An error evaluation technique for the angle of incidence in a rotating element ellipsometer using a quartz crystal

1999 J. Phys. D: Appl. Phys. 32 2246

(http://iopscience.iop.org/0022-3727/32/17/315)

J. Phys. D: Appl. Phys. **32** (1999) 2246–2249. Printed in the UK PII: S0022-3727(99)97060-2

An error evaluation technique for the angle of incidence in a rotating element ellipsometer using a quartz crystal

Y F Chao, M W Wang and Z C Ko

National Chiao Tung University, Electro-optical Engineering, Hsinchu, Taiwan

E-mail: `yschao@cc.nctu.edu.tw`

Received 28 August 1998, in final form 14 May 1999

Abstract. The error in the angle of incidence for a rotating element ellipsometer is evaluated using a uniaxial quartz crystal. At a fixed angle of incidence with respect to the surface of reflection, the ratio of reflectance in the parallel to that in the perpendicular electromagnetic field is measured by rotating the quartz crystal through a full cycle. We determine the deviation in the angle of incidence by comparing the experimentally measured reflectance ratio to its calculated value.

1. Introduction

The reflection technique has been widely used to determine the refractive indices of materials, even in powder form [1–4]. It was proved [4] that the ratio of reflectance in the parallel (R_p) to that in the perpendicular (R_s) electromagnetic field is insensitive to the condition of the surface. The ellipsometric parameter tan ψ is the square root of the reflectance ratio [5]; it can be easily obtained using ellipsometric measurements. Ellipsometry is a powerful technique for determining the optical properties of materials, such as isotropic multilayer thin films [5] and anisotropic crystals [6]. Rotating-element ellipsometry [7] is now known to be more amendable to automation than the conventional null ellipsometry. It has been known [8] that the beam deviation in the rotating-element ellipsometry can cause serious errors in the angle of incidence (AI), which is one of the crucial angles in ellipsometric measurements. Although both McCrackin *et al* [9] and Chao *et al* [10] have systematically align the azimuthal angles of the polarizer and analyser to the incident plane in an ellipsometric system with a high degree of accuracy, no one has dealt with AI deviation.

Because the analytical Fresnel reflection coefficients for uniaxial crystals have been developed explicitly [11], it is possible to calculate the ellipsometric parameter ψ as a function of the azimuthal angle for a known uniaxial crystal at a given incident angle. For aligning an incident angle in a PSA ellipsometry [12], we measure the ellipsometric parameter ψ of a quartz crystal by rotating it around its normal line for a full cycle. A deviation in AI can be clearly observed by comparing the improved values of ψ with its calculated values. Thus AI deviation can be determined by fitting the measured parameter to its calculated value. Because of this evaluation, we are able to use a dichroic sheet polarizer to substitute a prism polarizer. Taking the AI deviation into consideration, the deduced refractive index of glass (BK7) is comparable to that specified in the vendor's catalogue.

According to our analysis, only a uniaxial crystal with small difference between the ordinary (n_o) and extraordinary (n_e) refractive indices can resolve the small error of AI. This conclusion is borne out numerically in this paper. We have also applied this technique to measure the angle between the normal to a cleavage plane and the optical axis of a yttrium orthovanadate (YVO4) crystal to explore the feasibility of this PSA system.

2. The reflectance ratio of a uniaxial crystal

The reflected χ_r and incident χ_i polarization states are related by [6]

$$\chi_r = \frac{(r_{sp}/r_{ss}) + \chi_i}{(r_{pp}/r_{ss}) + (r_{ps}/r_{ss})\chi_i} \qquad (1)$$

where r_{xy} is the Fresnel reflection coefficient for the parallel (p, i.e. x) and perpendicular (s, i.e. y) polarizations. According to [11], we summarize the analytical expressions for these Fresnel reflection coefficients for uniaxial crystals in appendix. The complex pseudoreflectance ratio was defined [6] as $\langle\rho\rangle = \chi_i/\chi_r$ for anisotropic media, while in general ρ is defined as [5]

$$\rho = \tan\psi\, e^{i\Delta}$$

thus

$$\tan^2\psi = \left|\frac{\chi_i}{\chi_r}\right|^2. \qquad (2)$$

0022-3727/99/172246+04$30.00

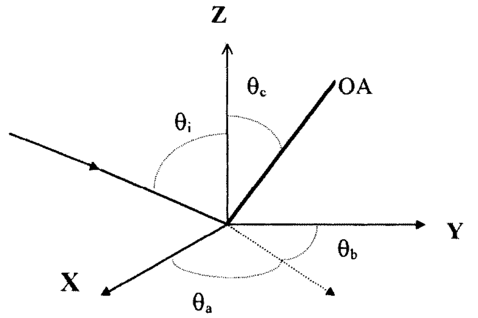

Figure 1. The reflection geometry: θ_i is the incident angle, the xy plane is the reflecting face of the crystal, the zx plane is the incident plane, the z-axis is the normal line. OA is the optical axis of the crystal.

Since the cross terms vanish in an isotropic medium, $\tan\psi$ [5] equals $|r_{pp}/r_{ss}|$, which is the conventional expression for the ellipsometric parameter. The reflection geometry for a uniaxial crystal is shown in figure 1. A simple model for anisotropic crystals was proposed by Aspnes [13]: the measured ellipsometric parameters for a particular θ_a equal those of the effective isotropic sample whose refractive index is given by its dielectric tensor projection onto the sample surface along the incident direction. This implies that

$$\tan^2\psi = \frac{I_{rp}}{I_{rs}} \qquad (3)$$

where I_{rp} represents the reflected intensity parallel to the incident plane and I_{rs} represents the reflected intensity perpendicular to the incident plane,for $P = 45°$, i.e. $\chi_i = 1$. According to equation (3), one can obtain $\tan\psi$ simply by measuring the reflected intensities I_{rp} and I_{rs}. If the optical axis of a non-absorbent uniaxial crystal is parallel to the reflection surface, i.e. $\theta_c = 90°$, then the ellipsometric parameter ψ can be characterized by a twofold symmetry with respect to the azimuthal angle θ_a. Since we are only interested in determining the AI in a PSA ellipsometry, we simulate the ellipsometric parameter function $\psi(\theta_a)$ for a uniaxial crystal with n_o and n_e as its ordinary and extraordinary refractive indices, respectively. Furthermore, we assume the optical axis of the sample crystal is parallel to the reflection surface so as to obtain the twofold symmetry for comparison.

Two types of crystal are simulated to examine their resolving power in AI. The difference of one crystal's ordinary and extraordinary refractive indices is about one order of magnitude lower than that of the other crystal. The function $\psi(\theta_a)$ is simulated for $\chi_i = 1$, i.e. $P = 45°$, and optimized [12] by $\chi_i = -1$, i.e. $P = -45°$, to eliminate the error caused by the misalignment of the polarizer; according to equation (3), one can obtain

$$\tan\psi = \left[\left.\frac{I_{rp}}{I_{rs}}\right|_{p=45°}\left.\frac{I_{rp}}{I_{rs}}\right|_{p=-45°}\right]^{1/4}. \qquad (4)$$

The angle of incidence θ_i in figure 1 is set to be 45° and 44.94° for quartz crystal ($n_o = 1.544$ and $n_e = 1.553$) and 45° and 44.5° for yttrium orthovanadate crystal (YVO4, $n_o = 1.9929$, $n_e = 2.2154$), as shown in figures 2(a) and 2(b), respectively.

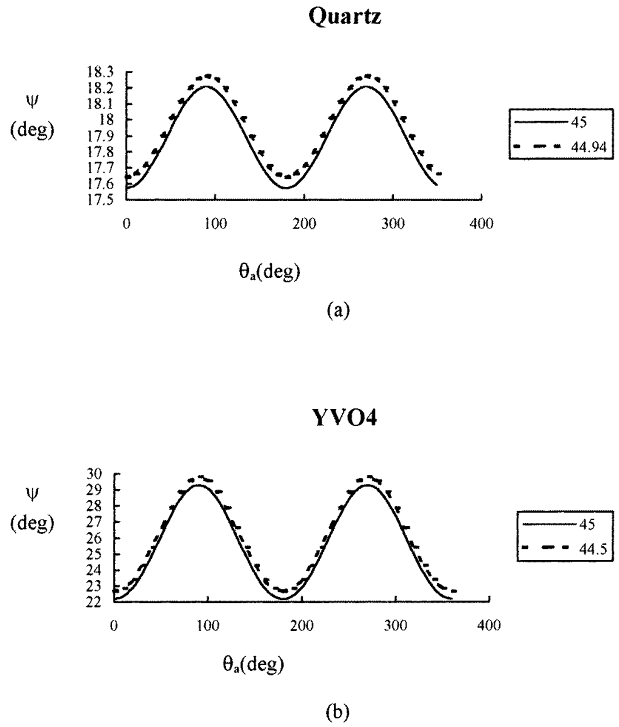

Figure 2. The numerically simulated ψ as a function of azimuthal angle of θ_a while $\theta_c = 90°$: (a) quartz crystal, $n_o = 1.544$, $n_e = 1.553$ at incident angles of 45° (full curve) and 44.94° (dashed curve); (b) yttrium orthovanadate crystal, $n_0 = 1.9929$, $n_e = 2.2154$ at the incident angles of 45° (full curve) and 44.5° (dashed curve).

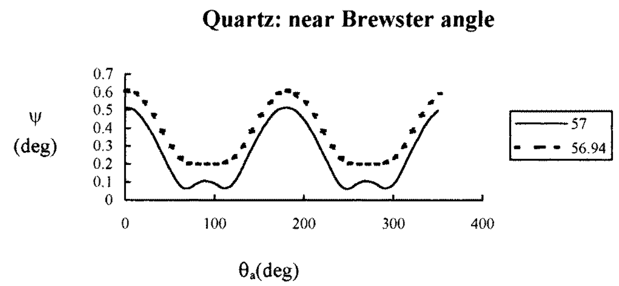

Figure 3. The numerically simulated ψ as a function of azimuthal angle of θ_a while $\theta_c = 90°$, for quartz crystal of $n_o = 1.544$, $n_e = 1.553$ at incident angles of 57° (full curve) and 56.94° (dashed curve), respectively.

These two figures indicate that the resolving power of quartz crystal is about one order of magnitude higher than that of YVO4 crystal. The resolving power is even higher when the AI is at the Brewster angle (the numerical simulated curves for $\theta_i = 57°$ are shown in figure 3). However, the reflectance of the parallel electromagnetic field is too low to be practical for measurement by this intensity ratio technique, especially for a non-absorbent material.

3. Experimental procedures

Figure 4 depicts the experimental set-up. The light (L: HeNe laser) passes through a polarizer (P: dichroic sheet polarizer of extinction ratio 10^{-4}) whose azimuthal angle is set to be

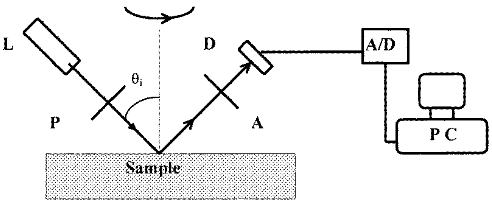

Figure 4. A schematic set-up of the PSA ellipsometer: L, light source (He–Ne laser); P, polarizer; A, analyser; D, detector.

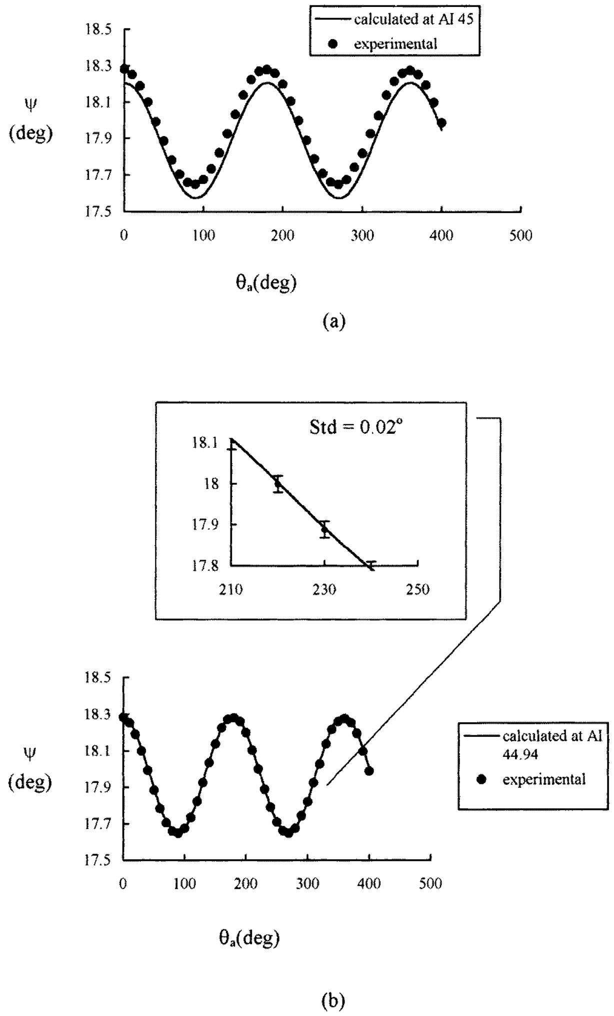

Figure 5. ψ versus θ_a for quartz crystal of $\theta_c = 90°$ with $n_o = 1.544$, $n_e = 1.553$: (a) $\theta_i = 45°$ (full curve, calculated; •, measured) and $\theta_a = 0°$; (b) $\theta_i = 44.94°$ (full curve, calculated; •, measured) and $\theta_a = -1.78°$. Insert: portion of the main plot, the error bars show the standard deviation of the measured value to its calculated value.

45° with respect to the incident plane of the sample. The AI (θ_i) is taken to be 45°. The analyser (A) is mounted on a stepping motor controlled rotator. A sample (quartz crystal, surface flatness $\lambda/4$; BK7 glass, surface flatness $\lambda/4$; and YVO4 crystal-CASIX, surface flatness 2λ) is mounted on a rotatable holder and measured at 10° intervals. All intensities are measured using a power meter (D) (Newport 818-SL), digitized by a multimeter (Keithley 195A), and stored in a PC for calculating the ellipsometric parameters. The reflectance ratios are obtained by the ratio of intensity at $A = 0°$ to that at

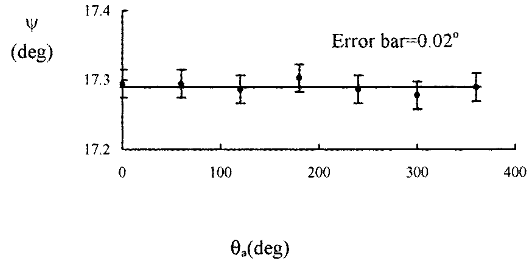

Figure 6. ψ versus θ_a for BK7. The standard deviation is 0.02°. The line indicates the mean value which is 17.29°.

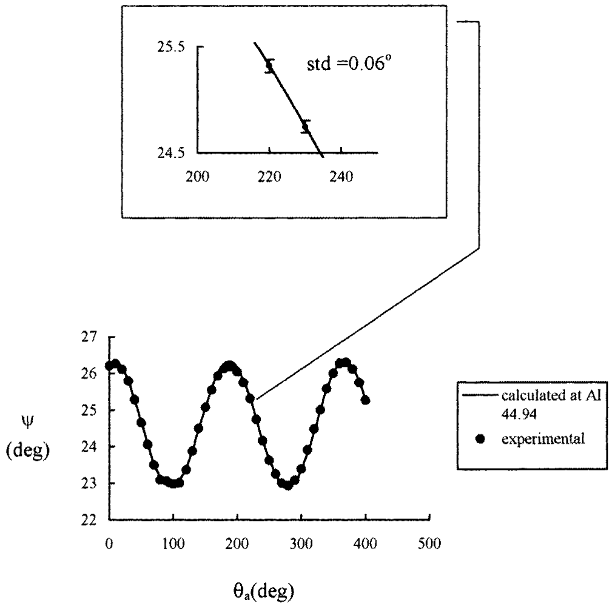

Figure 7. ψ versus θ_a for YVO4. $\theta_c = 136.01°$ with $n_o = 1.9929$ and $n_e = 2.2154$, $\theta_i = 44.94°$ (full curve, calculated; •, measured) and $\theta_a = 7.24°$. Insert: portion of the main plot, the error bars show the standard deviation of the measured value to its calculated value.

$A = 90°$. Prior to the measurements, all the azimuthal angles of the polarizer and the analyser are systematically aligned according to [10] using an optical flat thick platinum plate. Moreover, the reference zeros of the polarizer and analyzer are confirmed using the technique of [12].

4. Results

In parallel to our experiment, we numerically analyse the improved value of ψ, using equation (4) incorporated with the analytical solution of reflection coefficients for uniaxial crystals. The improved ψ is found to be free from the misalignment of the polarizer and differs by only 0.002° if there is a 0.5° misalignment in the analyser. However, the azimuthal deviations of the polarizer and analyser with respect to the incident plane can be as low as 0.005° in the PSA ellipsometric system [12]. Comparing the measured ψ of the quartz crystal with its calculated value, as shown in figure 5(a), we conclude that the systematic error is mainly caused by the deviation of AI. The deviation is found to be $-0.06 \pm 0.01°$ by fitting the experimental data to the calculated values. Furthermore, the azimuthal angle θ_a of the optical axis is found to be $1.78 \pm 0.01^{\circ}$ to the incident plane; as shown in figure 5(b), the standard deviation between the measured value and calculated value is 0.02° after the adjustment of the azimuthal angle. The refractive index of BK7 deduced [5] from $\tan\psi$ (figure 6) is 1.517 ± 0.002 at an incident angle of 44.94° and is 1.521 ± 0.002 at an incident angle of 45°. Since the refractive index of BK7 is 1.515 (Schott: optical glass No 1000), this error evaluation can improve the measurement of glass. The angle (θ_c) between the normal to the cleavage plane and the optical axis of YVO4 crystal was specified as 135° by the vendor (CASIX). Under the corrected incident angle 44.94°, θ_c was obtained to be 136.01° and θ_a (figure 1) to be $7.24 \pm 0.01^{\circ}$, as shown in figure 7.

5. Concluding remarks

The small difference between the ordinary and extraordinary refractive indices of quartz crystal allows us to resolve the deviation in the angle of incident for rotating-element ellipsometry. The other two primary errors in a rotating PSA ellipsometric system, the azimuthal misalignment of polarizer and degradedness of a polarizer, can be reduced by the intensity ratio technique [12]. Since all three primary errors in a rotating PSA ellipsometry can be reduced, we can employ a low cost dichroic sheet polarizer in place of a prism polarizer without loosing its accuracy in a PSA ellipsometric system.

In addition to determining the deviation of incident angle in a rotating element ellipsometry, the following three parameters can be obtained by fitting the measured $\tan\psi$ to the analytic solution of uniaxial crystals: the absolute value of n_o, n_e and the directions of optical axis (θ_a and θ_c) in the laboratory frame. Since the resolving power of the system can be increased as the incident angle moves closer to the Brewster angle (the reflected intensity at 50° will be about 0.4% of the incident intensity), the system can be improved by using a sensitive detector or a higher power light source. It is our interest to extend the current experimental system to measure a material which consists of both linear and circular birefringence.

Acknowledgment

This work is supported by National Science Council of the Republic of China under grant NSC 86-2112-M-009-026.

Appendix. Fresnel reflection coefficients of uniaxial crystals

The reflection geometry is shown in figure 1. The direction of the optical axis is specified by angles θ_a and θ_c relative to the laboratory xyz; if $\vec{c}$ is the unit vector of optical axis, we can express it as

$$\vec{c} = (\alpha, \beta, \gamma)$$

where $\alpha = \cos\theta_a \sin\theta_c$, $\beta = \sin\theta_a \sin\theta_c$ and $\gamma = \cos\theta_c$. Let the incident wavevector be $K\vec{i} + q_1\vec{k}$, where $K = k\, n_i \sin\theta_i$, $q_1 = k\, n_i \cos\theta_i$, for a wavenumber $k = \omega/c$ at an incident angle θ_i. According to [11], we summarized the Fresnel reflection coefficients for uniaxial crystals of ordinary refractive index $n_o = \sqrt{\varepsilon_o}$ and extraordinary refractive index $n_e = \sqrt{\varepsilon_e}$ as follows:

$$r_{ss} = [(q_1 - q_e)AE_y^e - (q_1 - q_e)BE_y^o]/D$$

$$r_{sp} = 2n_i k(AE_x^o - BE_x^o)/D$$

$$r_{pp} = 2q_t[(q_1 + q_e)E_x^o E_y^e - (q_1 + q_o)E_x^o E_y^o]/D - 1$$

$$r_{ps} = 2n_i k(q_e - q_o)E_y^o E_y^e/D.$$

The ordinary and extraordinary modes have wavevector normal components q_o, and q_e related to the medium as

$$q_e = (\sqrt{d} - \alpha\gamma K \Delta\varepsilon)/(\varepsilon_o + \gamma^2 \Delta\varepsilon)$$

$$q_o = \varepsilon_o k^2 - K^2 \qquad q_t = q_1 + K \tan\theta_i$$

where $\Delta\varepsilon = \varepsilon_e - \varepsilon_o$ and

$$d = \varepsilon_o[\varepsilon_e(\varepsilon_o + \gamma^2\Delta\varepsilon)k^2 - (\varepsilon_e - \beta^2\Delta\varepsilon)K^2].$$

The corresponding electric field vectors $\boldsymbol{E}^o$ and $\boldsymbol{E}^e$ are noted as

$$\boldsymbol{E}^o = N_o(-\beta q_o, \alpha q_o - \gamma K, \beta K)$$

$$\boldsymbol{E}^e = N_e(\alpha q_o^2 - \gamma q_e K, \beta\varepsilon_o k^2, \gamma(\varepsilon_o k^2 - q_e^2) - \alpha q_e K)$$

where N_o and N_e are the normalization factors, respectively. For simplicity, we also state the collective parameters as follows:

$$A = (q_o + q_1 + K \tan\theta_i)E_x^o - KE_z^o$$

$$B = (q_e + q_1 + K \tan\theta_i)E_x^e - KE_z^e$$

$$D = (q_1 + q_e)AE_y^e - (q_1 + q_o)BE_y^o.$$

References

[1] Beatie J R 1955 *Phil. Mag.* **46** 235
[2] Taft E A and Philipp H R 1965 *Phys. Rev.* A **138** 197
[3] Tomaselli V P, Rivera R, Edeweard D C and Moeller K D 1981 *Appl. Opt.* **20** 3961
[4] Stagg B J and Charalampopoulos T T 1991 *Appl. Opt.* **30** 4113
[5] Azzam R M A and Bashara N M 1992 *Ellipsometry and Polarized Light* (Amsterdam: North-Holland)
[6] Alonso M I and Garriga M 1995 *Appl. Phys. Lett.* **67** 596
[7] Collins R W 1990 *Rev. Sci. Instrum.* **61** 2029
[8] Zeidler J R, Kohles R B and Bashara N M 1974 *Appl. Opt.* **13** 1938
[9] McCrankin F L, Passaglia E, Stromberg R R and Steinberg H L 1963 *J. Res. Natl Bur. Stand.* A **67** 363
[10] Chao Y F, Wei C S, Lee W C, Lin S C and Chao T S 1995 *Japan. J. Appl. Phys.* **34** 5016
[11] Lekner J 1991 *J. Phys.: Condens. Matter* **3** 6121
[12] Chao Y F, Lee W C, Hung C S and Lin J J 1998 *J. Phys. D: Appl. Phys.* **31** 1968
[13] Aspnes D E 1980 *J. Opt. Soc. Am.* **70** 1275