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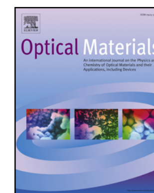

Efficacy of Raman mapping over ellipsometric spectroscopy and XRD for characterization of structurally heterogeneous PLD nc-Si thin films

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ABSTRACT

The present report demonstrates the efficacy of Raman mapping over X-Ray diffraction (XRD) and spectroscopic ellipsometry (SE) in structural characterization of nanocrystalline Silicon (nc-Si) films having spatial heterogeneity in crystallinity and structure. The Si films were deposited via pulsed-laser deposition (PLD) technique at room temperature and elevated substrate temperature (T_s) of 400 °C and 700 °C, under vacuum (~10^{-6} mbar). XRD showed that all films were polycrystalline while SE studies showed that only at 700 °C, the film was nanocrystalline in nature otherwise largely amorphous at lower T_s. This discrepancy in results is well explained by Raman maps which confirmed the presence of micron sized structures composed of nc-Si embedded in otherwise uniform matrix comprised of amorphous Silicon (a-Si) for films fabricated at room temperature and 400 °C whereas at 700 °C the uniform background matrix is dominated by nc-Si. The real and imaginary parts of pseudo dielectric function (ε_1,ε_2) versus energy spectra obtained for films grown at RT, 400 °C, and 700 °C were modeled and fitted to substrate/film/top SiO_2 layered structure. The simulated (ε_1,ε_2) patterns are in excellent agreement with the recorded spectra. From the SE studies the band gap and thickness of the films were estimated. Band gap energy was found to vary from 1.35 to 1.30 eV approximately with increasing T_s from room temperature to 400 °C, then gradually increases to 1.55 eV at 700 °C. The variation of other optical constants of the films as a function of T_s also reflects the structural transition from amorphous-like to nanocrystalline-like with increasing T_s.

1. Introduction

Si based thin films are technologically very useful material for its compatibility with microelectronics industry. Crystalline silicon (c-Si) has some drawback for solar cell applications due to high cost and low absorption coefficient in visible light spectrum. However, nanocrystalline silicon (nc-Si) overcomes these limitations. It absorbs light better than c-Si, has less disordered structures than amorphous silicon (a-Si), and, hence exhibit better electronic and optoelectronic properties. Solar cells with the nc-Si layers have been proven to be efficient and low-cost technology [1,2]. The stability and efficiency of the nc-Si films are important for device applications and both these factors are determined by the microstructural characteristics of the film [3]. A variety of nanostructured silicon, viz. nano-crystalline Si, nano structured amorphous Si (α-Si), nc Si or α-Si embedded in amorphous SiO_2 matrix, porous Silicon (po-Si) etc., show intense visible and near infrared photoluminescence [4,5]. Si based thin films have been reported to be used as micro electro mechanical system (MEMS) based memory devices and antireflective surface structuring [6]. Among all the deposition techniques, PLD offers the advantages of fabrication of stoichiometric as well as non-stoichiometric thin films in a single step by controlling the deposition parameters [7]. It does not require any hazardous gas and hence environment friendly. The nc-Si and microcrystalline silicon (μc-Si) grown by PLD could be structurally heterogeneous comprising of Si nanocrystallites (≈2–20 nm) and their aggregates (~few hundred nm in μc-Si), dispersed on amorphous Si matrix having disorder in the form of voids and grain boundaries [8]. Also the structural evolution of PLD nc-Si films can be manipulated by altering the deposition parameters to yield tailorable optical properties, e.g., like optical band gap, refractive index, extinction coefficient etc. X-Ray diffraction (XRD), Spectroscopic Ellipsometry (SE) and Raman spectroscopy are basic non-destructive optical tools routinely employed to study the microstructural properties of nc-Si films [9–12]. The XRD and SE studies provide the average picture of crystalline properties of films whereas micro Raman spectroscopy can probe the crystalline property and bonding structures locally and can unveil the structural inhomogeneity. The shape and peak position of the first order Raman scattering of Si provides information about average nanocrystallite size

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as well as stress present in it [13,14].

In the present manuscript, the structural and optical properties of nanostructured Si thin films deposited at room temperature (RT) and elevated substrate temperature (T_s) of 400 °C and 700 °C have been presented. XRD, SE and Raman spectroscopy were employed to study the microstructural properties of these PLD Si films. The main aim of this work is to demonstrate the efficacy of micro Raman spectroscopy to probe local nc-Si clusters, over XRD and SE studies which give global overview only. The micro Raman mapping technique was employed to distinguish the amorphous and crystalline domains within each film which XRD and SE failed to do. Moreover, the thickness, band gap, refractive index and extinction coefficient of the films were estimated from SE studies by using appropriate models and dispersion laws. The film stoichiometry obtained from SE is in agreement with that of the Raman spectroscopic results confirming the appropriateness of the model used for the former analysis.

2. Experimental methods

The Si films were deposited onto glass substrates (*Corning 7059*) by PLD technique at substrate temperature of RT, 400 °C and 700 °C. A high power Q-switched Nd:YAG laser (532 nm, 10 ns duration, 10 Hz) was focused to provide a laser fluence of ~2.5 J/cm^2 onto the Si (100) target under vacuum (~10^{-6} mbar). The films were deposited for 30 min deposition time. XRD (*Seifert* 3003 TT) spectra were recorded for analyzing the crystallinity of the Si thin films using the Cu K_α line. The 2θ was scanned in the range of 20° and 70° in angular steps of 0.03°. The Raman spectra of Si thin films were recorded at room temperature (RT) using micro-Raman setup (*Lab-Ram HR 800*) in back scattering geometry. The 488 nm line of Ar ion laser was used as excitation source. The Raman maps were recorded by scanning the film surface over an area of 30 × 30 μm^2 with a step size of 1 × 1 μm^2. The thickness of the thin film was measured by stylus profilometer (*Veeco Dektak 150*). The SE measurements were carried out over the spectral range of 1.32–5.00 eV using Variable Angle Spectroscopic Ellipsometer (*Semilab SOPRA: GES*-5E) equipped with goniometer at incident angles of 65°, 70°, and 75°. Data acquisition and analysis was performed using spectroscopy ellipsometry analyzer (*SEA*) software. The analysis of SE data was carried out by screening several realistic physical models for semiconductors to obtain best fit [15–18]. In the present work, a model with a combination of dispersion laws of Tauc-Lorentz and Lorentz gave the best fit for the films deposited at RT and 400 °C whereas for the 700 °C, Bruggeman's effective medium approximation (BEMA) was observed to be more appropriate model [10,18,19]. The optical band gap, refractive index, extinction coefficient and thickness of the films were also estimated from SE studies. The film thickness was compared to that of measured from surface profilometer.

3. Results and discussion

3.1. XRD spectra of Si thin films

Fig. 1(a) shows the XRD patterns of the Si thin film deposited at RT, 400 °C 700 °C and that of the bare substrate. All the thin films show characteristic XRD peaks for the Si (111), Si (220) and Si (311) planes at around 2θ ~ 28.45°, 47.37° and 56.20°, respectively confirming the polycrystalline nature of the films [20]. The broad peak observed in all samples from 20° to 35° could be attributed largely to amorphous Corning glass (SiO_2) substrate along with small contribution from a-Si present within the films (as ascertained from Raman studies discussed later). The respective individual contribution cannot be distinguished because of the nearly overlapping XRD peaks for both. It was observed that intensity of the peaks corresponding to c-Si initially increased from RT to 400 °C and thereafter slightly decreased at 700 °C. Fig. 1(b) exhibits the plot of integrated intensities corresponding to the peaks Si (111), Si(220) and Si(311) as a function of T_s, while the inset shows the

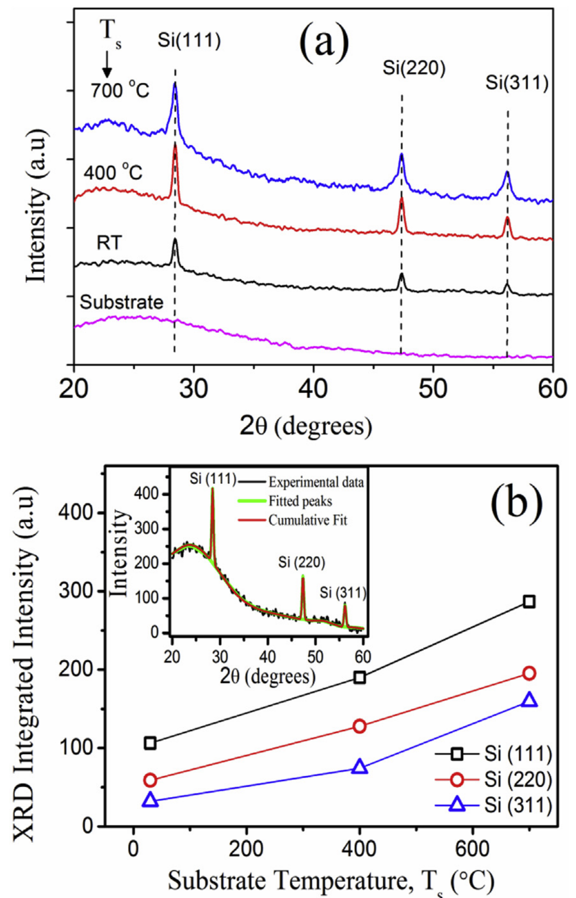

Fig. 1. (a) XRD spectra of the substrate and PLD Si thin films fabricated at laser fluence of 2.5 Jcm^{-2} at different T_s and (b) Plot of integrated intensity of XRD peaks corresponding to Si(111), Si(220) and Si(311) as a function of T_s. Inset shows de-convoluted XRD spectrum of Si film fabricated at RT.

de-convoluted XRD spectrum of Si film deposited at RT. The integrated intensities of XRD peaks of all the films were extracted from respective de-convoluted spectra. From this figure it was observed that the integrated intensities gradually increased with increasing T_s from RT to 700 °C inferring the increase in crystallinity of the film. An increased in surface diffusion at elevated substrate temperature might have enhanced the film nucleation sites resulting in the increase in crystallinity.

3.2. Ellipsometric studies of Si thin film

In any ellipsometric experiment, the changes in the state of the polarization of a monochromatic beam, upon reflection from the sample surface, is measured. The changes are expressed in terms of two parameters, ψ and Δ, by the following equation [21,22],

$$\rho = \frac{r_p}{r_s} = \left|\frac{r_p}{r_s}\right| e^{i(\Delta_p - \Delta_s)} = tan\psi e^{i\Delta} \tag{1}$$

where r_p and r_s are the Fresnel reflection coefficients for a plane wave polarized parallel (p) and perpendicular (s) to the plane of incidence, respectively while Ψ and Δ are the ellipsometric angles. Ψ is the angle whose tangent gives the ratio of the magnitude of the reflection coefficient of electric field components along the p and s polarizations and Δ gives the difference between the phase shifts of electric field components of p and s polarizations, Δ_p and Δ_s, experienced upon reflection. Thus the ellipsometric parameters, cos Δ and tan ψ, are the directly measurable quantities which are recorded as a function of wavelength at different angles of incidence. In the particular case of an abrupt interface between two semi-infinite media the ellipsometric data are

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