Surface characterization of thin film devices and optical elements

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Optical techniques provide a convenient and non-destructive approach for characterization of surface of thin films and optical elements. This article describes various advanced techniques like spectrophotometry, laser calorimetry, spectroscopic phase-modulated ellipsometry and Zygo interferometry which have been applied to various thin films and optical elements to characterize not only surface features but to also probe inhomogeneity, contamination and inclusions in surface sub-layers.

Introduction

Development of thin film multilayer devices and generation of precision optical surfaces for laser systems and spectroscopic equipment form an integral part of the activities of the Spectroscopy Division at BARC. Characterization facilities have been set up which can provide information about surface roughness, inhomogeneity and surface profile of a variety of thin films and optical elements. These are briefly described below.

Spectrophotometric and calorimetric techniques for measuring surface roughness

It is well known that transmittance and reflectance spectra of a dielectric thin film produce an interference pattern. Any variation in the film thickness, refractive index or modulation of surface roughness and inhomogeneity modifies the interference profile in a unique fashion. Some of the commonly occurring surface features are depicted in Figure 1, such as linear, triangular, rectangular and sinusoidal roughness variation. Other types of surface roughness can be approximated by any of the above forms, simplifying the computational aspect of the data analysis. The transmittance ($T_{\Delta d}$) at a specified wavelength $\lambda$ of a rough thin film having random roughness variation $\pm \Delta d$ can be expressed as:

$$T_{\Delta d} = \frac{1}{\varphi_2 - \varphi_1} \int_{\varphi_1}^{\varphi_2} \frac{A}{B - C \cos(\varphi) + D} d\varphi,$$

where $A = 16n_f^2n_s$, $B = (n_f + 1)^3$, $C = 2(n_f^2 - 1)(n_s^2 - n_f^2)$, $D = (n_f - 1)^3(n_f - n_s^3)$, and $\varphi_i = 4\pi n_f d_i/\lambda$, $\varphi_1 = 4\pi n_f (d - \Delta d)/\lambda$, $\varphi_2 = 4\pi n_f (d + \Delta d)/\lambda$; $d$ is the mean thickness of the film and $n_f$, $n_s$ are the refractive indices of the film and the substrate, respectively. Here, $\Delta d$ refers to actual variation of thickness from the average thickness $d$ as shown in Figure 1 and must not be confused with standard deviation of calculated values or a RMS deviation used conventionally.

A simulated transmission curve of a film with and without surface roughness is shown in Figure 2. As can be seen from the figure, though peak positions remain the same, the peak amplitudes as well as widths get considerably modulated. With a proper model, the experimental spectrum can be very well fitted to determine the nature and extent of roughness present in optical thin films. Using this technique the surface behaviour of several dielectric films such as TiO$_2$, ZrO$_2$, MgO, Al$_2$O$_3$, HfO$_2$ and composite films deposited under various process parameters has been determined. The transmittance and reflectance spectra of the films have been recorded on a Cary-2390 double beam spectrophotometer with a built-in specular reflectance attachment. This technique has also been extended to determine the surface inhomogeneity in thin films. The variations of refractive index on the surface and/or along the growth direction of the thin film also alter the nature of transmittance and reflectance spectrum. The modulated transmittance is given by:

$a$: linear; $b$: triangular; $c$: square wave; and $d$: sinusoidal roughness.

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The mean thickness ($d$) and the refractive index ($n$) of the film are taken as 1000 nm and 3.5, respectively.

$$\delta T_{\delta\ell}(\lambda) = \frac{4\pi n}{\lambda} T(k) \sin \left\{ \frac{2\pi n(d-z)}{\lambda} \right\} + i \frac{n}{n} \sin \left( \frac{2\pi (d-z)}{\lambda} n(z)dz \right), \quad (2)$$

where $k = 2\pi/\lambda$, and $r(\lambda)$ is the complex Fresnel amplitude; other symbols are explained in Figure 3. In case of a homogeneous film the maxima and minima of the interference peaks are dispersive. But in case of inhomogeneity, these peaks get considerably modulated. The nature and extent of this modulation can be analysed numerically to find the distribution of inhomogeneity within or near the surface of the film. A software ‘SPECTR’ has been developed in-house to analyse such surface inhomogeneities from the spectrophotometric measurements. This is illustrated in Figure 3 for a composite film (ZrO$_2$ + MgO + Al$_2$O$_3$).

This technique has severe limitation when the surface layer is very thin (2–4 nm), and surface inclusion and contamination are present. Such a thin surface layer has little effect on the optical properties of the composite film. We have another sensitive technique to derive the properties of such a sub-layer, which is based on laser calorimetry. In this technique absorption-induced thermal properties are detected using a thin gold film detector as well as AD-590 high-sensitive transducer. From the analysis of experimental data using appropriate models, information about various parameters, such as extinction coefficients and thickness of the surface inclusions is obtained.

**Surface characterization by phase-modulated spectroscopic ellipsometry**

In the reflection ellipsometry, the variation of the amplitude and the phase difference between the perpendicular ($p$) and the parallel ($s$) components of the reflected light polarized with respect to the plane of incidence are recorded. The effect of reflection is measured by the two quantities $\psi$ (which measures the amplitude ratio) and $\Delta$ (which measures the relative phase change). These are given by:

$$\rho = r_p/r_s = \tan \psi \exp(i\Delta), \quad (3)$$

where $r_p$ and $r_s$ are the reflection coefficients for the $p$ and $s$ component of the wave, respectively.

The technique of phase modulated ellipsometry has been employed to study the surfaces of e-beam evaporated ZrO$_2$ films. ZrO$_2$ is a hard material with high refractive index suitable for application in optical thin film devices. It has been prepared in our laboratory by e-beam gun evaporation technique on glass substrates at 350°C. Figure 4 shows the experimental measured curve for the ellipsometric parameters ($\psi$ and $\Delta$) as a function of wavelength for a representative ZrO$_2$ film. The data have been fitted with a theoretical model where dispersion of the refractive index $n$ for ZrO$_2$ in the spectral range considered is assumed to be described by the Sellmier’s dispersion relation:

$$n^2(\lambda) = A_s + B_s\lambda^2(\lambda^2 - \lambda_0^2), \quad (4)$$

where $\lambda$ is the wavelength in nanometer, $\lambda_0$ is the wavelength of the oscillator, $B_s$ is the oscillator strength and $A_s$ is the contribution of the ultra-violet term.

The best fit theoretical spectra are also shown in Figure 4. The best fit has been obtained with a three-layer sample structure (also depicted in the figure). The film is found to grow as a compact layer up to a thickness of 125 nm and then voids start appearing in the sample. The deposition is terminated by a surface layer of 57 nm. The best fit values for the dispersion relation obtained are: $A_s = 1.3020$, $B_s = 1.969157$ and $\lambda_0 = 110.183$ nm, which gives the refractive index dispersion of ZrO$_2$ films as shown in the inset of Figure 4.

Apart from carrying out characterization of dielectric films, we have also applied the ellipsometric technique for other surface-related studies, namely the nature of passive films on stainless steel (SS304) surface and surface modification of glass by trichloro-phenyl-silane deposited as monolayers through self-assembly technique.

**Precise mapping of optically plane surfaces by Zygo phase measuring interferometer**

In precision optical equipment, the surface figure of the optical elements needs to be determined accurately. It is a measure of how closely an optical surface conforms to its intended shape. For example, the optical flats for Fabry–Perot etalons have to be matched to within $\lambda/50$ or...
Figure 3. Effect of surface refractive index inhomogeneity on the modulation of transmittance and reflectance.

Figure 4. Surface layer structure of e-beam evaporated ZrO$_2$ film as determined by fitting to the experimental spectra of $\psi(\lambda)$ and $\Delta\lambda$. O, Experimental spectrum of $\psi(\lambda)$; –, best fit theoretical curve with three-layer sample structure; A, experimental spectrum of $\Delta\lambda$; ---, best fit theoretical curve with three-layer sample structure. (Inset), Refractive index ($n$) dispersion of e-beam evaporated ZrO$_2$ film.
Figure 5. Isometric phase contours recorded for a plane surface of Fabry–Perot plates. The surface is convex in shape with peak-to-valley (P–V) of 0.028(10) and root-mean-square deviation of 0.0048(10) from the plane surface (λ = 633 nm).

better which corresponds to 100 Å in the visible region. A phase measuring Zygo interferometer has been employed for mapping of optical surfaces.

The Zygo interferometer basically measures the deviation of the actual surfaces from the ideal shape (say flat) which are available as masters11. These deviations are called surface form errors (figure errors). The surface errors are in the form of bumps or holes at the centre of the surface and zonal bumps and holes at various locations on the entire surface. The surface quality is estimated in the form of peak-to-valley (P–V) deviation of the actual surface and the root-mean-square (RMS) deviation from the best fit plane. The interferometer has been used for matching the two surfaces of an etalon with an accuracy of λ/50 in our laboratory. Figure 5 shows typical contours/isometric phase recorded for a plane surface for one of the Fabry–Perot plates using Zygo interferometer. The phase maps show the shape of the actual surfaces along with P–V deviation and the RMS deviations from the ideally plane surface.