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An optical in-situ method for layer growth characterization

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An optical method is presented which allows the in-situ characterization of growing films on solid substrates. The technique is based on the changes in reflectivity of p-polarized light at the substrate Brewster angle. The changes in the reflectivity are shown to be large enough to monitor layer growth as well as to reveal the film thickness and the optical constants of the film.

1. Introduction

At present, the characterization of the growth of thin films is being intensively pursued [1,2]. In-situ techniques such as combined XPS (X-ray photoelectron spectroscopy) and LEISS (low energy ion scattering spectroscopy) [3], AES (Auger electron spectroscopy), RHEED (reflection high energy electron diffraction) and ellipsometry are frequently employed. These methods are rather complex and need considerable technical equipment. In the present work we present a simple method which makes use of the relaxation of the Brewster angle condition for p-polarized light [4,5] of a substrate material upon film growth. The applicability of the method will be demonstrated for a film growing on top of a silicon substrate taking into account the experimental limitations through angle divergence and depolarization effects.

For p-polarized light, Si exhibits a reflectivity minimum (pseudo-Brewster angle) at $\varphi_B = 75.637°$ ($\epsilon_{s1} = 15.25$, $\epsilon_{s2} = 0.17$) for illumination with a He/Ne laser ($\lambda = 632.8$ nm). We consider the change in reflectivity R_p at this fixed angle due to the growth of a layer on silicon. To simplify matters, it has been assumed that this layer is homogeneous and grows layer by layer (Frank–van der Merwe growth mechanism) [6].

2. Measurement procedure

The measurement principle is shown in fig. 1. A He/Ne laser (1–3 mW) is used as light source. The output power has to be stabilized and a beam-splitting unit is needed to measure the light intensity accurately enough. The polarizer needs an extinction ratio better than 10^6 for the p-polarized component in order to achieve a high resolution of the reflectivity minimum of the Si substrate. This condition is fulfilled by a Glan–Thompson prism for instance. The detection of the beam reflected at the sample is done by a cooled Si photodiode with an additional low noise amplifier. The configuration allows detection of changes in R_p of the order of 1×10^{-5}. The investigation of the initial stages of deposition is

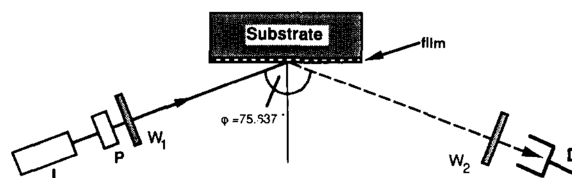

Fig. 1. Schematical diagram of the experimental setup; L: He/Ne-laser; P: polarizer; W1, W2 are the windows into the vacuum chamber; D: detector. Angle of incidence $\varphi_B = 75.637°$ (Brewster angle for the silicon substrate at 632.8 nm with $\epsilon_{s1} = 15.25$ and $\epsilon_{s2} = 0.17$).

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usually made with a lock-in technique for enhanced sensitivity.

The laser beam divergence is about 1 mrad ($\approx 0.06°$) and the polarizer extinction ratio $(R_s/R_p) \approx 10^{-6}$. While passing the polarized light through tempered window glasses into the vacuum chamber a reduction the polarization extinction ratio (stress induced birefringence) to about 10^{-5} has to be assumed.

3. Model consideration

The change in reflectivity R_p of a homogeneous film growing layer by layer can be calculated using Fresnels equations for a three layer system [7] (ambient/film/substrate):

$$rr_p = \frac{r_{01_p} + r_{12_p}\, e^{-i2\phi_1}}{1 + r_{01_p} r_{12_p}\, e^{-i2\phi_1}}, \tag{1}$$

where r_{01_p} and r_{12_p} are the reflectivity coefficients

$$r_{01_p} = \frac{\epsilon_f \cos\varphi_0 + \sqrt{\epsilon_a}\sqrt{\epsilon_f - \epsilon_a \sin^2\varphi_0}}{\epsilon_f \cos\varphi_0 + \sqrt{\epsilon_a}\sqrt{\epsilon_f + \epsilon_a \sin^2\varphi_0}} \tag{2a}$$

and

$$r_{12_p} = \frac{\epsilon_s\sqrt{\epsilon_f - \epsilon_a \sin^2\varphi_0} - \epsilon_f\sqrt{\epsilon_s - \epsilon_a \sin^2\varphi_0}}{\epsilon_s\sqrt{\epsilon_f - \epsilon_a \sin^2\varphi_0} + \epsilon_f\sqrt{\epsilon_s - \epsilon_a \sin^2\varphi_0}} \tag{2b}$$

for p-polarized light at the ambient/film and film/substrate interfaces, respectively. The phase factor ϕ_1 in eq. (1) is given by

$$\phi_1 = \frac{2\pi d_{f1}}{\lambda}\sqrt{\epsilon_f - \epsilon_a \sin^2\varphi_0}\,. \tag{3}$$

The film's thickness is d_{f1}, φ_0 is the angle of incidence and ϵ_s, ϵ_f, ϵ_a are the complex dielectric functions of the substrate, film and the ambient, respectively.

In fig. 2a, $R_p = rr_p \times rr_p^*$ is plotted for transparent films ϵ_f with $\epsilon_{f1} = 8$, 6, 4 and 2 for a thickness range between 0 and 5000 Å. A pronounced modulation of R_p is calculated which has the highest modulation depth for the smallest ϵ_{f1} value. Fig. 2b shows changes in reflectivity during the first 100 Å film growth.

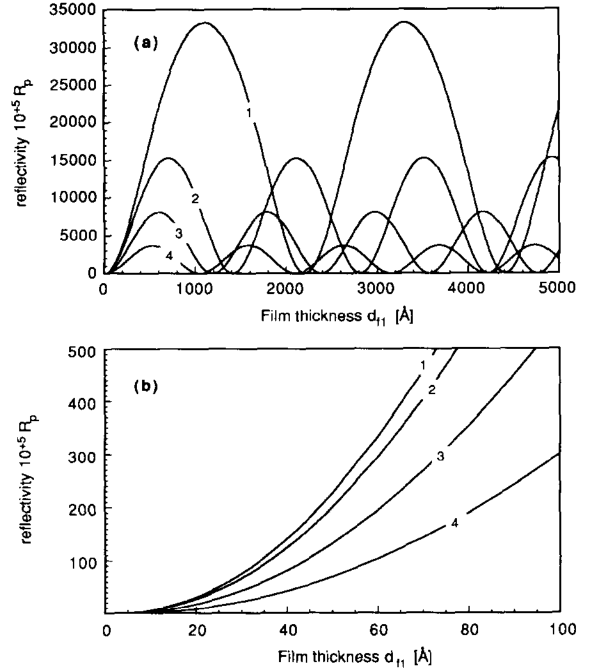

Fig. 2. (a) Calculated changes in reflectivity R_p by growing films (optical constant ϵ_f) on Si-substrate (transparent films ϵ_f with 1: $\epsilon_{f1} = 3$; 2: $\epsilon_{f1} = 6$; 3: $\epsilon_{f1} = 8$ and 4: $\epsilon_{f1} = 10$). Angle of incidence $\varphi_B = 75.637°$. (b) Changes in reflectivity for film growth up to 100 Å.

The occurring minima and maxima as well as the modulation height in the reflectivity are related to the optical constant of the grown film. The sensitivity of the technique is highly dependent on the magnitude between the occurring minima and maxima during the growth. At the beginning of the growth process, after having chosen the angle of incidence at the Brewster angle of silicon, the determined reflectivity is close to the first reflection minimum, with a maximal change in reflectivity. Therefore, a high sensitivity range during the layer growth (seven orders of magnitude) should be available to determine the conditions of growth.

Fig. 3 shows changes in reflectivity between the occurring maxima and minima, as a function of the optical constant of the grown transparent film ϵ_f. For optical constants of the film close to the optical constant of the substrate ($\epsilon_{s1} = 15.25$), the reflectivity range between the occurring minima and maxima decreases and is close to zero if

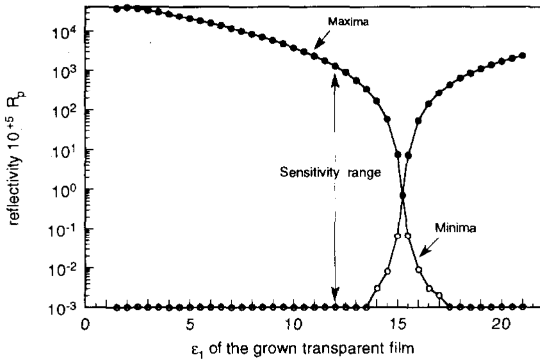

Fig. 3. Changes in reflectivity for the minima and maxima versus optical constant ϵ_{f1} of the grown film at $\varphi_B = 75.637°$.

the optical constants of the film are equal to the optical constant of the substrate.

Fig. 4 shows a contour plot, where changes in the calculated reflectivity minima and maxima positions are drawn as a function of the optical constants of the layer versus thickness of the film. Using both the value of reflectivity R_p (fig. 3) and the positions of the reflectivity maxima and minima (fig. 4), the optical constant ϵ_{f1} of the film can be extrapolated.

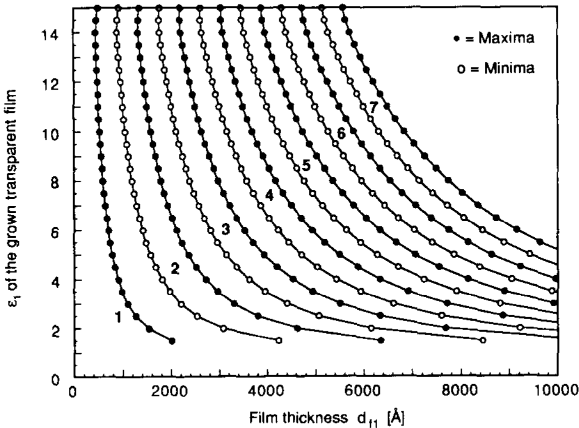

Fig. 4. Contour families of $R_p|_{max}$ and $R_p|_{min}$ in the ϵ_{f1} plane versus film thickness d_{f1}. $R_p|_{max}$ and $R_p|_{min}$ are the reflectivity maxima and minima positions determined during the film growth, respectively (experimental conditions: $\varphi = 75.637°$, $\lambda = 632.8$ nm, silicon substrate: $\epsilon_{s1} = 15.25$ and $\epsilon_{s2} = 0.17$). Curves 1 to 7 indicate the order of the occurring reflectivity maxima (minima).

4. Experimental limitations in the determination of layer growth

The sensitivity of the method during layer growth is dependent on the resolution of the occurring reflection minima which are in the order of 10^{-8} R_p. The depolarization ratio (R_s/R_p) of a Glan–Thompsen prism is better than 10^{-6}. Even for high quality stress-free window material, the polarization ratio is reduced by approximately one order of magnitude. The total depolarization can be estimated to be 10^{-5} multiplied by the reflectivity R_s of the ambient/film/substrate system.

Estimation of the error induced by the angle divergence of the light beam can be analyzed as follows: around the Brewster angle the reflectivity can be expressed as

$$R(\varphi) = R|_{\varphi_B} + \frac{\Delta R}{\theta^2}(\varphi - \varphi_B)^2. \tag{4}$$

Here φ is the angle of incidence, φ_B the Brewster angle of the substrate, $R|_{\varphi_B}$ the reflectivity at this angle and ΔR the difference between $R|_{\varphi_B-\theta}$ and $R|_{\varphi_B}$. The interval of the angle, defined by 2θ, lies in the order of 0.2° up to over 2°, depending on the absorption of the material [8]. The experimentally resolved reflectivity $\bar{R}$ at the Brewster angle can therefore be calculated as

$$\bar{R}|_{\varphi_B} = \frac{1}{2\alpha}\int_{\varphi_B-\alpha}^{\varphi_B+\alpha} R(\varphi)\ \mathrm{d}\varphi = R|_{\varphi_B} + \frac{\Delta R \alpha^2}{3\theta^2}$$
$$= R|_{\varphi_B} + \frac{|R|_{\varphi_B-\alpha} - R|_{\varphi_B}|}{3}. \tag{5}$$

Here, 2α is the interval of angle divergence with $\alpha \leq \theta$. For any other angle of incidence φ inside the interval $[\varphi_B \pm \theta]$ the experimentally resolved reflectivity can be estimated by

$$\bar{R}|_{\varphi_x} = R|_{\varphi_B} + \frac{\Delta R}{6\theta^2\alpha}\left[(\varphi_{x+\alpha} - \varphi_B)^3 + (\varphi_{x-\alpha} - \varphi_B)^3\right] < R|_{\varphi_x}$$
$$+ \frac{|R|_{\varphi_{x+\alpha}} + R|_{\varphi_{x-\alpha}} - 2R|_{\varphi_x}|}{6}, \tag{6}$$

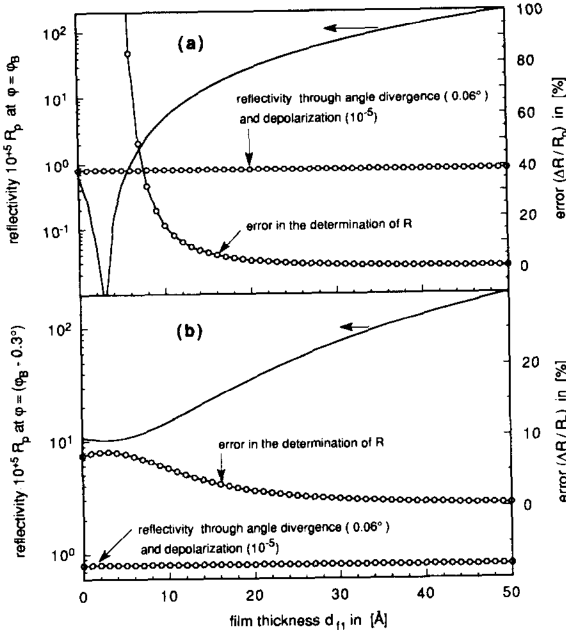

Fig. 5. (a) Changes of reflectivity during the initial stages of film growth on a Si-substrate at the Brewster angle $\varphi_B = 75.637°$ ($\lambda = 632.8$ nm). Due to depolarization and angular divergence, a large error in reflectivity occurs. (b) Changes in reflectivity by monitoring the changes of the reflected intensity at the angle $\varphi = (\varphi_B - 0.3°)$. The calculated error due to experimental limitation is reduced to less than 10% during the initial growth.

where $\varphi_{x \pm \alpha}$ lies inside the angle interval $[\varphi_B \pm \theta]$.

For any angle of incidence φ outside the interval $[\varphi_B \pm \theta]$ the reflectivity can be estimated through a linear function for angle divergences in the order of 0.2° or less. A linear function approximation, however, means that no significant error occurs during a distribution of symmetrical angle divergence. For a more analytical approximation an additional, nearly Gaussian angle divergence distribution can be assumed.

In a first approach, the worst possible conditions (eqs. 5 and 6) were assumed with a distribution of linear angle divergence of 0.06° for the He/Ne-laser. Fig. 5a shows the changes in reflectivity during the layer growth up to 50 Å. The angle of incidence was chosen at the Brewster angle φ_B. Under the assumed experimental limitations, a large error in the determination of the reflectivity occurs below 10 Å thickness. Fig. 5b shows the reflectivity behavior during layer growth for an angle of incidence 0.3° smaller than the Brewster angle.

Here, the reflectivity for the uncoated substrate starts at $R_p \approx 10^{-4}$. Therefore, the error in the determination of the reflectivity is 5–10% below 10 Å thickness. The limitations are strongly related to the depolarization ratio R_s/R_p. In case of depolarization ratios higher than 10^{-5}, the influence of beam divergence can be neglected.

While growing a layer up to several micrometers, a change of the angle of incidence occurs. The change in the angle, β, can be estimated by $\beta = \arctan(d_{f1}/L_{SD} \cos \varphi)$, where d_{f1} is the thickness of the layer; L_{SD} being the distance from the sample to the detector and φ the angle of incidence. For a film thickness of 2 μm and $L_{SD} = 20$ cm, changes in the angle of 0.002° can be observed. Compared to the 0.06° angle divergences of a He/Ne-laser, the occurring error due to the film growth can therefore be neglected.

5. Imperfect film growth

In a simplified approach, the changes in reflectivity due to imperfect layer growth can be modelled by replacing the complex dielectric function of the film, ϵ_f, with an effective complex dielectric function ϵ_e [9]:

$$\epsilon_e = \epsilon_a \frac{\epsilon_f(1 - 2q) + 2\epsilon_a(1 + q)}{\epsilon_f(1 - q) + \epsilon_a(2 + q)}. \tag{7}$$

Here q describes an inhomogeneous film in which q varies from 0 to 1; ϵ_a, ϵ_f, ϵ_e are the complex dielectric functions of the ambient, the film and the effective medium, respectively. In the case of non-epitaxially growing layers, it should be possible to fit the observed behavior with the data of an effective medium. In principle, growth modes such as Frank–van der Merwe, Stranski–Krastanov and Vollmer–Weber [6] should be distinguishable.

6. Conclusion

The new method allows the correlation of film growth with the slope of the reflectivity R_p as a

function of d_{fl}. For larger thicknesses the optical constants of the film can be determined (figs. 2a and 2b). Further information on the growth process can be obtained from changes in the R_p (d_{fl}) function.

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