Ellipsometry of anisotropic media

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We examine various reflection ellipsometry arrangements to see what they measure when the reflecting stratification is anisotropic. We find that the simplest polarizer–sample–analyzer ellipsometer can determine the absolute squares and the real parts of $\rho_P = (r_{pp} + r_{sp} \tan P)/(r_{ps} + r_{ss} \tan P)$ or $\rho_A = (r_{pp} + r_{ps} \tan A)/(r_{sp} + r_{ss} \tan A)$, depending on whether the polarizer angle P or the analyzer angle A is varied, the other being held constant. For ellipsometer arrangements that use a compensator or a polarization modulator, the real and the imaginary parts of ρ_A are measured when the compensator or the modulator is placed between the polarizer and the reflecting specimen, while the real and the imaginary parts of ρ_P are determined when the compensator or the modulator is placed between the sample and the analyzer.

INTRODUCTION

Reflection ellipsometry[1,2] of isotropic materials measures the real and the imaginary parts of the complex ratio r_p/r_s. Here we answer the question: What does ellipsometry measure when the reflecting stratification is anisotropic? We may expect the answer to be some ratio involving the four reflection amplitudes r_{pp}, r_{ps}, r_{sp}, and r_{ss}. It turns out that all the configurations considered here measure either $\rho_P = (r_{pp} + r_{sp} \tan P)/(r_{ps} + r_{ss} \tan P)$ or $\rho_A = (r_{pp} + r_{ps} \tan A)/(r_{sp} + r_{ss} \tan A)$, where P and A are the angles between the p direction and the easy axes of the polarizer and the analyzer, respectively.

The essence of all ellipsometric methods is as follows. A polarizer produces a known proportion of in-phase p and s incident waves. The amplitude and the phase of these waves are altered by reflection, in a manner specified by the complex amplitudes r_{pp}, r_{ps}, r_{sp}, and r_{ss}. The reflected light is passed through an analyzer, which combines the components of the orthogonal p and s polarizations along the analyzer easy direction. The intensity then measured by a detector is the result of interference of the p and s components and thus contains information about the relative phases of the reflection amplitudes as well as about their magnitudes.

The simplest ellipsometer arrangement has just the polarizer, the sample, and the analyzer components mentioned above. This arrangement will be discussed first.

POLARIZER–SAMPLE–ANALYZER

Let P be the angle between the polarizer easy axis and the p direction. Then the p and s components of the electric field transmitted by the polarizer are $\cos P$ and $\sin P$ (we remove common factors where these are not experimentally relevant). After reflection, the p and s components are

$$r_{pp} \cos P + r_{sp} \sin P, \qquad r_{ps} \cos P + r_{ss} \sin P. \tag{1}$$

These components are combined by the analyzer. If this is set at angle A to the p direction, the field transmitted by the analyzer is

$$\begin{aligned}(r_{pp} \cos P &+ r_{sp} \sin P)\cos A \\ &+ (r_{ps} \cos P + r_{ss} \sin P)\sin A \\ &= \cos P \cos A[r_{pp} + r_{sp} \tan P \\ &\quad + (r_{ps} + r_{ss} \tan P)\tan A] \\ &= \cos P \cos A[r_{pp} + r_{ps} \tan A \\ &\quad + (r_{sp} + r_{ss} \tan A)\tan P].\end{aligned} \tag{2}$$

Let us define two ellipsometric ratios:

$$\rho_P = \frac{r_{pp} + r_{sp} \tan P}{r_{ps} + r_{ss} \tan P}, \qquad \rho_A = \frac{r_{pp} + r_{ps} \tan A}{r_{sp} + r_{ss} \tan A}. \tag{3}$$

Consider two intensity measurements as analyzer angles A_1 and A_2, and fixed polarizer angle P. Then

$$\frac{I(A_1)}{I(A_2)} = \left(\frac{\cos A_1}{\cos A_2}\right)^2 \left|\frac{\rho_P + \tan A_1}{\rho_P + \tan A_2}\right|^2. \tag{4}$$

Similarly, if two intensity measurements are made at polarizer angles P_1 and P_2, at fixed analyzer angle A, their ratio will be

$$\frac{I(P_1)}{I(P_2)} = \left(\frac{\cos P_1}{\cos P_2}\right)^2 \left|\frac{\rho_A + \tan P_1}{\rho_A + \tan P_2}\right|^2. \tag{5}$$

Measurements of this kind can thus give the absolute squares and the real parts of ρ_P and ρ_A, respectively. The signs of the imaginary parts of ρ_P and ρ_A are not determined.

POLARIZER–COMPENSATOR–SAMPLE–ANALYZER

A compensator is a crystal plate, or arrangement of plates, that produces a known phase difference between two orthogonal components. For example, a wave normally incident onto a uniaxial crystal will split into two orthogonal components that travel in the crystal with phase speeds

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c/n_o and $cn_\gamma/n_o n_e$, where n_o and n_e are the ordinary and the extraordinary refractive indices, respectively, of the crystal, γ is the cosine of the angle between the optic axis and the surface normal, and[3]

$$n_\gamma^{\,2} = n_o^{\,2} + \gamma^2(n_e^{\,2} - n_o^{\,2}). \tag{6}$$

When light traverses a crystal plate of thickness Δz, there results a phase difference of approximately

$$\delta_e - \delta_o \approx (\omega/c)(n_o n_e/n_\gamma - n_o)\Delta z \tag{7}$$

between the components along the e and o directions in the crystal. (Strictly speaking, the direction of $\mathbf{E}_e$ is not in the plane of the crystal surface unless γ is zero or unity, but the electric fields $\mathbf{E}_o$ and $\mathbf{E}_e$ are orthogonal, and $\mathbf{E}_o$ does lie in the plane of the crystal surface, so we should refer to the o direction and one perpendicular to it. The latter is loosely referred to as the e direction, and correctly so when the optic axis is parallel to the crystal surface.) Relation (7), although long accepted as exact (see, for example, Ref. 4, Sec. 14.4.3), is approximate because it does not allow for multiple reflections inside the crystal plate.[5] A real crystal plate also does not transmit all the light; Ref. 5 gives the actual transmission amplitudes t_o and t_e from which the transmission properties of arbitrarily polarized light may be obtained.

We now consider the polarizer–compensator–sample–analyzer ellipsometer.[6] After passing through the polarizer set at angle P to the p direction, the light passes through a compensator with its o direction at angle C to the p direction. The components along the o and e directions are

	Along o	Along e
After polarizer	$\cos(C - P)$	$-\sin(C - P)$
After compensator	$t_o \cos(C - P)$	$-t_e \sin(C - P)$

(8)

We now resolve along the p and s directions. The electric-field amplitudes along p and s are

$$\begin{aligned} E_p &= t_o \cos C \cos(C - P) + t_e \sin C \sin(C - P), \\ E_s &= t_o \sin C \cos(C - P) - t_e \cos C \sin(C - P). \end{aligned} \tag{9}$$

After reflection these become

$$r_{pp}E_p + r_{sp}E_s, \qquad r_{ss}E_s + r_{ps}E_p. \tag{10}$$

These s and p components are combined by the analyzer, set at angle A to the p direction. The field transmitted by the analyzer is thus

$$(r_{pp}E_p + r_{sp}E_s)\cos A + (r_{ss}E_s + r_{ps}E_p)\sin A. \tag{11}$$

The intensity measured is proportional to the absolute square of this quantity. In null ellipsometry this is made zero (in practice minimized) by adjustment of the polarizer, compensator, and analyzer angles P, C, and A, respectively. We define the ratio $\tau = t_e/t_o$ (the complex variable τ can also represent the effect of compensators other than the simple crystal plate discussed above) and the complex angle $D = D_r + iD_i$ by means of

$$\tan D = \tau \tan(C - P). \tag{12}$$

Then, from Eqs. (9), the ratio of E_s to E_p can be written as

$$E_s/E_p = \tan(C - D). \tag{13}$$

It follows that the zero of expression (11) occurs when

$$\rho_A = \tan(D - C). \tag{14}$$

Thus a null setting of the polarizer–compensator–sample–analyzer ellipsometer determines the real and the imaginary parts of $\rho_A = (r_{pp} + r_{ps} \tan A)/(r_{sp} + r_{ss} \tan A)$. In the isotropic case Eq. (14) reduces to [compare Eq. (3.33) of Ref. 2]

$$r_p/r_s = \tan A \tan(D - C). \tag{15}$$

POLARIZER–SAMPLE–COMPENSATOR–ANALYZER

In this ellipsometer the electric-field amplitudes along the s and p directions after the polarizer are $\cos P$ and $\sin P$, respectively, and after reflection these become

$$\begin{aligned} E_p' &= r_{pp} \cos P + r_{sp} \sin P, \\ E_s' &= r_{ss} \sin P + r_{ps} \cos P. \end{aligned} \tag{16}$$

The compensator o direction is at angle C to the p direction, so after the compensator the components along the o and e directions are

$$t_o(E_p' \cos C + E_s' \sin C), \qquad t_e(E_s' \cos C - E_p' \sin C). \tag{17}$$

Thus the electric field transmitted by the analyzer set at angle A to the p direction is

$$\begin{aligned} &t_o(E_p' \cos C + E_s' \sin C)\cos(C - A) \\ &\qquad - t_e(E_s' \cos C - E_p' \sin C)\sin(C - A). \end{aligned} \tag{18}$$

We again define $\tau = t_e/t_o$, with the understanding that complex number τ can represent any compensator, and we introduce the complex angle D' by means of

$$\tan D' = \tau \tan(C - A). \tag{19}$$

The field (18) is zero when

$$\rho_P = \tan(D' - C). \tag{20}$$

A null setting thus determines the real and imaginary parts of $\rho_P = (r_{pp} + r_{sp} \tan P)/(r_{ps} + r_{ss} \tan P)$. In the isotropic case, Eq. (20) reduces to [compare Eq. (3.55) of Ref. 2]

$$r_p/r_s = \tan P \tan(D' - C). \tag{21}$$

POLARIZER–MODULATOR–SAMPLE–ANALYZER

We turn now to polarization-modulation ellipsometry,[7,8] in which the polarization state of the light is varied sinusoidally, with synchronous (lock-in) detection of the intensity. The configuration used by Beaglehole[8] has been discussed already.[9] Here we consider a more general configuration, in which the polarizer and the analyzer have arbitrary orientation. When the polarizer pass direction is at angle P

to the p direction, the p and s components of the field passing through the polarizer are cos P and sin P, respectively. The birefringent modulator has its o and e directions along the p and s directions. After passing through the modulator, the p and s components are cos P and $\exp(i\delta)(\sin P)$, respectively, where

$$\delta(t) = M \sin(\Omega t). \tag{22}$$

M is the maximum phase shift induced by the modulator, and $\Omega/2\pi$ is the modulation frequency. [We omit a correction factor, to multiply $\exp(i\delta)$, which would allow for different transmission probabilities through the modulator of the orthogonal components, because in practice this is close to unity.] After reflection, the p and s components are

$$\begin{aligned} E_p &= r_{pp} \cos P + r_{sp} \exp(i\delta)\sin P, \\ E_s &= r_{ps} \cos P + r_{ss} \exp(i\delta)\sin P. \end{aligned} \tag{23}$$

The analyzer pass direction is at angle A to the p direction. Thus the final field amplitude is

$$\begin{aligned} E_p \cos A + E_s \sin A = \cos P \cos A(r_{sp} + r_{ss} \tan A) \\ \times [\rho_A + \exp(i\delta)\tan P]. \end{aligned} \tag{24}$$

For fixed polarizer and analyzer settings the intensity is thus proportional to

$$\rho_r^2 + \rho_i^2 + 2(\rho_r \cos\delta + \rho_i \sin\delta)\tan P + \tan^2 P, \tag{25}$$

where ρ_r and ρ_i are the real and the imaginary parts, respectively, of ρ_A. Now

$$\begin{aligned} \cos[M \sin(\Omega t)] &= J_0(M) + 2\sum_{n=1}^{\infty} J_{2n}(M)\cos(2n\Omega t), \\ \sin[M \sin(\Omega t)] &= 2\sum_{n=0}^{\infty} J_{2n+1}(M)\sin[(2n+1)\Omega t], \end{aligned} \tag{26}$$

so the DC, Ω, and 2Ω parts of the intensity are proportional, respectively, to

$$\begin{aligned} &\rho_r^2 + \rho_i^2 + 2\rho_r J_0(M)\tan P + \tan^2 P, \\ &4\rho_i J_1(M)\tan P \sin(\Omega t), \\ &4\rho_r J_2(M) \tan P \cos(2\Omega t). \end{aligned} \tag{27}$$

Thus polarization–modulation ellipsometry, with the modulator placed between the polarizer and the sample, measures the real and imaginary parts of $\rho_A = (r_{pp} + r_{ps} \tan A)/(r_{sp} + r_{ss} \tan A)$.

POLARIZER–SAMPLE–MODULATOR–ANALYZER

Finally, we look at the ellipsometer configuration in which the modulator is placed after the sample. The p and s components after reflection are

$$\begin{aligned} E_p' &= r_{pp} \cos P + r_{sp} \sin P, \\ E_s' &= r_{ps} \cos P + r_{ss} \sin P. \end{aligned} \tag{28}$$

After the modulator (aligned with its o and e directions along the p and s directions), the p and s components become E_p' and $E_s'\exp(i\delta)$, respectively, with $\delta(t)$ given by Eq. (22) as before. The field amplitude passing through the analyzer is thus

$$\begin{aligned} E_p' \cos A + E_s' \exp(i\delta)\sin A \\ = \cos P \cos A(r_{ps} + r_{ss} \tan P)[\rho_P + \exp(i\delta)\tan A]. \end{aligned} \tag{29}$$

For fixed polarizer and analyzer settings, the intensity is proportional to (here we write $\rho_P = \rho_r + i\rho_i$)

$$\rho_r^2 + \rho_i^2 + 2[\rho_r \cos\delta + \rho_i \sin\delta]\tan A + \tan^2 A, \tag{30}$$

and so the DC, Ω, and 2Ω signals are proportional, respectively, to

$$\begin{aligned} &\rho_r^2 + \rho_i^2 + 2\rho_r J_0(M)\tan A + \tan^2 A, \\ &4\rho_i J_1(M)\tan A \sin(\Omega t), \\ &4\rho_r J_2(M)\tan A \cos(2\Omega t). \end{aligned} \tag{31}$$

Thus, when the modulator is placed between the sample and the analyzer, the polarization–modulation ellipsometer determines the real and the imaginary parts of $\rho_P = (r_{pp} + r_{sp} \tan P)/(r_{ps} + r_{ss} \tan P)$.

CONCLUSION

Ellipsometry of anisotropic media determines the complex ratios ρ_P or ρ_A. Measurement (with compensator or modulator) at three polarizer or three analyzer settings is sufficient to determine three ratios of the reflection amplitudes, for example r_{pp}/r_{ss}, r_{ps}/r_{ss}, and r_{sp}/r_{ss}. Each of the individual reflection amplitudes carries the same arbitrary phase, which depends on conventions such as the choice of origin, and is not determined experimentally.

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