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Determination of optical constants of thin films from transmittance trace

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ABSTRACT

A simple method is depicted in this communication to determine the optical constants of transparent thin films from transmittance versus wavelength traces, showing no fringes, for evaluating thickness. The strength of this technique is apparent when applied to $Zn_{1-x}Mg_xO$ films.

1. Introduction

Accurate knowledge of the wavelength dependent optical constants of thin films is very much essential from both academic and technological standpoints for obtaining fundamental information about its optical behaviour and material properties and in order to put it into device application. Apart from ellipsometric studies, simultaneous determination of the thickness (d) of the thin film and obtaining information about its complex refractive index ($\eta = n + ik$, where n = refractive index and k = extinction coefficient) is quite difficult experimentally as in most practical cases the transmittance traces are devoid of interference fringes which are generally employed to determine the thickness of the films. This problem is more aggravated in the case of nanocrystalline materials in thin film form. Hence, to overcome this inadequacy, several alternative theoretical models and experimental methods involving the determination of n, k and d of thin films from optical spectra were developed [1–11]. In this regard, different numerical techniques were proposed by considering homogeneous, isotropic plane parallel model of thin film and subsequently modified by considering the inhomogeneity and roughness of the films [1–3]. Most of the theoretical techniques developed so far required the knowledge of:

i. both transmittance (T) and reflectance (R) versus wavelength (λ) data [8,9], or,
ii. a dispersion relation for the wavelength dependent refractive index [10–12], or,
iii. special conditions like the presence of fringes in the optical spectra of the thin film [13,14] or a combination of any of these.

All the methods mentioned earlier, except the one proposed by Bhattacharyya et al. [4], thickness values were either supplied or determined from the fringes appearing in the transmittance spectra. Although the method presented by Bhattacharyya et al. did not require thickness as input value but it needed to record the transmittance (T), reflectance (R) measured from film side and reflectance (R_1) from the substrate side. Besides this, all the above spectra have to be taken from the same area of the film that made the technique a bit complicated one. Thus, requirement of an effective theoretical model which would involve the least amount of experimental data to yield useful results is still on the hot anvil. We present here an approach for the evaluation of the optical constants like n, k, dielectric constant along with thickness (d) just by recording the transmittance spectra, containing no fringes, of a transmitting film.

2. Theoretical consideration

The Kramers–Kronig (KK) model seemed to serve this purpose elegantly. Apart from the fact that KK model uses only a single transmittance spectrum, it does not require any dispersion relation for the wavelength dependent refractive index, thus making it a superior choice for calculating the optical constants of thin film. The only difficulty regarding the KK theory is that of recording the transmittance spectra over a wide range of wavelength is necessary to obtain the best results. Measurements in a moderate range of wavelength may also yield quite useful results. But, one needs to have the thickness value as input for evaluating the optical constants. Xue et al. [15] employed the KK theory to obtain the spectral dependence of the real part of the refractive index and the extinction coefficient and hence the real and imaginary part of the complex dielectric constant for transparent ZnO:Al thin films with different Al doping concentration. But, in their computation the thickness of the films was measured separately and taken as input to obtain the

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optical parameters. In this section, we put forward another method for the simultaneous computation of the optical parameters of transparent thin film along with its thickness by using the same KK approach.

The real part of the refractive index, $n(\lambda)$ of semiconductor materials may be related to the optical absorption coefficient $\alpha(\lambda)$ and using the KK model [16] one may write:

$$n(\lambda) = 1 + \frac{1}{2\pi^2}\int_0^\infty \frac{\alpha(\psi)d\psi}{1-\frac{\psi^2}{\lambda^2}} \tag{1}$$

where ψ is the running variable for the wavelength in the wavelength range [0, ∞]. Henceforth, the wavelength dependent extinction coefficient $k(\lambda)$ may be related to the absorption coefficient as:

$$k(\lambda) = \frac{\alpha(\lambda)\lambda}{4\pi}. \tag{2}$$

Correspondingly, the real and imaginary part of the dielectric constants ($\varepsilon = \varepsilon_1 + i\varepsilon_2$) may be related to the real part of the refractive index and the extinction coefficient as:

$$\varepsilon_1 = n^2(\lambda) - k^2(\lambda) \tag{3}$$

and

$$\varepsilon_2 = 2n(\lambda)k(\lambda). \tag{4}$$

Hence, by knowing the values of the absorption coefficient over the whole wavelength range, one may use Eqs. (1) and (2) to evaluate the complex refractive index of the thin film and compute the complex dielectric constants. But, the main problem lies in knowing the optical absorption coefficient value over the wavelength range 0 and ∞. In practice, any UV–VIS-NIR spectrophotometer would permit to estimate α in only a finite optical range. Also, the optical absorption coefficient is related to the transmission coefficient as:

$$T = \exp(-\alpha(\psi)d). \tag{5}$$

Hence, the crux of the problem lies in accurate estimation of the film thickness in order to derive meaningful information about the optical constants of the thin film. This would be possible by using the method described below.

The wavelength range [0, ∞] may be split up into three different parts, viz., the low wavelength limit ranging from [0, λ_1], the estimable wavelength range [λ_1, λ_2] and the higher wavelength limit [λ_2, ∞]. The estimable wavelength range would mean here the range of the spectra as recorded by UV–VIS-NIR spectrometer. Then, writing Eq. (1) in the form:

$$n(\lambda) = 1 + I \tag{6}$$

where,

$$I = \frac{1}{2\pi^2}\int_0^\infty \frac{\alpha(\psi)}{1-\frac{\psi^2}{\lambda^2}}d\psi. \tag{7}$$

The integral I may be split up into three parts, $I = I_1 + I_2 + I_3$, where,

$$\begin{aligned} I_1 &= \frac{1}{2\pi^2}\int_0^{\lambda_1} \frac{\alpha(\psi)}{1-\frac{\psi^2}{\lambda^2}}d\psi, \\ I_2 &= \frac{1}{2\pi^2}\int_{\lambda_1}^{\lambda_2} \frac{\alpha(\psi)}{1-\frac{\psi^2}{\lambda^2}}d\psi, \text{ and} \\ I_3 &= \frac{1}{2\pi^2}\int_{\lambda_2}^{\infty} \frac{\alpha(\psi)}{1-\frac{\psi^2}{\lambda^2}}d\psi \end{aligned} \tag{8}$$

the integrals in each of the wavelength range needs to be evaluated separately and then using Eq. (6) one may estimate $n(\lambda)$. Once $\alpha(\lambda)$ is known for a wavelength (λ), the calculation of $k(\lambda)$ becomes trivial.

In the lower wavelength limit where $\psi < \lambda_1$, the optical absorption coefficient for direct band gap semiconductor near the band edge, where the photon energy is greater than the band gap energy (E_g), is given by:

$$\alpha(\psi) = A\left(h\nu - E_g\right)^{1/2} \tag{9}$$

where h is the Plank's constant, $\nu = c/\psi$, c being the speed of light, and

$$A = \frac{q^2\left(\frac{2m_e m_h}{m_e + m_h}\right)^{3/2}}{nch^2 m_e}. \tag{10}$$

Here, m_e is the electron effective mass, m_h is the effective hole mass, q is the electronic charge, and n is the refractive index of the bulk material. Hence, putting this value of α in I_1 (Eq. (8)), the contribution of integral in the lower wavelength limit can be estimated.

$$I_1 = \frac{1}{2\pi^2}\int_0^{\lambda_1} \frac{A\left(\frac{hc}{\psi} - E_g\right)^{1/2}}{1-\frac{\psi^2}{\lambda^2}}d\psi \tag{11}$$

Integrating in the region $\frac{hc}{\psi} > E_g$, we obtain the analytical expression:

$$I_1 = \frac{A\lambda}{4\pi^2}\sqrt{\frac{E_g\lambda + hc}{\lambda}}\left[\pi - 2\,\mathrm{Arctg}\left\{\sqrt{\frac{\lambda}{E_g\lambda + hc}}\left(\frac{hc}{\lambda_1} - E_g\right)\right\}\right. \\ \left. - 2\sqrt{\frac{hc - E_g\lambda}{hc + E_g\lambda}}\ln\left[\frac{\sqrt{\left(hc - E_g\lambda\right)/\lambda} + \frac{hc}{\lambda_1} - E_g}{\sqrt{\left(hc - E_g\lambda\right)/\lambda} - \frac{hc}{\lambda_1} + E_g}\right]\right]. \tag{12}$$

In the estimable wavelength range [λ_1, λ_2], the absorption coefficient can be calculated using Eq. (5) by taking any arbitrary value of d. Putting this value of $\alpha(\psi)$ into Eq. (8), we obtain after integration,

$$I_2 = \frac{1}{2\pi^2}\sum_{\substack{\psi = \lambda 1 \\ \psi \neq \lambda}}^{\lambda_2} \frac{\alpha(\psi)}{\left(1 - \psi^2/\lambda^2\right)}\Delta\psi \tag{13}$$

where $\Delta\psi$ is the computation steps.

At the point $\psi = \lambda$, the value is,

$$\underset{\psi\to\lambda}{Lim}\left[\frac{1}{2\pi^2}\frac{\alpha(\psi)\Delta\psi}{\left(1-\frac{\psi^2}{\lambda^2}\right)}\right] = \frac{\alpha(\lambda)\lambda}{4\pi^2}. \tag{14}$$

Since the optical absorption is small in the higher wavelength limit and there are no other absorption band in this region, hence, $\alpha(\psi)$ is almost constant. Taking $\alpha(\psi) = \alpha(\lambda_2)$, and putting in Eq. (1), we obtain:

$$I_3 = -\frac{\alpha(\lambda 2)\lambda}{4\pi^2}\ln\left[\frac{\lambda + \lambda_2}{\lambda - \lambda_2}\right]. \tag{15}$$

Then, summing up I_1, I_2 and I_3 and using Eqs. (6) and (7), the real part of the refractive index may be calculated.