879

IN SITU MEASUREMENT OF ZINC OXIDE THIN FILM THICKNESS AND OPTICAL LOSSES

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ABSTRACT:

We report a simple and accurate interferometric method for simultaneous, *in situ* measurement of both the thickness and the depth resolved optical losses of thin transparent films. The experimental arrangement is simple, requiring only a laser and a detector regardless of the substrate. The originality of our method is based on the evaluation of the optical losses from the envelopes of the laser signal, while the thickness is measured conventionally by counting the number of signal periods. The technique has been tested while growing ZnO films in a planar RF magnetron sputtering system. Using the above method, the evolution of optical losses is easily observed during deposition, leading to the possibility of a real time control of the film quality. Both theoretical calculations and experimental results are presented in this letter.

Highly oriented piezoelectric zinc oxide films are of great interest for many acoustooptic components, such as high performance acoustooptic modulators which are used in optical switching, Q-switching and mode locking of lasers. Film reproducibility remains a problem despite considerable research into ZnO deposition. *In situ* techniques for evaluation of the film properties can be used to improve reproducibility [1-2]. Ellipsometry allows measurement of both the refractive index and film thickness, but involves an extensive experimental arrangement [3-5]. Transmittance and reflectance methods are also widely used [6-9], and ultrasonic diagnostics have been reported [10,11]. In this letter, we report an interferometric method which is used to determine both the thickness and the depth resolved optical losses of ZnO films during deposition. Optical losses are useful characteristics because it has been shown that they are related to the electromechanical coupling constant k_t of thin film transducers [12]. Note that the losses are not only due to absorption in the film, but are also a result of the scattering of light by the columnar structure of sputtered films.

The first part of this study deals with theoretical considerations that illustrate how the thickness and the optical losses of the film can be determined. In the second part, experimental results are presented. In particular we describe the method used to evaluate the depth resolved optical losses in real time. The film is assumed to be transparent at the interrogating wavelength, λ, so that a Fabry-Pérot interference signal can be obtained, leading to an *in situ* measurement of both the optical thickness of the film and the deposition rate. The physical film thickness can then be determined from a knowledge of the refractive index. Conversely, an independent measurement of thickness allows the calculation of the actual index.

We now consider a homogeneous, isotropic, and transparent film deposited on a reflecting substrate. We assume that the optical losses due to the surface roughness remain constant during deposition. Let d and n be the thickness and the refractive index of the film respectively. An S polarized laser beam of wavelength λ is incident on the film at an angle θ_0 as shown in figure 1-a (a P polarised beam is less sensitive to thickness change). In this case, the normalized power reflectivity is:

$$S = \frac{I}{I_0} = \frac{R + e^{-2\alpha p} - 2re^{-\alpha p}\cos(\varphi)}{1 + Re^{-2\alpha p} - 2re^{-\alpha p}\cos(\varphi)} \qquad 1$$

with R and r respectively the power and amplitude Fresnel coefficients at the vacuum → film interface, α the average losses per unit length, $p=2d/\cos\theta_1$ the ray path through the film for a double traversal, and $\varphi = (4\pi d/\lambda)(n^2-\sin^2\theta_0)^{1/2}$ the phase difference between two consecutive elementary reflected rays. Figure 1-b shows the theoretical reflected signal obtained with the following parameters corresponding to orientated polycrystalline ZnO; n=1.923, deposition rate≈11.17 nm/mn, $\theta_0=74°$, $\alpha=47$ mm^{-1}. The value of the refractive index we use here is lower than the bulk value, because sputtered films are generally less dense than the monocrystalline material. The increase in film thickness corresponding to one period of the reflected signal is:

$$\Delta d = \frac{\lambda}{2\sqrt{n^2 - \sin^2\theta_0}} \qquad 2$$

Therefore, the film thickness is given by the total number of signal periods. In order to calculate the optical losses, we have to consider the envelopes of the signal. Signal *maxima* occur when $\cos\varphi = 1$, then the film thickness is equal to $d_k=k\lambda/2(n^2-\sin^2\theta_0)^{1/2}$, where k is an integer. *Minima* occur when $\cos\varphi = -1$, *i.e.* $d_k=(2k+1)\lambda/4(n^2-\sin^2\theta_0)^{1/2}$. Let S_M and S_m be the envelopes of the *maxima* and *minima* respectively. The envelopes are given by:

$$S_M = \frac{R + e^{-2\alpha p} - 2r\,e^{-\alpha p}}{1 + R\,e^{-2\alpha p} - 2r\,e^{-\alpha p}}$$

$$S_m = \frac{R + e^{-2\alpha p} + 2r\,e^{-\alpha p}}{1 + R\,e^{-2\alpha p} + 2r\,e^{-\alpha p}} \qquad 3$$

Because r is negative, S_m can be equal to zero (visibility equal to 1) as shown on figure 1-c. Therefore, the average losses per unit length can be estimated by measuring the thickness d_0 for which the visibility is equal to 1. From equation (3) the losses are:

$$\alpha = \frac{-\ln\sqrt{R}\cdot\cos(\theta_1)}{2\,d_0} \qquad 4$$

We consider next a film deposited on a transparent semi-infinite substrate whose refractive index is n_0 as shown in figure 2-a. In this case, the normalized reflected intensity is:

$$S = \frac{I}{I_0} = \frac{R_1 + R_2\,e^{-2\alpha p} + 2r_1 r_2\,e^{-\alpha p}\cos(\varphi)}{1 + R_1 R_2\,e^{-2\alpha p} + 2r_1 r_2\,e^{-\alpha p}\cos(\varphi)} \qquad 5$$

where the subscripts 1 and 2 refer to the vacuum → film and the film → substrate interfaces respectively. Figure 2-b shows the theoretical signal obtained with the same parameters as those used for figure 1-b, with n_0=1.46 (fused quartz). The increase in film thickness corresponding to one period of the signal remains the same as that of a reflecting substrate, but *maxima* now occur when $\cos\varphi = -1$ and *minima* when $\cos\varphi = 1$. Hence the *maxima* and *minima* envelopes are described by:

$$S_M = \frac{R_1 + R_2\,e^{-2\alpha p} - 2r_1 r_2\,e^{-\alpha p}}{1 + R_1 R_2\,e^{-2\alpha p} - 2r_1 r_2\,e^{-\alpha p}}$$

$$S_m = \frac{R_1 + R_2\,e^{-2\alpha p} + 2r_1 r_2\,e^{-\alpha p}}{1 + R_1 R_2\,e^{-2\alpha p} + 2r_1 r_2\,e^{-\alpha p}} \qquad 6$$

It can be shown that the signal for a transparent substrate can not exhibit a visibility of 1 in the presence of loss. Therefore, the average losses of a transparent substrate are more difficult to evaluate than those of a reflecting substrate (see below). Figure 2-b also shows that the signal amplitude is lower for a transparent substrate than for a reflecting one. This is because an important part of the incident energy crosses the film, and then propagates into the substrate which is considered to be semi infinite (it is possible to calculate the reflectivity of a system involving a non infinite, or multilayer, substrate using Abeles formalism [13], but it is not the purpose of this study).

This optical thin film evaluation method has been used whilst sputtering zinc oxide on a variety of substrates. We have developed this technique to precisely control the thickness and to monitor the film structure during deposition. Figure 3 shows a signal recorded during the deposition of a 9270 nm thick film on a gold coated silicon wafer. For oriented polycrystalline ZnO, equation (2) implies that $\Delta d = 190$ nm. Therefore the recorded 48.8 periods correspond to a 9270 nm thick zinc oxide film. The thickness for which the visibility is equal to 1 is d_0=3325.5 nm, and equation (4) leads to optical losses of α=47 mm^{-1}. This experimental signal is in good agreement with the theoretical signal of figure 1. However, we acknowledge that the film structure varies during deposition, consequently the optical losses may not be considered constant. By using the envelopes of the recorded signal, we can deduce the depth-resolved optical losses as a function of film thickness. For the envelope of *maxima*, equation (3) can written as:

$$X^2\,(S_M R - 1) + X\,2r(1 - S_M) + (S_M - R) = 0 \qquad 7$$

where $X = e^{-\alpha p}$. For each period of the recorded signal, we measure S_M and the corresponding film thickness, and the optical losses are deduced from the positive solution X^+. Alternatively, the same calculation can be made by considering the envelope of *minima*. Figure 4 shows the average of the real time losses calculated from the measured *maxima* and *minima* envelopes. It can be seen, that the film quality decreases for thicknesses between 1580 and 3290 nm, and then increases from 3290 to 4170 nm. The quality of the film then remains constant between 4170 and 8000 nm. Losses are very high at the beginning of the film deposition, probably because the light is greatly scattered by the initially poorly structured film. The losses of a similar film measured by spectrophotometry are consistent with those obtained using our *in situ* method as shown in the insert of figure 4. Figure 5 shows a comparison between the signals obtained with a reflecting (gold coated silicon wafer) and a transparent substrate (fused quartz), and both compare favourably with the theoretical curves of figures 1-b and 2-b.

In conclusion, theoretical and experimental studies of an interferometric method for the evaluation of both thin film thickness and depth-resolved optical losses have been presented. Due to a large angle of incidence (of the order of 74° with respect to the normal), the thickness variation corresponding to a period of the signal is small, leading to a very accurate film thickness measurement. The method can be used with a large variety of substrates, and can also be applied to different deposition techniques. One possible and important application is the on-line control of deposition parameters to facilitate production of low-loss optical films and high quality acoustic transducers.

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FIGURE CAPTION:

Figure 1: Theoretical results for a reflecting substrate.

a) Schematic diagram of multiple reflections from a transparent film on a reflecting substrate.

b) Theoretical laser signal versus time. The parameters used are: n=1.923 (polycristalline ZnO); rate=11.17 nm/mn; θ_0=74°; α=47 mm^{-1}.

c) Envelopes of the *maxima* and *minima*. d_0 is the thickness for which the visibility is equal to 1.

Figure 2: Theoretical results for a transparent substrate.

a) Schematic diagram of multiple reflections from a transparent film on a transparent substrate.

b) Theoretical laser signal versus time. The parameters used are the same than for figure 1-b, the refractive index of the substrate is n_0=1.46 (fused quartz).

Figure 3: Experimental signal recorded while sputtering a 9270 nm thick ZnO film (48.8 periods) on a reflecting substrate. The thickness for which the visibility is equal to 1 is d_0=3325.5 nm, leading to the optical losses α=47 mm^{-1}.

Figure 4: Real time losses versus film thickness recorded during the deposition of the film. This curve is obtained by averaging the results for envelopes of both *maxima* and *minima*. The insert shows the absorption spectrum obtained with a more lossy film. The orders of magnitude are comparable.

<u>Figure 5:</u> Comparison between experimental signals obtained with a reflecting substrate (upper curve) and a transparent one (lower curve). The signals are not drawn to the same scale.

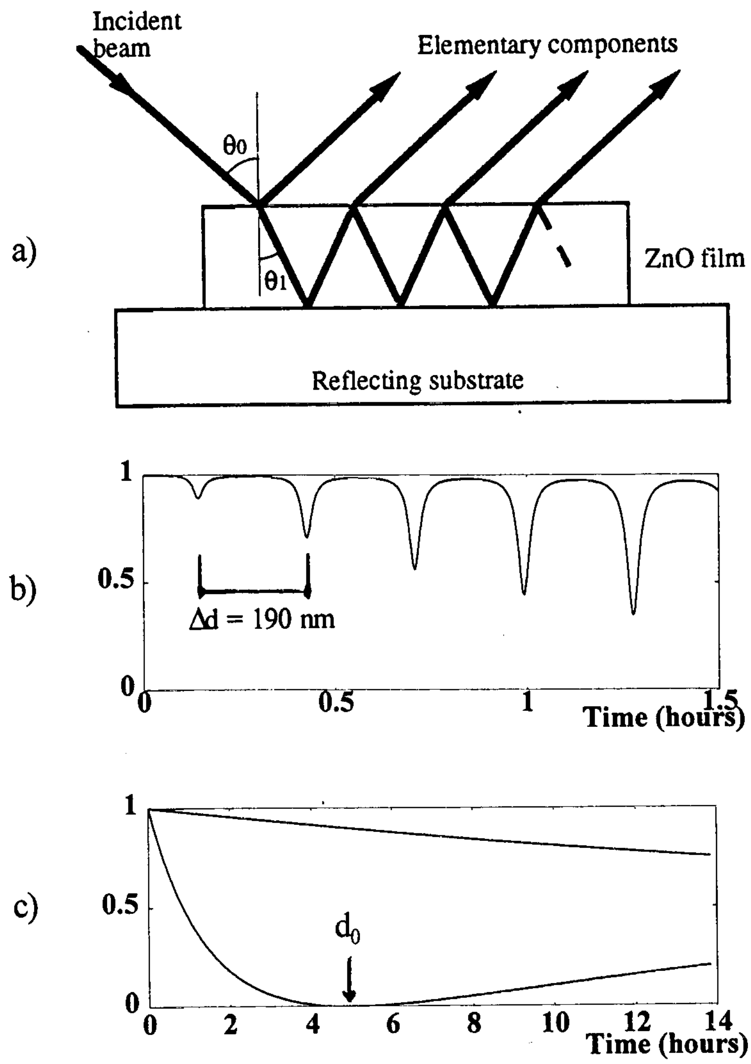
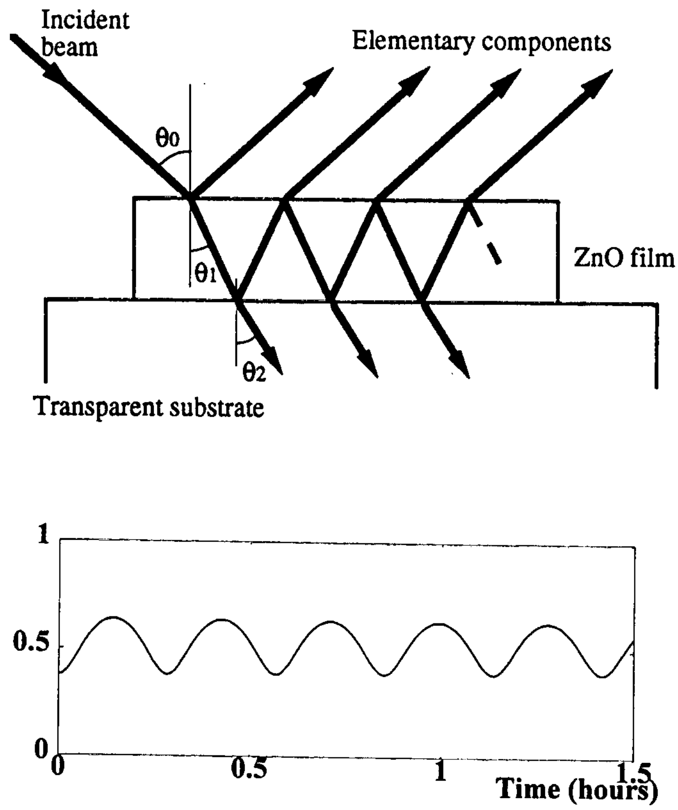

Incident beam
Elementary components
θ0
θ1
a)
ZnO film
Reflecting substrate
b)
1
0.5
0
Δd = 190 nm
0
0.5
1
1.5
Time (hours)
c)
1
0.5
0
d0
0
2
4
6
8
10
12
14
Time (hours)

Fig. 1

a)

b)



Incident beam
Elementary components
θ0
θ1
θ2
ZnO film
Transparent substrate
1
0.5
0
0
0.5
1
1.5
Time (hours)

Fig. 2

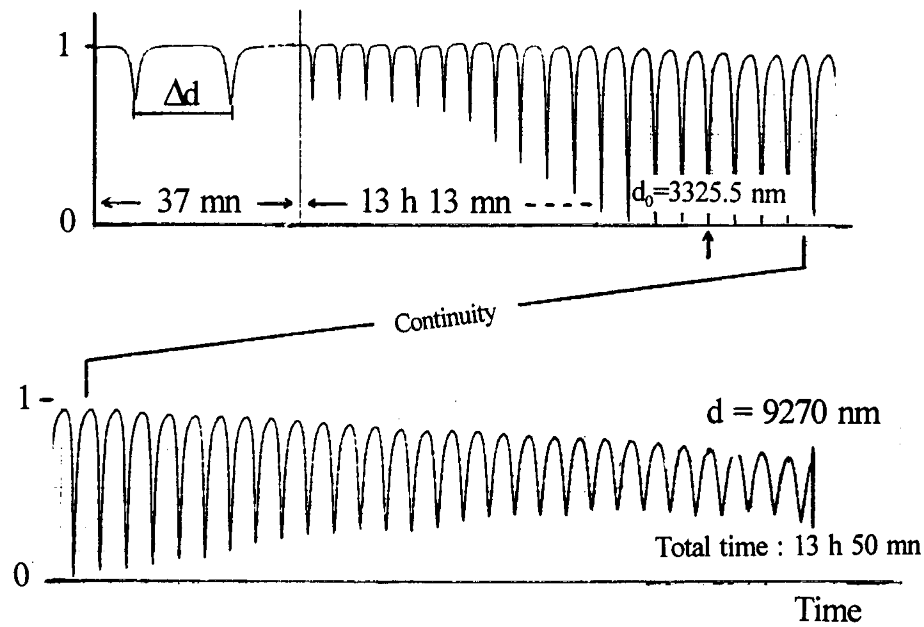

1
0
Δd
← 37 mn →
← 13 h 13 mn ----
d0=3325.5 nm
Continuity
1
0
d = 9270 nm
Total time : 13 h 50 mn
Time

Fig. 3

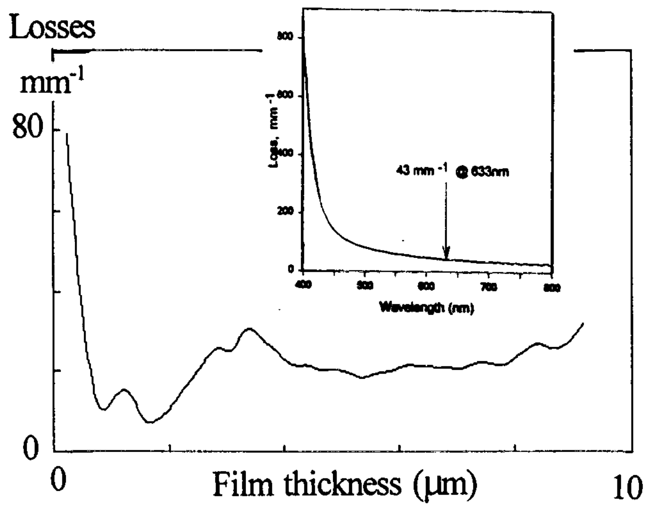

Losses
mm-1
80
0
0
Film thickness (µm)
10
43 mm -1 @ 633nm
Wavelength (nm)

Fig 4

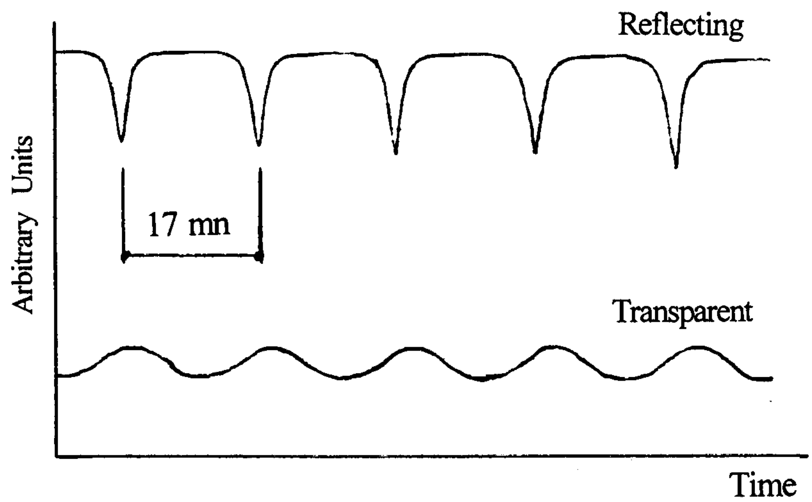


Fig. 5