of having to calibrate the various optical components, incident angle, incident wavelength, and sample plane tilt of an ellipsometer. Furthermore, once these calibrations are made, one is still faced with the ambiguities in \( n, \kappa, \) and \( d \) which can result from a single measurement of \( \delta \) and \( \psi \). For example, the optical thickness of the \( \text{V}_2\text{O}_5 \) sample, about \( \lambda/4 \) for the source radiation, falls in the region of highest sensitivity for \( \delta \) and \( \psi \) (where ambiguities are smallest).\(^{29}\) As seen in the \( \delta-\psi \) map for this sample in Fig. 10, for varying values of \( n \) and \( d \), \( \delta \) and \( \psi \) are well-separated, with a minimum change in \( \psi \) of half-of-a-degree for an 8%
change in \( d \). Yet a change in \( \kappa \) of 20\%, from 0.05 to 0.06, changes \( \psi \) less than a tenth-of-a-degree. Even though the verniers for an ellipsometer's polarizers are certainly that accurate, the incident angle and sample plane tilt on less expensive ellipsometers cannot be specified that well. So \( \kappa \) can be difficult to measure with an ellipsometer, even in the most sensitive region of the \( \delta-\psi \) map. And if the sample thickness is near \( \lambda/2 \), ambiguities in both \( \delta \) and \( \psi \) become largest, making it difficult to determine \( n \) and \( d \) also.

![Figure 10: \( \delta-\psi \) map for ellipsometry of a V\(_2\)O\(_5\) thin film sample.
Thickness was varied from 86nm - 93nm in increments of 1 nm; \( n = 1.9, 2.0, 2.1; \kappa = 0.05 - 0.09 \) in 0.01 increments.](image)
In interferometry, only calibration of the angle-of-incidence and incident polarization is required; there are no second polarizers or wavelength sensitive optical components. More importantly, \( n, \kappa, \) and \( d \) can be obtained from a single test set-up which yields three independent parameters rather than two. Finally, the measurements can be performed with inexpensive, low power light sources, a single axis piezomirror, two photorefractive crystals, two detectors, and an oscilloscope while still achieving the equivalent accuracy of a much more expensive ellipsometer.

However, ambiguities in the values of \( n, \kappa, \) and \( d \) also exist for interferometry. Very thin films with large absorption coefficients can exhibit multiple solutions for \( n, \kappa, \) and \( d \) just as ellipsometry does.\(^2,3^0\) Suggestions for overcoming this problem include performing measurements at multiple incident angles and varying wavelengths, using symmetric refractive index bounding layers, and immersing the film sample in liquids of different refractive index. These tactics would also work for ellipsometry to some degree, but interferometry still yields one more parameter than does ellipsometry for each measurement set-up. Furthermore, the interferometric ambiguities can be substantially reduced by measuring the sign of \( \Delta \), as it is periodic with respective to the effective film thickness, \( \beta = (2\pi/\lambda)nd\cos\theta \).\(^3^0\) Knowledge of the sign of \( \Delta \) can reduce the ambiguities by a factor of two.

The accuracy of \( \Delta \) can further be improved by measuring the phase difference between \( D_1 \) and \( D_2 \) directly. For example, one can display
the individual detector outputs on an oscilloscope which is time-
synched to the piezomirror driver. The time delay between the two
signals can then be measured, resulting in an order of magnitude
increase in sensitivity over the original phase angle measurement. In
addition, improvements in accuracy for $\alpha^2$, $\beta^2$, $\delta^2$, and $\gamma^2$ can be
obtained by employing time-averaging data acquisition techniques using
a personal computer.

Other improvements can be achieved by including in the
computational analysis of the interferometric data the effects of
internal reflections from the film-substrate and film-air interfaces.
It has previously been assumed that these reflections could be ignored
because they were such a small percentage of the total power reflected
or transmitted. Also, the sensitivity of the interferometer would
improve if there was no path length difference between the two arms so
that fewer fringes occurred in the output plane.
SECTION V: CONCLUSION

A phase-conjugate interferometric method of thin film analysis obtains three independent parameters from a single measurement set-up with which to find $n$, $\kappa$, and $d$ of a single layer film. The PCM's are self-aligning, and the test set-up requires little calibration. This technique can be extended to multi-layer film analysis by varying the angle-of-incidence, incident wavelength, or incident polarization for each set of three measurements. It should also be possible to map index-of-refraction profiles because of the many independent parameters available for analysis. Furthermore, by using a slightly different set-up, films deposited on opaque substrates can be analyzed.
LIST OF REFERENCES


