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Thin Solid Films 313–314 (1998) 394–397

thin solid films

Spectroscopic ellipsometry characterization of indium tin oxide film microstructure and optical constants

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Abstract

Indium tin oxide (ITO) is a transparent conducting oxide in wide use today. ITO can be difficult to work with since this material displays a complicated (graded) microstructure, and the optical properties of ITO can vary widely with deposition conditions and post-deposition processing. For this reason it is common to characterize ITO films via optical measurements. However, accurate results are difficult to obtain due to the graded microstructure of the film introducing variations in the refractive index throughout the film thickness. Thus the typical ITO film does not have a single, well-defined set of optical constants due to grading in the microstructure. Several optical models for ITO will be presented which include the graded microstructure of the material and work reasonably well in fitting spectroscopic ellipsometry data for ITO film thickness, index grading, and optical constants. Since the film thickness, optical constants, and microstructure grading are all intermixed in the experimental data the issue of determining a unique best-fit optical model for ITO will also be discussed. © 1998 Elsevier Science S.A.

Keywords: Spectroscopic ellipsometry; Indium tin oxide; ITO; Optical constants; Refractive index; Graded microstructure

1. Introduction

Indium tin oxide is a widely used transparent conducting oxide. ITO is useful to the flat panel display and optical coating industries because ITO has both good optical transparency and electrical conductivity [1–4]. However, these properties are very sensitive to deposition conditions and post-deposition annealing. Optical monitoring is a useful way of tracking film properties.

The microstructure of ITO changes the electrical and optical properties (refractive index) in a complicated manner. ITO commonly grows with a graded microstructure which introduces grading into the film optical properties (refractive index and extinction coefficient).

Variable angle spectroscopic ellipsometry (VASE) measures changes in polarization of light as a function of angle and wavelength when light is reflected from or transmitted through a sample. Details of the VASE technique are discussed elsewhere [5–9]. This work investigates the effectiveness of different optical models that can be applied to the analysis of ITO films. Sensitivity to the graded microstructure of ITO is also shown, and optical models are developed that account for the grading.

2. Experiment

The ITO film studied here was deposited onto a fused silica substrate. The film was RF sputter de-

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0040-6090/98/$19.00
PII S0040-6090(97)00853-5

posited at 45 W from an ITO target in an argon atmosphere at 5 mtorr pressure. The film was then annealed at 350°C for ~8 h.

Data were acquired using a J.A. Woollam Company VASE® ellipsometer. Optical modeling and data analysis were done using the Woollam Company WVASE32™ software package [10].

Ellipsometric Ψ and Δ data were acquired at three angles of incidence (50°, 60°, and 70°) over the spectral range 300–1700 nm (0.73–4.13 eV) in steps of 10 nm. Intensity transmission data were also acquired with the VASE® at normal incidence over the same spectral range as the ellipsometry data. Multiple angles and wavelengths were fit simultaneously in the optical models.

3. Data analysis and discussion

Three unknown quantities need to be determined: ITO film thickness, refractive index (n) at each wavelength, and the extinction coefficient (k) at each wavelength. Also, it is desirable to model gradients in the refractive index, which add additional fit parameters to the optical models.

At each measured wavelength Ψ and Δ data are measured at multiple incident angles. This helps greatly in adding information content to the data set (information about index grading and film thickness, for example). Thus Ψ and Δ data measured at multiple angles are quite valuable, but it should be noted that adding additional experimental data (e.g. intensity transmission) measured independently of Ψ and Δ greatly helps in determining a unique optical model. Intensity data acquired in transmission mode is very different from polarization data acquired in reflection mode (Ψ and Δ) and thus contains different information. This transmission data can be analyzed simultaneously with ellipsometric data, or used for comparison with the results obtained from Ψ and Δ fits.

3.1. Cauchy models

To determine an approximate film thickness and refractive index it is desirable to find a region of the measured spectral range where the film is transparent (or nearly so). This allows simpler models with fewer parameters to be used in fitting the data. Fortunately, ITO is nearly transparent over the visible spectrum, so it is possible to assume $k = 0$ over part of the visible spectrum and fit Ψ and Δ for the two unknowns of film thickness and refractive index (n).

Fig. 1 shows Ψ and Δ fits to the experimental data over the spectral range 370–650 nm. The ITO refractive index was described using the Cauchy dispersion relation:

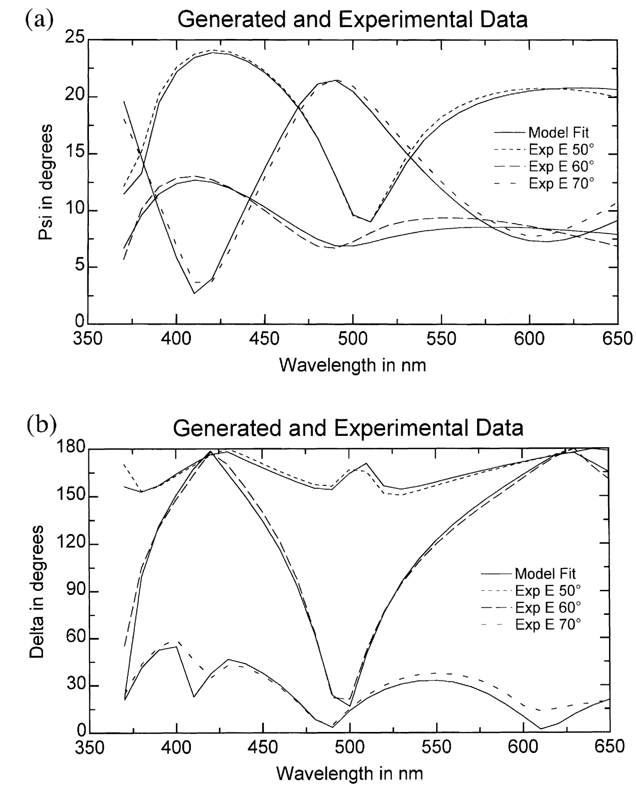

Fig. 1. Ellipsometric data and fit for the range 370–650 nm. The ITO film thickness was determined to be 288.4 nm. The data fits are far from perfect, with the misfit attributed primarily to grading in the refractive index of the film and secondarily to small amounts of absorption in the film.

$$n(\lambda) = A + B/\lambda^2 + C/\lambda^4,$$

and $k(\lambda) = 0$ at all measured wavelengths, where A, B, and C are fit coefficients and λ is the wavelength in microns. The substrate (fused silica) optical constants were taken from the literature and were not allowed to vary during the fits [11].

The transparent region was determined from examination of the intensity transmission data. If transmission data are not available, it is still possible to find the transparent spectral range by fitting the film thickness and Cauchy relation to a narrow spectral range (typically 500–700 nm in the visible). Once an acceptable fit is achieved, the spectral range can be slowly extended to include longer and shorter wavelengths until regions are found where the Cauchy relation no longer fits the data.

The Cauchy model presented above is useful for obtaining an approximate film thickness and set of optical constants. Index grading and absorption (if needed) were not included in this initial model, so the fits are not perfect and the results are only approximate. However, Cauchy fits of this type can be used as a starting point for building more complicated models such as the Lorentz models discussed below.

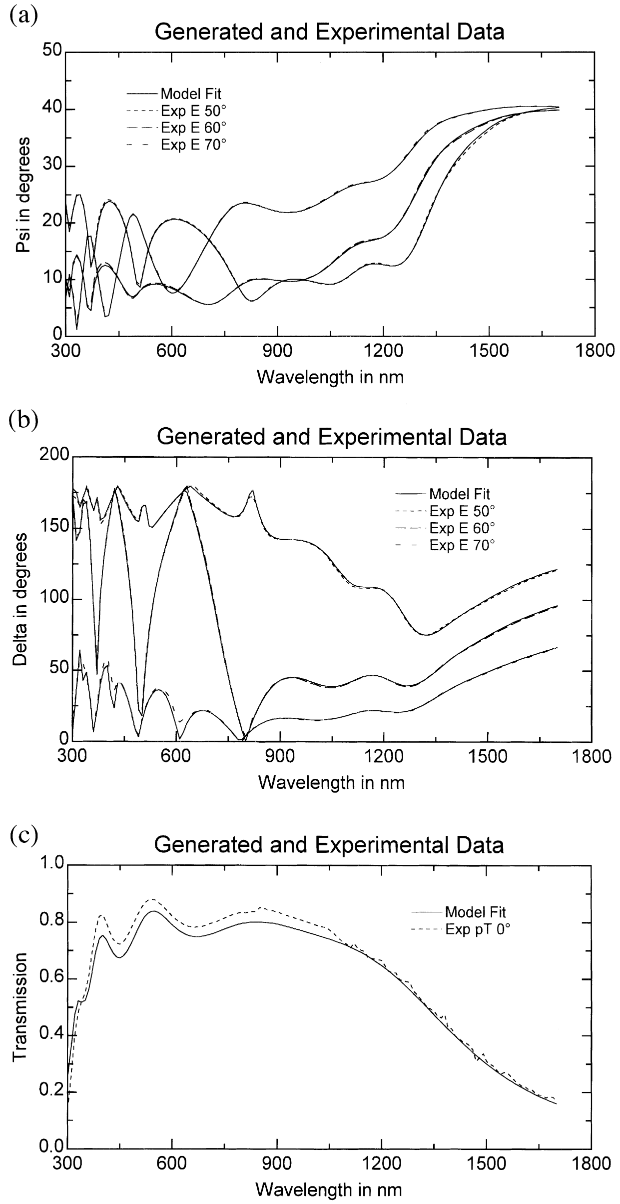

Fig. 2. (a) and (b) Ellipsometric data and fit for the range 300–1700 nm. The ITO film thickness was determined to be 284.4 nm. A graded profile was used to model the ITO optical constants. The best-fit model was used to generate the transmission data in (c) for comparison with experiment. Note that the Lorentz model generates too much absorption between 350 and 1100 nm. This is a limitation of using Lorentzian broadening.

3.2. Lorentz oscillator models

It is usually desired to obtain information about the free carrier conductivity of the film by fitting for absorption at longer wavelengths. This absorption is due to free carriers in the material (Drude edge). A more exact determination of film thickness and optical constants can be achieved by modeling the ITO refractive index with one or more Lorentz (harmonic) oscillators of the form:

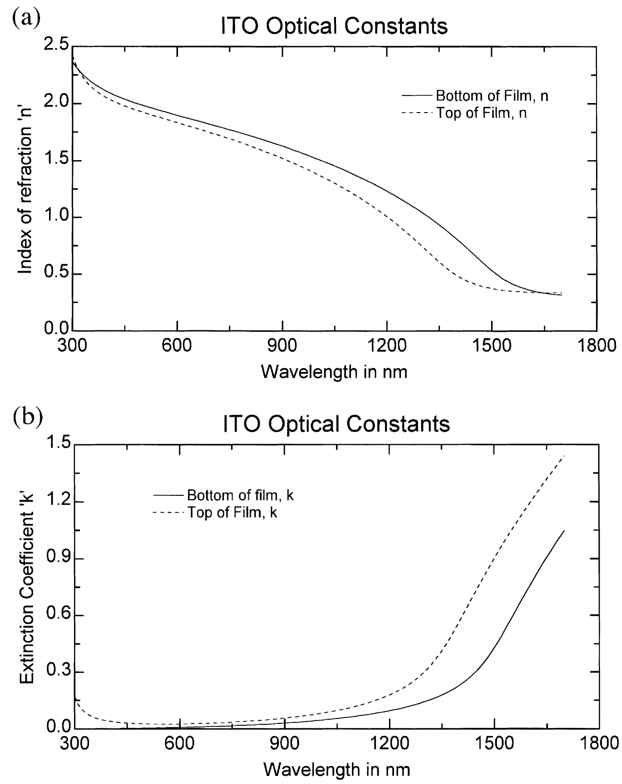

Fig. 3. Refractive index and extinction coefficient of ITO determined from the fits in Fig. 2. Note that the extinction coefficient is lower at the bottom of the film, suggesting that the film is more conductive at the film-air interface.

$$\tilde{\varepsilon}(E) = \tilde{n}^2(E) = \varepsilon(\infty) + \sum_n \frac{A_n}{E^2 - E_n^2 + i\Gamma_n E},$$

where ε is the complex dielectric function and n is the complex index of refraction. The fit parameters in this expression are the dielectric function at infinite energy $\varepsilon(\infty)$, and the amplitude (A_n), center energy (E_n), and broadening (Γ_n) of each oscillator [12,13]. The Lorentz model allows us to simulate the optical constants of the ITO over both the transparent and absorbing spectral range (300–1700 nm used here) with a minimum number of parameters while maintaining Kramers-Kronig consistency between the real and imaginary parts of the refractive index [13,14].

3.3. Graded layers

Using two sets of Lorentz oscillators (four total oscillators) it is possible to model the ITO refractive index at the top and bottom of the film. The total film thickness is divided into multiple sublayers of equal thickness. The optical constants at any point are described as a mixture of the optical constants at the top and bottom of the film using the Bruggeman effective medium approximation (EMA) [15,16]. The EMA mixing fraction is graded linearly between the bottom and top of the film.

The graded model consists of a single ITO layer thickness and two sets of Lorentz oscillators describing the optical constants at the bottom and top of the film. It is important to minimize correlation between the oscillator parameters of the top and bottom layers, or else the best fit optical constants may not be unique. Correlation is minimized by fitting only a minimum number of parameters in the model, and by simultaneously fitting as many data sets as possible. Dispersion models such as the Cauchy and Lorentz describe optical constants over a wide spectral range using mathematical functions. Only the variable parameters of the functions are being fit, rather than n and k at each wavelength. This greatly reduces the number of fit parameters in the model. Also, simultaneously fitting ellipsometric data at multiple angles of incidence greatly reduces correlation between fit parameters and helps ensure a unique best-fit model. Intensity transmission data can be simultaneously analyzed with psi and delta if more information is required to obtain a unique solution.

Fig. 2a and b show Ψ and Δ fits over the spectral range 300–1700 nm. The best-fit optical constants at the top and bottom of the film are shown in Fig. 3. The ITO extinction coefficient is lower at the bottom of the film, suggesting that the film is more conductive near the surface. This is consistent with a graded microstructure. The film is more dense at the top and conduction is enhanced because the film is more continuous at the film-ambient interface.

Fig. 2c shows a comparison of experimental transmission data with that generated from the model. Note that the model generates transmission values that are too low over the spectral range 350–1100 nm. This shows a limitation of the Lorentz model. At both the top and bottom of the film two oscillators were used: one at zero energy to model absorption in the infrared, and another above 4 eV to model the direct band gap. The Lorentzian functions have long 'tails' which add together through the visible spectral range which results in higher absorption and lower transmission values. This problem can be minimized by fitting over a smaller spectral range (500–1700 nm), which avoids modeling the direct gap. This also simplifies the model (two oscillators at high energy can be removed), but comes at the expense of working over a reduced spectral range and avoiding the direct gap.

Lorentzian broadening was used here, but dispersion models based on Gaussian broadening should work better for modeling ITO since the tails are much less pronounced. Dispersion models of this type are being considered for future work on ITO.

4. Conclusion

ITO films are commonly both graded and absorbing. Cauchy fits provide a means for determining the approximate film thickness and refractive index over the visible spectral range where the ITO is nearly transparent. More exact fits can be achieved over a wider spectral range by modeling the film with a graded profile and using Lorentz oscillators to describe the refractive index at the top and bottom of the film. Adding intensity transmission data to the analysis helps ensure a unique solution for the optical model.

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