Determination of the optical parameters of a-Si:H thin films deposited by hot wire–chemical vapour deposition technique using transmission spectrum only

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Abstract. Three demonstration samples of intrinsic hydrogenated amorphous silicon (a-Si:H) films were deposited using hot wire–chemical vapour deposition (HW–CVD) technique. The optical parameters and the thickness were determined from the extremes of the interference fringes of transmission spectrum in the range of 400–2500 nm using the envelope method. The calculated values of the refractive index (n) were fitted using the two-term Cauchy dispersion relation and the static refractive index values (n_0) obtained were 2.799, 2.629 and 3.043 which were in the range of the reported values. The calculated thicknesses for all samples were cross-checked with Taly-Step profilometer and found to be almost equal. Detailed analysis was carried out to obtain the optical band gap (E_g) using Tauc's method and the estimated values were 1.99, 2.01 and 1.75 eV. The optical band gap values were correlated with the hydrogen content (C_H) in the samples calculated from Fourier transform infrared (FTIR) analysis. An attempt was made to apply Wemple–DiDomenico single-effective oscillator model to the a-Si:H samples to calculate the optical parameters. The optical band gap obtained by Tauc's method and the static refractive index calculated from Cauchy fitting are in good agreement with those obtained by the single-effective oscillator model. The real and the imaginary parts of dielectric constant (ε_r, ε_i), and the optical conductivity (σ) were also calculated.

1. Introduction

Hydrogenated amorphous silicon (a-Si:H) films have attracted considerable attention in modern electronic devices such as thin-film transistors, solar cells, active matrix displays

and image sensors [1]. The optical parameters and the thickness of a-Si:H films govern the performance of the device. Therefore, accurate measurement of the thickness and optical parameters of a-Si:H layer is of extreme importance for device fabrication. For example, in a-Si:H-based solar cells, the active (intrinsic) layer should be as thin as possible to minimize the recombination of the carriers and at the same time it should be thick enough to absorb almost all incident light [2]. Moreover, accurate determination of the refractive index also plays an important role in designing anti-reflection coating of solar cells [3].

Various methods have been used to determine the optical parameters and the thickness of films. These methods are classified into two groups: the first depends on the transmission or reflectance measurements [4] while the other comprises interferometric methods with two and multiple beam interferometry [5] and prism coupling technique [6]. The latter methods provide accurate measurements of optical properties of thin films. However, they require special equipments for the measurements and need experimental expertise [7]. One of the widely used techniques to calculate the optical properties and thickness of films was put forth by Swanepoel [8]. This is relatively a simple procedure that depends on the method suggested by Manifacier *et al* [9] to calculate the optical parameters and the thickness of films using only the transmission spectrum.

In this paper, we report the systematic investigation of the determination of optical properties and thickness of a-Si:H films proposed by Swanepoel [8]. The optical properties include refractive index (n), extinction coefficient (k), absorption coefficient (α), optical conductivity (σ), dielectric constant (ε), optical band gap (E_g) etc. The a-Si:H thin films were grown by HW–CVD technique. An attempt has been made to apply the single-effective oscillator model proposed by Wemple and DiDomenico [10,11] to a-Si:H films. To check the validity of the single-effective oscillator model to a-Si:H films, detailed analysis has been carried out to obtain the optical band gap (E_g) by Tauc's method [12]. Furthermore, the film thickness values are also confirmed by Taly-Step profilometer. We found that the optical band gap values obtained by Tauc's method were in good agreement with those obtained by the single-effective oscillator model.

2. Experimental methods

In the present study, three demonstration samples (denoted by A, B and C) of intrinsic hydrogenated amorphous silicon (a-Si:H) films were deposited in a HW–CVD system, details of which are described in [13]. Films were deposited using pure silane (SiH$_4$) (Matheson Semiconductor Grade) as a source gas simultaneously on Corning #7059 glass to study the optical properties and c-Si wafers (5–10 Ω-cm, p-type) for Fourier transform infrared (FTIR) spectroscopic analysis. The deposition conditions for the samples A, B and C are listed in table 1.

The substrates were placed on the substrate holder and then the deposition chamber was evacuated to a base pressure of $<10^{-6}$ mbar. After that, the substrate temperature was brought to 200°C by appropriately setting the in-built thermocouple and temperature controller. The pressure during the deposition was kept constant using the throttle valve. The deposition was carried out for the desired time and the films were allowed

Table 1. Deposition conditions for a-Si:H films.

Parameter	Sample		
	A	B	C
SiH$_4$ flow rate (SCCM)	1.2	2.0	2.0
Deposition pressure (mTorr)	70	70	20
Substrate temperature (°C)	200	200	200
Filament temperature (°C)	1900	1900	1900
Deposition time (min)	10	10	10

to cool to ∼100°C in vacuum. Then substrate cooling was put on which further cooled the substrates to the room temperature in vacuum. Then films were taken out for characterization.

3. Results and discussion

3.1 *Calculation of film thickness and refractive index*

Figure 1 shows the transmission spectra of a-Si:H demonstration samples A, B and C as a function of wavelength. The spectra were recorded using a double beam UV-visible spectrophotometer (Hitachi-330, Japan) in the wavelength range 400–2500 nm. The spectrophotometer was set with a slit width of 1 nm and hence no slit correction was required. The transmission measurements were carried out in different parts of the film, scanning the entire sample, and a good reproducibility of the respective samples was generally observed. The thickness of the substrate was several orders of magnitude larger than the film thickness. For a uniform film, the interference effects give rise to oscillating curves as shown in figure 1. These interference fringes were used to estimate the optical parameters and the thickness of the films.

According to Swanepoel's method [8] which is based on the idea of Manifacier *et al* [9] of creating the envelopes of interference maxima and minima, as a first approximation, the refractive index (n_1) of the film in the regions of medium and weak absorption can be estimated using the expression

$$n_1 = \left[N_1 + \left(N_1^2 - s^2 \right)^{1/2} \right]^{1/2},$$ (1)

where

$$N_1 = 2s \times \frac{T_\mathrm{M} - T_\mathrm{m}}{T_\mathrm{M} T_\mathrm{m}} + \frac{s^2 + 1}{2},$$

and T_M and T_m are the transmission maximum and the corresponding minimum at a certain wavelength (λ), one being measured and the other being calculated using a suitable
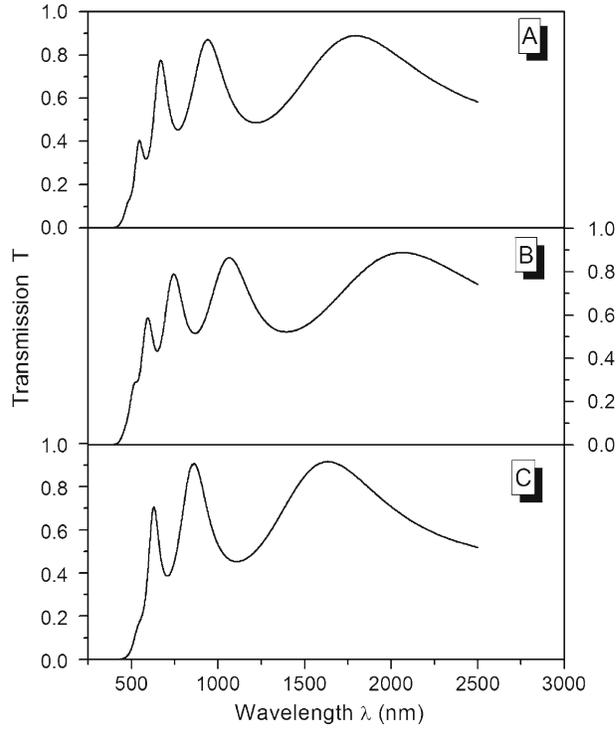
Figure 1. Optical transmission spectra for a-Si:H samples A, B and C.

computer program in order to construct two smooth envelopes that cover the original transmission spectrum.

The refractive index of the substrate (s) can be calculated by measuring the transmission of the substrate (T_s) alone and using the well-known equation [14]

$$s = \frac{1}{T_s} + \left(\frac{1}{T_s} - 1\right)^{1/2}. \tag{2}$$

The refractive index values (n_1) estimated for sample A using eq. (1) are shown in table 2.

These values can be further improved after calculating the film thickness (d). If $n_{e1,2}$ are the refractive indices at wavelengths corresponding to the adjacent maxima and minima, λ_1 and λ_2, then the film thickness is given by the expression

$$d = \frac{\lambda_1 \times \lambda_2}{2\left(\lambda_1 n_{e2} - \lambda_2 n_{e1}\right)}. \tag{3}$$

The values of thickness determined using eq. (3) for sample A are listed in table 2. The last value usually deviates from other values and hence should be rejected because it

Table 2. Values of T_M, T_m, s, n_1, d_1, m_0, m, d_2, n_2 for sample A. The underlined values are calculated as mentioned in the text. The values of refractive index and film thickness are calculated according to the envelope method (similar procedure has been adopted for samples B and C).

λ (nm)	T_M	T_m	s	n_1	d_1 (nm)	m_0	m	d_2 (nm)	n_2
1794	0.890	<u>0.527</u>	1.533	2.792	–	0.94	1	321	2.871
1225	<u>0.877</u>	0.486	1.533	3.035	–	1.46	1.5	312	2.941
940	0.871	<u>0.466</u>	1.533	3.035	299	1.96	2	310	3.009
767	<u>0.810</u>	0.454	1.533	3.006	331	2.38	2.5	319	3.069
669	0.766	<u>0.382</u>	1.533	3.363	278	3.05	3	298	3.212
584	<u>0.519</u>	0.319	1.533	3.247	305	3.37	3.5	315	3.271
546	0.404	<u>0.300</u>	1.533	3.490	366	3.88	4	312	3.495
					$\overline{d}_1 = 303$ nm			$\overline{d}_2 = 312$ nm	

corresponds to the strong absorption of the transmission spectrum, whereas eq. (1) is valid for medium and weak absorption only [8]. The average value of $d_1(\overline{d}_1)$ then can be used with the values of λ and n_1 to estimate the values of the interference order (m_0) using the interferometric equation

$$2nd = m\lambda. \tag{4}$$

The values of m_0 subsequently must be corrected to the exact integers or half integers (m) as shown in table 2. These corrected values of m are now used to calculate new values of thickness (d_2) which are also shown in table 2. The average thickness ($\overline{d}_2$) can now be used along with the values of λ and m to calculate the improved values of n which are listed in table 2 as n_2. These values are fitted to the two-term Cauchy dispersion relation which can be used for extrapolation to shorter wavelengths [15]

$$n = a + \frac{b}{\lambda^2}. \tag{5}$$

The above procedure was done for samples B and C and values of thickness and refractive index were obtained. The values of a and b for a-Si:H samples A, B and C are listed in table 3.

Table 3. Two-term Cauchy parameters, the calculated thickness by envelope method and thickness measured by mechanical Taly-Step profilometer for three a-Si:H samples A, B and C.

Sample	a (n_0)	b	d_{Envelope} (nm)	d_{Taly} (nm)
A	2.799	1.848×10^5	312	300
B	2.629	1.469×10^5	387	400
C	3.043	2.167×10^5	259	250
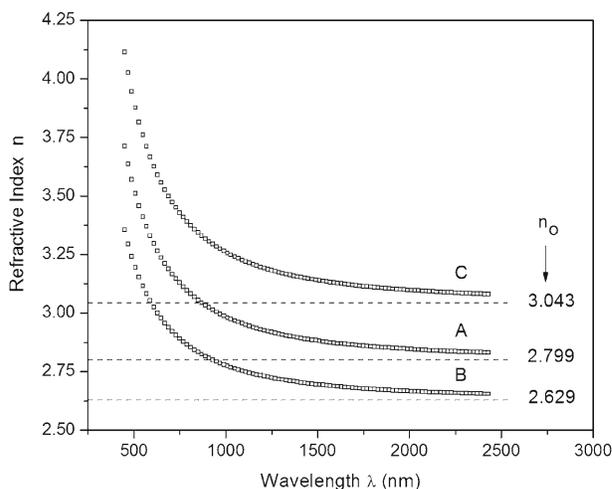
Figure 2. Variation of refractive index as a function of wavelength for a-Si:H samples A, B and C.

The correlation coefficient (r) of the fitted dispersion relation was more than 0.995 for all the three samples. Figure 2 shows the refractive index as a function of wavelength for a-Si:H samples A, B and C. As can be seen from the figure the refractive index decreases with increase in the wavelength. This decrease in refractive index indicates that the HW–CVD deposited a-Si:H material shows normal dispersion behaviour. The values of the static refractive index (n_0) of a-Si:H films estimated by envelope method are in the range of the reported values in [16,17]. Therefore, the envelope method proposed by Swanepoel which uses the optical transmission spectrum only to calculate the thickness and the refractive index is certainly advantageous and accurate for our a-Si:H thin films, whereas, the conventional method for estimating the refractive index of thin films requires the wavelength positions of the interference maxima in the transmission spectrum and independent measurement of thickness [18,19]. Note that in the present study, the Taly-Step profilometer (Taylor–Hobson) was used to cross-check the thickness calculated from the envelope method.

The values of thickness for samples A, B and C deduced by the envelope method are almost equal to the thickness measured by Taly-Step profilometer as depicted in table 3.

3.2 *Determination of the absorption coefficient, extinction coefficient and optical band gap*

The absorption coefficient (α) can be calculated using the equation [8]

$$\alpha = \frac{1}{d} \times \ln(x), \qquad (6)$$

where d is the thickness and x is the absorbance which is given by the well-known formula proposed by Connell and Lewis [20]

$$x = \frac{P + \left[P^2 + 2QT_\alpha(1 - R_2R_3)\right]^{1/2}}{Q},$$ (7)
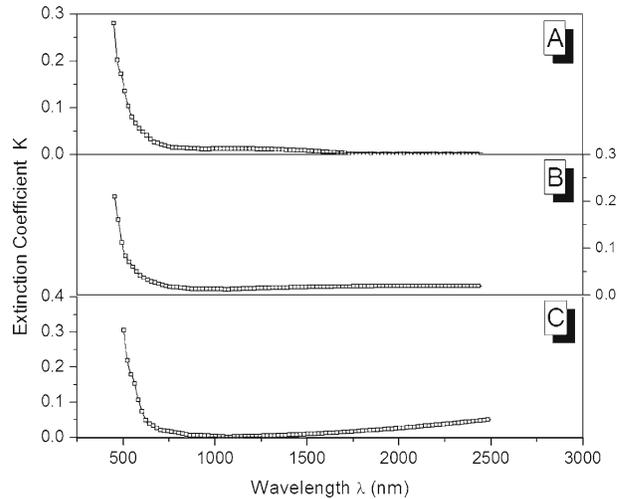
where

$$P = (R_1 - 1)(R_2 - 1)(R_3 - 1)$$

and

$$Q = 2T_\alpha(R_1R_2 + R_1R_3 - 2R_1R_2R_3).$$

R_1, R_2 and R_3 are the reflectance of the air–film, film–substrate and substrate–air interfaces, respectively and are given by

$$R_1 = \left[\frac{1-n}{1+n}\right]^2, \quad R_2 = \left[\frac{n-s}{n+s}\right]^2 \text{ and } R_3 = \left[\frac{s-1}{s+1}\right]^2.$$

In the spectral region with the interference fringes, T_α is the geometric mean of T_M and T_m and is given by [21]

$$T_\alpha = (T_M \times T_m)^{1/2}.$$ (8)

Figure 3. Variation of extinction coefficient as a function of wavelength for a-Si:H samples A, B and C.

The extinction coefficient is a measure of the fraction of light lost due to scattering and absorption per unit distance of the penetration medium. It can be estimated from the values of α and λ using the relation [22]

$$K = \frac{\alpha\lambda}{4\pi}. \tag{9}$$

Figure 3 shows the dependence of extinction coefficient on the wavelength for the three a-Si:H samples A, B and C. As seen from the figure, the extinction coefficient decreases with the increase in the wavelength. The decrease in extinction coefficient with increase in wavelength shows that the fraction of light lost due to scattering and absorbance decreases. Furthermore, the optical absorption edge can be seen clearly for all three a-Si:H samples under investigation. Our results are very similar to the results of Marquez *et al* [23] and have similar trend as those shown by El-Sayed and Amin [24]. Recently, Sharma and Katyal [25] have also observed similar behaviour of extinction coefficient for a-$(As_2Se_3)_{90}Ge_{10}$ glassy alloy.

The absorption coefficient of amorphous semiconductors in the high-absorption region, assuming parabolic band edges and energy-independent matrix elements for intraband transitions is given by Tauc [12] as

$$(\alpha E)^{1/2} = A(E - E_g), \tag{10}$$

where A is a constant which is almost independent of the chemical composition of the semiconductor, E is the photon energy and E_g is the optical band gap. The optical band gap is defined as the E axis intercept of the linear part of the plot of $(\alpha E)^{1/2}$ vs. E (Tauc's plot). Figure 4 shows Tauc's plots for the a-Si:H samples A, B and C. It can be seen from the figure that the estimated values of optical band gap for samples A, B and C are 1.99, 2.01 and 1.75 eV, respectively.
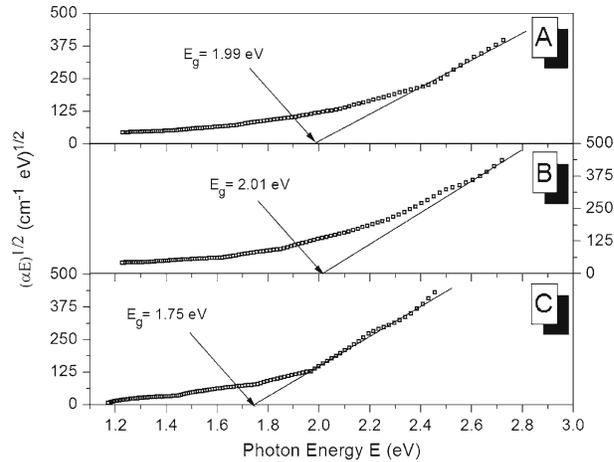
Figure 4. Variation of $(\alpha E)^{1/2}$ as a function of photon energy for a-Si:H samples A, B and C.

For a-Si:H the dependence of the optical band gap and the refractive index on the hydrogen content is well known. It has been reported that the a-Si:H with more hydrogen content has lower density, lower refractive index and higher optical band gap [26]. The hydrogen content in the present study was estimated from the FTIR spectra using the method proposed by Brodsky *et al* [27] and was found to be 5.6, 6.4 and 4.2 at.% for the a-Si:H samples A, B and C, respectively. It can be noticed that the results of refractive index and optical band gap are consistent with the estimated values of the hydrogen content in the samples.

### 3.3 *Determination of dielectric properties and optical conductivity*

The complex dielectric constant is a fundamental intrinsic property of the material. The real part of the dielectric constant shows how much it will slow down the speed of light in the material, whereas the imaginary part shows how a dielectric material absorbs energy from an electric field due to dipole motion. The knowledge of the real and the imaginary parts of the dielectric constant provides information about the loss factor which is the ratio of the imaginary part to the real part of the dielectric constant. The real and the imaginary parts of the dielectric constant can be estimated using the relations [28]

$$\varepsilon_{\mathrm{r}} = n^2 - k^2 \quad \text{and} \quad \varepsilon_{\mathrm{i}} = 2nk. \tag{11}$$

Figures 5 and 6 show the variation of the real and the imaginary parts of the dielectric constant as a function of photon energy for the three a-Si:H samples A, B and C. It can be seen clearly from the figures that both the real and imaginary parts of the dielectric constant increase with increase in photon energy. The real part of the dielectric constant increases sharply with increase in photon energy whereas the imaginary part increases
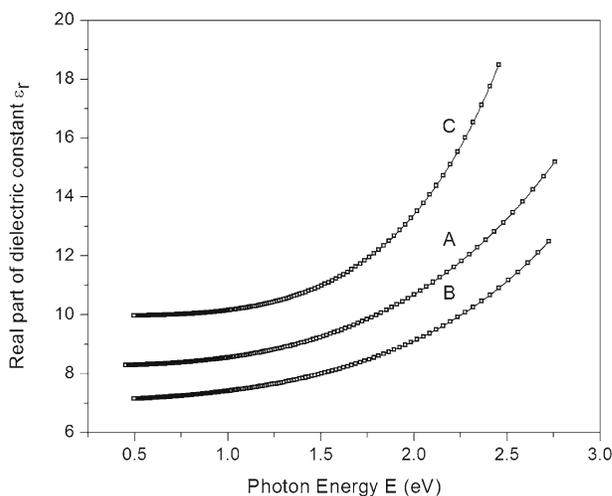
Figure 5. Variation of real part of dielectric constant as a function of photon energy for a-Si:H samples A, B and C.
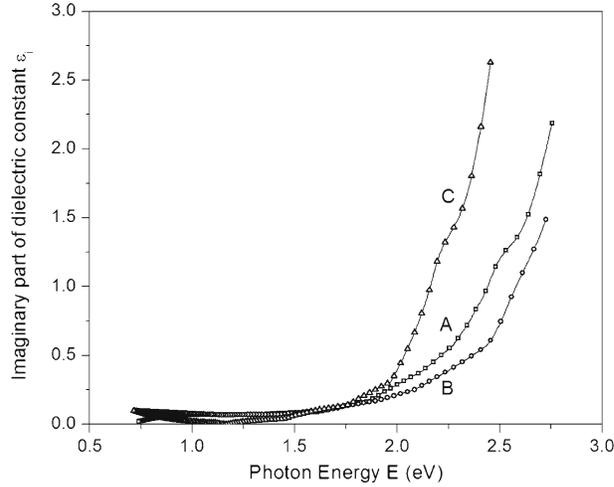
Figure 6. Variation of imaginary part of dielectric constant as a function of photon energy for a-Si:H samples A, B and C.

slowly in the lower energy region but increases sharply in the higher energy region. These results indicate that in a-Si:H the loss factor increases with increase in photon energy.

The optical response of a material is mainly studied in terms of the optical conductivity (σ) which is given by the relation [25]

$$\sigma = \frac{\alpha n c}{4\pi}, \tag{12}$$

where c is the velocity of light, α is the absorption coefficient and n is the refractive index.

It can be seen clearly that the optical conductivity directly depends on the absorption coefficient and the refractive index of the material. Figure 7 shows the variation of the optical conductivity as a function of photon energy for the a-Si:H samples A, B and C. It can be seen clearly from the figure that the optical conductivity increases abruptly after ~ 2 eV for all a-Si:H samples under investigation. The sudden increase in optical conductivity can be attributed to the increase in absorption coefficient.

3.4 *The single-effective oscillator model*

According to the single-effective oscillator model proposed by Wemple and DiDomenico [11,12], the optical data can be described to an excellent approximation by the relation

$$n^2 - 1 = \frac{E_d E_0}{E_0^2 - E^2}, \tag{13}$$

where $E \,(= h\upsilon)$ is the photon energy, n is the refractive index, E_0 is the single-effective oscillator energy and E_d is the dispersion energy which is a measure of the average strength of the interband optical transitions. Plotting $(n^2 - 1)^{-1}$ against E^2 gives the
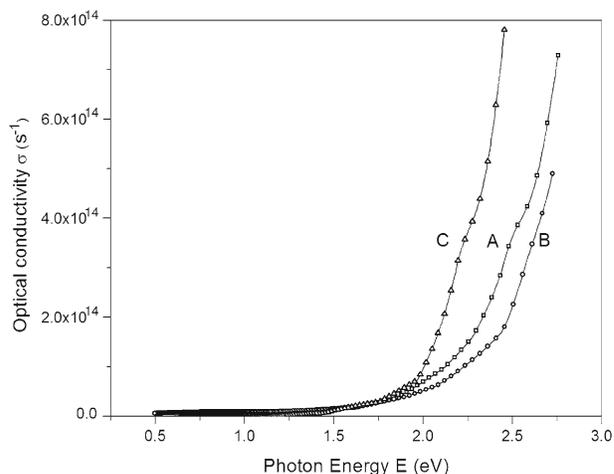
Figure 7. Variation of optical conductivity as a function of photon energy for a-Si:H samples A, B and C.
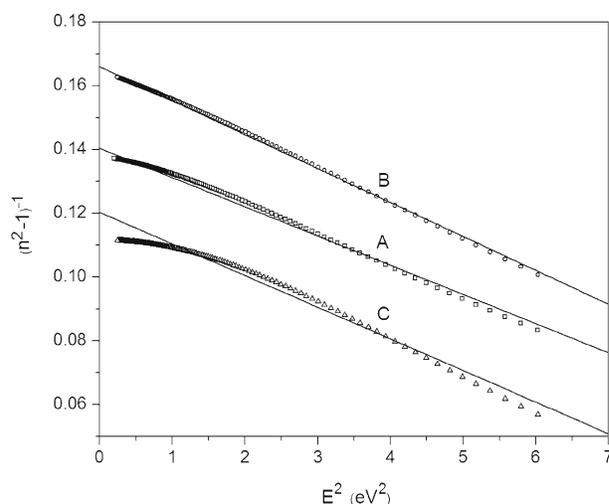
Figure 8. Variation of $(n^2 - 1)^{-1}$ as a function of E^2 for a-Si:H samples A, B and C.

oscillator parameters by fitting a straight line. Figure 8 shows the plot of $(n^2 - 1)^{-1}$ vs. E^2 for the a-Si:H samples A, B and C. The values of E_0 and E_d can be then calculated from the slop $(E_0 E_d)^{-1}$ and the intercept on the vertical axis (E_0/E_d). The calculated values of E_0 and E_d are listed in table 4. Furthermore, the values of the static refractive index (n_0) can be calculated by extrapolating the Wemple–DiDomenico dispersion equation to $E \to 0$. The calculated values of n_0 are 2.85, 2.65 and 3.1 for the a-Si:H samples A, B and C, respectively and these values are in good agreement with the values obtained

Table 4. The values of static refractive index (n_0), single oscillator energy (E_0), dispersion energy (E_d) and optical band gap (E_g) for a-Si:H samples A, B and C.

Sample	n_0	E_0 (eV)	E_d (eV)	E_g (eV)
A	2.85	3.91	27.88	1.96
B	2.65	3.95	23.80	1.97
C	3.10	3.69	31.80	1.84

from Cauchy fitting (see figure 2). In addition, the optical band gap (E_g) can also be determined from the Wemple–DiDomenico dispersion parameter E_0 using the relation $E_g = E_0/2$, giving alternative values 1.96, 1.97 and 1.84 eV for the a-Si:H samples A, B and C, respectively. These values are also in good agreement with that determined from Tauc's plot extrapolation (see figure 4).

4. Summary and conclusions

We have analysed the transmission spectrum of a-Si:H thin films deposited by HW–CVD technique in the range 400–2500 nm for the calculation of optical parameters and thickness. The calculated thickness for all samples using the envelope method were cross-checked with thickness measurements done by Taly-Step profilometer and they were in good agreement. The refractive index and the extinction coefficient decreased with the increase in wavelength and the absorption edge was observed in all samples. The decrease in refractive index with increase in the wavelength indicates that HW–CVD-deposited a-Si:H shows normal dispersion behaviour. The values of the static refractive index obtained for all samples were in the range of the reported values. Thus, the envelope method used to estimate the thickness and the refractive index is certainly advantageous and accurate in the case of a-Si:H thin films. The real and imaginary parts of the dielectric constant increase with increase in photon energy. These results indicate that for a-Si:H the loss factor increases with increase in photon energy. The optical conductivity also increases with increase in photon energy and this can be attributed to the increase in absorption coefficient with increase in photon energy. Finally, an attempt was made to apply Wemple–DiDomenico single-effective oscillator model to the a-Si:H samples to calculate the optical parameters. The optical band gap obtained by Tauc's extrapolation and the static refractive index from Cauchy fitting were in good agreement with those obtained by the Wemple–DiDomenico single-effective oscillator model. The results of the refractive index and the optical band gap were correlated to the hydrogen content in the a-Si:H films and found to be consistent.

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