Optical Monitoring of Thin-films Using Spectroscopic Ellipsometry

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Key Words: in-situ monitoring; E-gun evaporation; ellipsometric optical monitoring; optical properties of materials

ABSTRACT

Spectroscopic Ellipsometry (SE) offers a precise technique for measuring thin film properties. Advanced SE instrumentation has been demonstrated as an excellent technique for monitoring the growth of optical films for sputtering applications. We have recently extended this technique for PVD E-gun evaporated films. In this paper we will show how an SE system was integrated into a standard optical coating system with an electron beam source and used for in situ film thickness monitoring of the growth of single layer films. During deposition of the films, the SE provided real time determination of the film thickness and refractive index. Additionally, the SE data was analyzed to study the homogeneity of the films. Finally a model was developed for in situ control of multi-layer stacks and demonstrated for a 3-cavity 17-layer band pass filter.

INTRODUCTION

Ellipsometry measures the change in polarization state of a probe beam reflected off (or transmitted through) a sample [1]. Ellipsometric results are often presented in terms of the ellipsometric parameters Psi (Ψ) and Delta (Δ), as defined in equation (1). In (1), r_p and r_s are the complex Fresnel reflection coefficients for p- and s-polarized light.

$$\tan(\Psi)\cdot e^{i\Delta} = r = \frac{r_p}{r_s} \qquad (1)$$

The ellipsometric measurement has a number of advantages over traditional intensity reflection and transmission measurements [2]. Since a ratio is measured, accurate ellipsometric measurements can be obtained even in low light conditions, and the data are not sensitive to absolute intensity fluctuations in the light source or baseline intensity drifts (which for in situ measurements could be caused by window coating). Ellipsometry also measures two parameters (Ψ and Δ) at each wavelength, which enables a direct determination of material properties under certain conditions: substrate optical constants (both n and k) can be directly inverted from Ψ and Δ, and film index and thickness can be directly determined for an ideal transparent film. The additional information content provided by the two ellipsometric parameters also provides increased sensitivity to non-ideal film properties such as index gradients and surface roughness. As a final advantage, the ellipsometric Δ parameter, which essentially quantifies the relative 'phase' difference of the reflected beam, is highly sensitive to ultra thin films, even down to the sub-monolayer regime [3]. These advantages, along with its non-invasive nature, make ellipsometry ideally suited for in situ applications.

Spectroscopic ellipsometry (SE) performs the ellipsometric measurement over a range of wavelengths. This further increases the information content of the measurement, enabling the analysis of complex multilayer structures. This also means the coating designs can be entered directly without calculating optimum monitor wavelengths as in discrete single wavelength monitoring. SE has been used in a large variety of applications [4], including technologically important materials and structures in the semiconductor, data storage, optical coating, and display industries, with both ex situ and in situ modes of operation. In situ SE is particularly powerful, in that it enables sample characterization throughout the entire process [5] (as opposed to ex situ measurements, which are only sensitive to the final state of the sample). In situ SE monitoring has been applied to many deposition techniques (e.g., MBE, MOCVD, sputtering, etc.), though relatively little work has been reported on evaporated coatings.

As the direct measurables from an SE system are ellipsometric parameters Ψ and Δ vs. wavelength, data analysis is typically required to determine material properties such as film thickness and optical constants. The details of the analysis procedure can be found in the literature [6,7]. Basically, a layered optical model is built to represent the structure of the sample, and model parameters (such as layer thicknesses, indices of refraction, etc.) are adjusted (or 'fit') via a non-linear regression algorithm to minimize the difference between the optical model generated and experimentally measured SE data. Some examples of such 'data fits', along with the resulting film parameters, will be presented in this paper.

45th Annual Technical Conference Proceedings (2002) ISSN 0737-5921

EXPERIMENTAL DETAILS

A spectroscopic ellipsometer system [8] was integrated with a PVD E-gun evaporation deposition system [9]. An existing Integrity® 36 chamber in Denton Vacuum's Application Laboratory was modified to accommodate the M-2000 source and receiver. Original plans were for having the monitor plane the same as was available for the standard Lambda Pro optical monitor. However installation involved drilling and welding in standard 2-3/4 inch conflat flange mounted windows so that the ellipsometric incident and reflected beams would be at 75°. It turned out that plenum of the chamber interfered with the desired position. Therefore it was necessary to lower the plane of the monitor 4.5 inches. This resulted in an arrangement as shown in figure 1, where the source was mounted on the chamber door and the receiver on the back right side of the chamber.

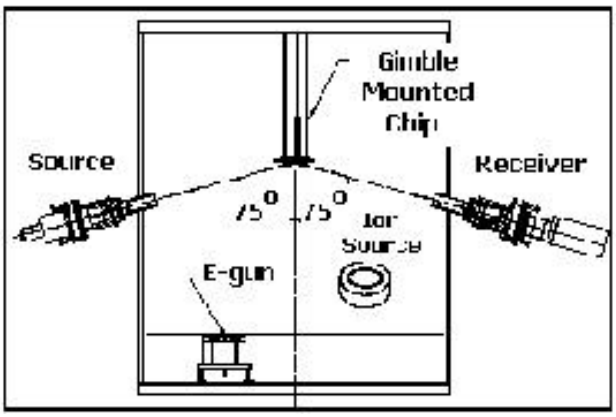

Figure 1. Physical arrangement of the M2000 ellipsometer integrated with the I-36 chamber.

The in situ SE instrumentation employed a simple quartz tungsten halogen (QTH) light source, a rotating compensator optics configuration, and a parallel readout spectrometer/CCD detector system. This enabled the simultaneous acquisition of 400 channels of SE data covering a 370 – 1000nm spectral range. To minimize the amount of data collected during long deposition runs, a data acquisition period of ≈3 seconds was used. The rotating compensator ellipsometer (RCE) configuration has many advantages [10]. In particular, the RCE can accurately measure the ellipsometric parameters Ψ and Δ over the entire range of possible values (Ψ = 0 – 90°, Δ = 0 – 360°). This advantage is especially important for optical coating applications, in which the uncoated glass substrate exhibits Δ values near 0° [11], and Ψ/Δ can vary over the entire range during coating deposition [12].

Initial film deposition was performed on silicon wafers, which is traditionally the substrate of choice for ellipsometric measurements. While accurate layer thicknesses and indices of refraction were extracted for the coatings on silicon, it was found that comparable accuracy could be obtained by using frosted-back glass microscopic slides as substrates. The glass microscope slides are low cost, and as the substrate can influence the initial film nucleation and growth mechanism, the glass slide provides a more representative substrate (compared to silicon) for the glass substrates used in most optical coatings applications. Ex situ SE characterization of multiple slides indicated that their ellipsometric properties are highly reproducible, and therefore the slides could be used as 'reference samples' for the in situ SE measurements. The in situ angle of incidence and vacuum window birefringence (which can severely degrade the accuracy of the SE data, unless properly corrected for [13]) were determined by fitting the reference optical model (determined from the ex situ SE measurements) to in situ SE data acquired on the glass microscope slide before each deposition. A typical in situ SE data fit to the glass microscope slide is shown in figure 2 (note that the model delta curve is non-zero; this is due to a ≈15Å roughness layer on the glass slide). The estimated accuracy in the determined angle of incidence and window birefringence was ≈0.01° and ≈0.1° respectively.

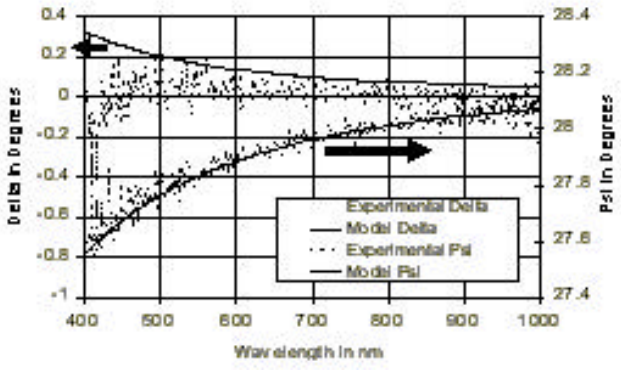

Figure 2. In situ SE data fit to the glass microscope slide before deposition (the angle of incidence and window birefringence were accurately determined in this analysis)

The frosted-back of the slides prevented incoherent reflections from the backside of the substrate from entering the detector. While it is possible to account for such backside reflections in the optical model, ex situ SE measurements on non-frosted slides at normal incidence indicated varying amounts of stress-induced birefringence in the slides; suppressing the backside reflection by frosting the back surface of the slide effectively eliminates the effects of residual substrate birefringence on the measurement.

45th Annual Technical Conference Proceedings (2002) ISSN 0737-5921

RESULTS

Single Layer Films

Single layer films of SiO_2 (10 Å/sec), Al_2O_3 (10 Å/sec), and TiO_2 (8 Å/sec), were deposited on frosted-back glass microscope monitor slides. The high deposition rates were due to the low position of the monitor plane. Deposition rates at the sample plane were half of the rates at the monitor plane. The starting temperature was 40º C and Denton Vacuum's CC-105 ion source parameters chosen so as to make high index films. Final layer thicknesses and average indices of refraction of the films were determined by analyzing the in situ SE data acquired at the end of the run, and from the analysis of ex situ reflectivity data. The optical model used for the in situ SE data analysis consisted of a substrate (using ex situ determined optical constants for the frosted-back glass microscope slide), a transparent layer in which the index dispersion was modeled by a 3-term Cauchy expression ($n = A + B/\lambda^2 + C/\lambda^4$), and a surface roughness layer (the optical constants of which were calculated assuming a mixture of 50% film and 50% void, using the Bruggeman Effective Medium Approximation [7]).

The ex situ optical properties of the films on the monitor slide was determined by spectroscopic reflectivity (R) measurements using relationships as defined previously [14]. This data could then be described dispersively using the same 3-term Cauchy expression.

Table 1 summarizes the layer thicknesses (d) extracted by both techniques from 3 deposition runs. The thickness agreement is excellent for the TiO_2 and Al_2O_3 films. A likely explanation for the discrepancy in the SiO_2 film thicknesses is that the in situ and ex situ characterizations were performed at different regions of the monitor sample; due to the location of the stationary monitor sample in the deposition chamber, thickness variations of ≈10% were observed across the sample. In addition, the SE analysis returns a surface roughness thickness (SRT), which is not accounted for in the reflectivity analysis (approximately half of the SE roughness value should be added to the SE thickness for a more appropriate comparison with the reflectivity-determined thickness). In this study, the surface roughness values were not confirmed by an independent technique, though others have shown good agreement between SE-determined roughness values and AFM measurements of roughness [10].

The index data for the three materials are shown in Figure 3. Excellent agreement is observed for the Al_2O_3 and SiO_2 films: the maximum index difference between the in situ SE and ex situ reflectivity techniques is less than 0.01 over the entire spectral range, and less than 0.005 over most of the spectrum. Larger differences (≈0.05) are observed for the TiO_2 film. While film non-uniformity on the monitor sample may also affect the index comparison (as previously described for the thickness results), changes in the film index of refraction upon exposure to atmosphere are another possible explanation. Unlike the ex situ reflectivity measurements, the in situ SE determination of index is performed under vacuum. Aging effects may also cause the film optical properties to change over time, further increasing the inconsistency with the in situ-determined index values. An additional study will be undertaken to more fully understand such behavior.

Table 1. Thickness comparison from R and SE data

	d (nm) ex situ R	d (nm) in situ SE	SRT (nm) In situ SE
TiO_2 film #2883	680	690	2.3
Al_2O_3 film #2881	656	653	6.0
SiO_2 film #2995	1350	1440	8.2

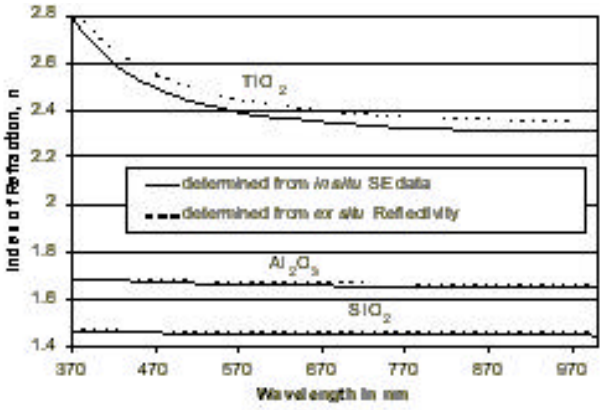

Figure 3. Index of refraction for 3 materials determined by in situ SE and ex situ reflectivity measurements (the curves for Al_2O_3 and SiO_2 lie essentially on top of each other).

Graded Index (Inhomogeneous) Films

To study the sensitivity of in situ SE to film index inhomogeneity, depositions were performed in which the index of refraction was intentionally varied. This was done by adjusting the chamber pressure in discrete steps (over a range of $3x10^{-4}$ Torr to $1.5x10^{-4}$ Torr) while the other deposition parameters were held constant. The decreasing pressure increments would result in increasing ion current density on the growing film causing an increasing refractive index (which is opposite of what is

usually observed when depositing films and trying to maintain a constant index).

The first inhomogeneous run consisted of a ≈0.8 μm TiO_2 film deposited with 6 discrete pressure changes. To determine the index variation throughout the run, a General Virtual Interface (GenVI) analysis was performed on the dynamic in situ SE data. While the details of the GenVI analysis are beyond the scope of this paper (and will be presented elsewhere), this type of analysis is similar in concept to the work of Aspnes and Collins [15,16]. However, while the common pseudo-substrate approximation (CPA) used in their work is nearly exact for semiconductor growth, it is *not* valid for the deposition of transparent dielectric materials. The GenVI analysis therefore uses exact expressions, as done by Urban [17], but in a manner which is more computationally efficient and better suited for real time in situ monitoring. In the GenVI analysis, the most recently acquired SE data points are used to determine the real time deposition rate, near-surface index of refraction, and surface roughness, independent of the previous deposition history. The rate and index vs. deposition time can then integrated, yielding an index profile vs. film thickness.

The index profile resulting from a GenVI analysis of the first inhomogeneous TiO_2 film is shown in Figure 4. Index steps directly corresponding to the deposition pressure changes are clearly seen in the graph. A standard layered optical model-based analysis (which included a gradient in the TiO_2 film) of the SE data acquired at the end of the run yielded index values at the bottom and top of the film, which were in good agreement with the GenVI analysis. However, due to the large amount of index inhomogeneity, a quantitative ex situ reflectivity analysis of the film properties was not possible.

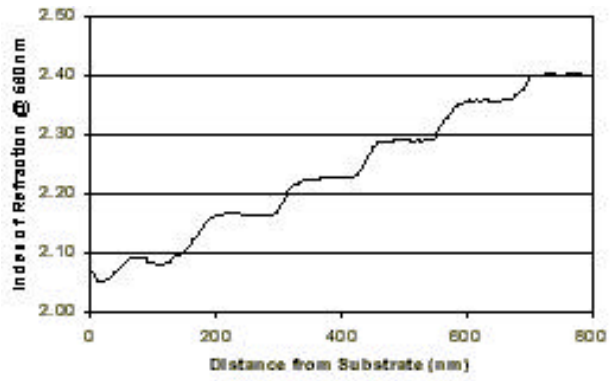

Figure 4. Index profile of 1st inhomogeneous TiO_2 film determined by a GenVI analysis of the in situ SE data.

To enable a better comparison of the in situ SE and ex situ R characterization techniques, a second inhomogeneous TiO_2 was deposited, but at a lower film thickness (≈0.35 μm) and with a reduced index gradient ($2x10^{-4}$ Torr to $1.5x10^{-4}$ Torr). The index profile of the film determined by a GenVI analysis of the in situ SE data is shown in Figure 5. Due to the lower amount of film inhomogenity and limited index precision provided by the current GenVI analysis algorithm, discrete index steps are not discernable in this curve. The high initial index value in the curve is most likely an artifact of the GenVI analysis, which could possibly be corrupted by the film nucleation process.

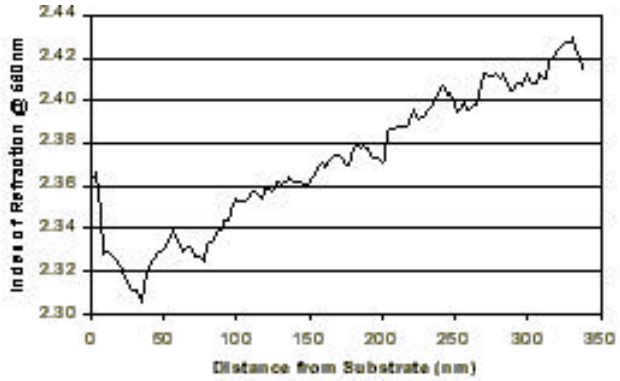

Figure 5. Index profile of 2nd inhomogeneous TiO_2 film determined by a GenVI analysis of the in situ SE data.

Fits from an optical model analysis of the in situ SE data acquired at the end of the run are shown in Figure 6 (for clarity, only the Ψ data fit is shown). Including an index gradient in the optical model is necessary to reproduce the peak heights in the ellipsometric Ψ data; index inhomogeneity induces a similar peak height effect in the ex situ R data.

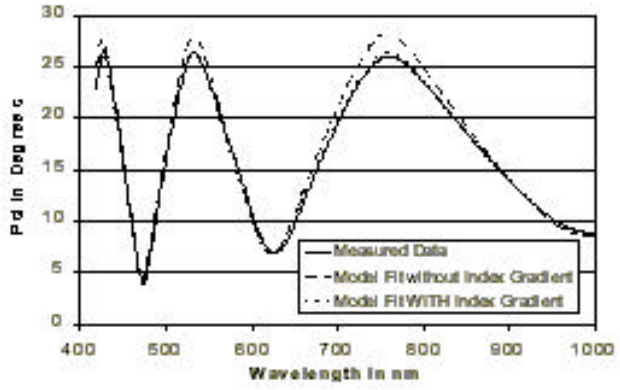

Figure 6. In situ SE data fits for the 2nd inhomogeneous TiO_2 film; the 'model fit with index gradient' curve lies essentially on top of the measured data.

45th Annual Technical Conference Proceedings (2002) ISSN 0737-5921

A comparison of the inhomogeneous film indices at the top and bottom of the film, determined by both in situ SE and ex situ R analysis, is presented in Figure 7. While the film inhomogeneity (i.e., the index difference from the top to the bottom of the film) determined by both techniques is in excellent agreement, the indices from the ex situ R analysis are ≈0.03 higher than the in situ SE values. As in the case with single TiO_2 films, this discrepancy could be explained by a change in film index upon exposure to atmosphere (moisture stable films will have a higher refractive index). It is important to note that the index profile determined from the dynamic GenVI analysis (Figure 5) is also in good agreement with the post-deposition SE and R analysis results.

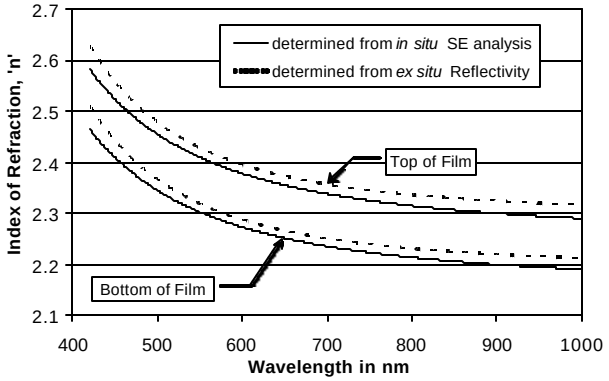

Figure 7. Film indices of refraction for 2nd inhomogeneous TiO_2 film deposition.

Real-time Deposition Control Runs

To demonstrate deposition control capabilities, 17-layer 3-cavity bandpass filter structures were deposited with real-time in situ SE feedback control. The design wavelength of the filters was 650nm, and the layer structure was: (HLHHLH)L(HLHHLH)L(HLHHLH), with H = ¼ wave TiO_2 and L = ¼ wave SiO_2. Deposition rates continued to be 8 Å/sec and 10 Å/sec respectively. Substrate temperature was 40 °C at the start of the deposition and increased to about 140 °C at the end of the deposition due to energy from the E-gun and the ion source

During the deposition, the current layer thickness, index of refraction, and surface roughness were determined by a real-time model fit of the in situ SE data. The total optical thickness (in waves, at the design wavelength) was then calculated from the current optical model. By linearly extrapolating the total optical thickness values obtained vs. time (using the 4 most recent values), the completion time for the current layer was predicted. At the predicted layer completion time, the sample shutter was automatically closed via a digital interface signal from the SE control electronics. The deposition control computer was then advanced to the next layer, and another layer was added to the optical model. Arbitrary structures can be deposited under ellipsometer control by simply entering the layer sequence in H-L notation (non quarter-wave layers can also be specified), along with the design wavelength, into the ellipsometer control software.

Typical fits to the in situ SE data during filter deposition are shown in Figures 8 and 9. The optical model fits are very good for the initial layers of the filter, but degrade as the film stack becomes thicker. Using inhomogeneous layers in the optical model and/or adding interfacial layers could possibly improve the fit. The features in the measured spectra of the thicker films (Figure 9) are also not as sharp as the model calculation. This can be due to both the finite bandwidth of the spectrometer (4nm), and film thickness non-uniformity (which was determined to be the dominate effect in this work). In future studies, spectrometer bandwidth effects will be added to the optical model, and thickness non-uniformity will be minimized by rotating the sample and by increasing the distance between sample and the e-gun source.

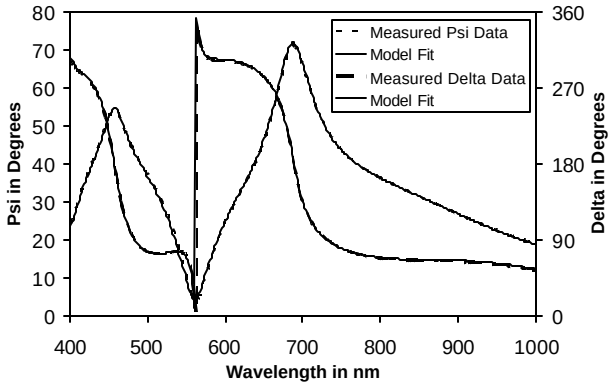

Figure 8. In situ SE model fit after the 4th layer.

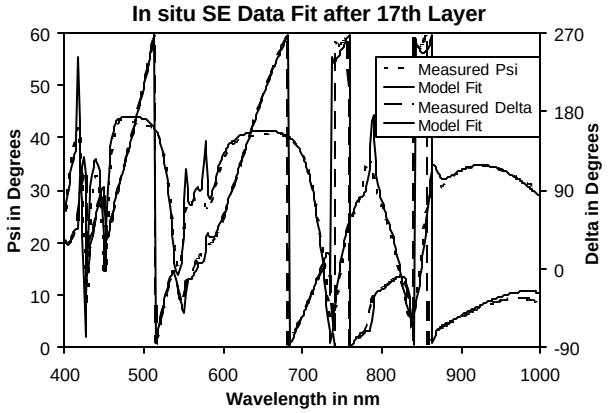

Figure 9. In situ SE model fit after the 17th layer.

45th Annual Technical Conference Proceedings (2002) ISSN 0737-5921

Ex situ measured reflectivity curves for 3 of the SE-controlled bandpass filters are shown in Figures 10 and 11. The measured spectra are in good overall agreement with the design curve (other groups have also achieved similar in situ SE control results [18]). Note that the center wavelength is shifted from the 650nm design value due to the non-normal (18°) angle of incidence. Unfortunately, the thickness variations across the sample (≈10%) made it impossible to quantify the SE control accuracy of the center wavelength. However, the shape of the bandpass is another indicator of the control accuracy: the unsymmetrical profiles of the measured filters indicate slight mismatches (<5nm) in the cavity layer thicknesses. This may be due in part to the imperfect optical model fits to the in situ SE data. However, other effects which could degrade the SE control accuracy, were observed. For example, the optical thickness of the stack appeared to change during the dwell times (≈2 min.) between layers, typically decreasing ≈0.002 waves for most layers, except for the last few layers in which it increased ≈0.001 waves. In the dwell times the deposition shutter was closed (while the e-gun was pre-melting the material for the next layer deposition), but the ion source was still on, potentially resulting in continued modification of the film optical properties. Also, the presence surface roughness in the optical model complicates the determination of the layer completion time. Should calculation of total optical thickness include the roughness layer, or does the roughness always 'ride on top' of the current layer, such that it should be disregarded when determining the layer completion (except for the last layer)? Are interfacial layers of intermediate index formed in between the H and L layers? These questions can hopefully be answered by a careful reanalysis of the in situ SE data. Furthermore, the apparent increase in TiO_2 index upon exposure to atmosphere should also be accounted for by the in situ SE control algorithm.

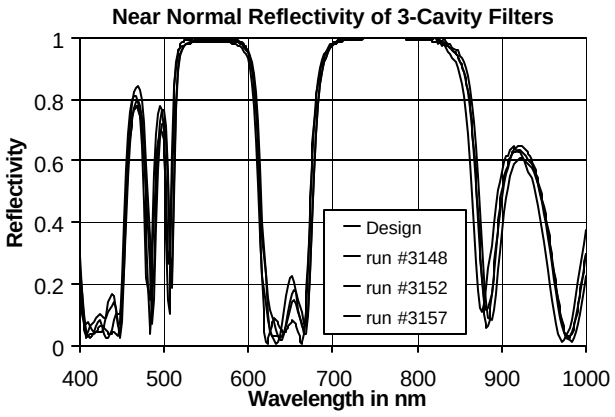

Figure 10. Reflectivity from 3 SE-controlled deposition runs (measured at 18° angle of incidence).

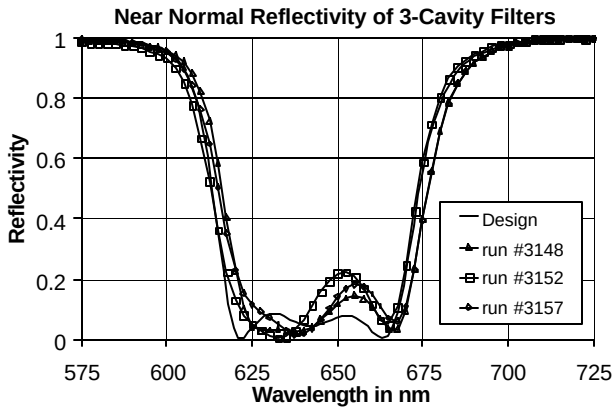

Figure 11. Reflectivity near the bandpass from 3 SE-controlled deposition runs (measured at 18° angle of incidence).

CONCLUSION

The result of this work shows that SE is a valuable tool for studying in real time the growth and modeling of evolving thin film layers. The SE parameters, Psi and Delta of the growing film, which are related to the amplitude ratio of the Fresnel reflection coefficients, can be accurately measured over a broad wavelength range, even in low light intensity situations. Furthermore, SE data analysis does not require the work to find the best monitor wavelength as need for traditional single wavelength level monitoring. Psi and Delta are stored as a function of time over a selectable broad wavelength range during the film growth. This stored data can be analyzed later at any time increment to study the film growth (optical properties and thickness). In situ final mean optical properties or initial and final optical properties can than be compared to the ex situ film properties. We report excellent comparisons for the in situ and ex situ properties of lower refractive index films. Variations of in situ and ex situ refractive index of TiO_2 films were due to the voids left in the film during growth. These voids fill with water when exposed to the humidity in the air, increasing the ex situ refractive index.

Controlled variations in pressure during the growth of TiO_2 films resulted in uniformly increasing refractive index that allowed us to develop an inhomogeneous film model. A layer stack model was developed and refined using a 5-layer single cavity band pass filter. This model was then tested by depositing a 3-cavity 17-layer band pass structure. The layered optical model structure used for real-time in situ SE data analysis is sufficiently accurate and robust such that first pass success of a complex functional coating design was achieved by

45th Annual Technical Conference Proceedings (2002) ISSN 0737-5921

simply inputting the theoretical filter design into the SE control software. There is very close agreement between the finished in situ SE deposition controlled filter and the design model. Further enhancements to the model are expected to result in additional manufacturing improvements.

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45th Annual Technical Conference Proceedings (2002) ISSN 0737-5921