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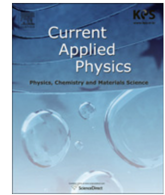

Low temperature plasma production of hydrogenated nanocrystalline silicon thin films

Tamila Anutgan*, Sema Uysal

Department of Physics, Karabuk University, 78050 Karabuk, Turkey

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ABSTRACT

The hydrogenated nanocrystalline silicon (nc-Si:H) thin films were produced by capacitively-coupled plasma enhanced chemical vapor deposition (PECVD) technique at low substrate temperatures ($T_s \approx 40$–200 °C). Firstly, for particular growth parameters, the lowest stable T_s was determined to avoid temperature fluctuations during the film deposition. The influence of the T_s on the structural and optical properties of the films was investigated by the Fourier transform infrared (FTIR), UV–visible transmittance/reflectance and X-ray diffraction (XRD) spectroscopies. Also, the films deposited at the center of the PECVD electrode and those around the edge of the PECVD electrode were compared within each deposition cycle. The XRD and UV–visible reflectance analyses reveal the nanocrystalline phase for the films grown at the edge at all T_s and for the center films only at 200 °C. The crystallinity fraction and lateral dark conductivity decrease with lowered T_s. FTIR analyses were used to track the hydrogen content, void fraction and amorphous matrix volume fraction within the films. The optical constants obtained from the UV–visible transmittance spectroscopy were correlated well with the FTIR results. Finally, the optimal T_s was concluded for the application of the produced nc-Si:H in silicon-based thin film devices on plastic substrates.

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1. Introduction

With the emerging flexible large-area electronics, the fabrication temperature of thin film devices (thin film transistors, solar cells, *pin* diodes, etc.) is continuously being lowered below the durability of plastic materials (<150 °C) [1,2]. As the active layer of these thin film devices, hydrogenated nanocrystalline silicon (nc-Si:H) is recently proposed instead of the industrially utilized hydrogenated amorphous silicon (a-Si:H) one, owing to its low photo-induced degradation, higher conductivity, etc. [2]. nc-Si:H film can be achieved by the post-deposition crystallization techniques of a-Si:H film, but they require high temperature processes which are not suitable for plastic substrates. That is why for plastic applications, nc-Si:H thin films should be obtained at low T_s by the direct deposition.

Previously, some depositions of intrinsic nc-Si:H thin films at low substrate temperatures (T_s) using various film growth techniques have been reported [3–8]. Mukhopadhyay et al. obtained nc-Si:H films in T_s range of 180–380 °C by radio frequency plasma enhanced chemical vapor deposition (RF PECVD) technique, revealing a void-rich structure and low crystallinity at $T_s = 180$ °C [3]. Waman et al. observed amorphous-to-nanocrystalline phase transition at $T_s = 300$ °C for nc-Si:H films grown by hot-wire CVD (HWCVD) at T_s ranging from 100 °C to 450 °C [4]. The RF sputtering deposition of nc-Si:H in a pure H_2 plasma at T_s varying from 50 °C to 250 °C lead to highly porous nanocrystalline phase with grain size (11–8 nm) and the disorder parameter decreasing from 260 meV to 129 meV with lowered T_s [5]. Cheng found that among the nc-Si:H films grown at T_s range of 90–230 °C by PECVD at 80 MHz excitation frequency, the highest crystallinity fraction was obtained at $T_s = 150$ °C, but the oxygen contamination affected its electrical properties [6]. Mani et al. showed that nc-Si:H films can be deposited at room temperature (RT) by an inductively-coupled plasma technique and the growth mechanism is related to H-induced crystallization of amorphous Si [7]. Chen et al. deposited nc-Si:H films by PECVD at very high RF power density (700 mW/cm^2) with T_s varied from 100 °C to 350 °C with 50 °C increments, and observed the increase in crystallinity for $T_s \geq 150$ °C, while the film grown at $T_s = 100$ °C was of amorphous phase [8]. Rahman obtained nc-Si:H films via pulsed PECVD technique at $T_s = 150$ °C, but their films possessed high conductivity due to the oxygen indiffusion from the ambient and T_s gradually rose ~40 °C above 150 °C during the film deposition process stemming from the heat transferred by the

* Corresponding author. Tel.: +90 370 433 83 74; fax: +90 370 433 83 34.
E-mail addresses: tamilaanutgan@karabuk.edu.tr, tamila.anutgan@gmail.com (T. Anutgan).

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bombarding radicals [9]. Other plasma depositions of nc-Si:H at RT were done with silane (SiH_4) heavily diluted by helium, argon and hydrogen (H_2) gas mixtures [10,11].

In this work, to fabricate nc-Si:H films, the industrially accepted capacitively-coupled PECVD process is utilized at high RF power density and high H_2 dilution of SiH_4. Unfortunately, as mentioned above, during the film deposition, high power density leads to the temperature rise in the substrates due to the high-energy ion bombardment. Therefore firstly, the lowest possible stable T_s for the previously optimized high RF power density [12] is determined by the film deposition at floating T_s. Afterward, the present study provides a systematic investigation on the effect of low T_s (80–200 °C) on the structural, optical and electrical properties of nc-Si:H thin films deposited in a capacitively-coupled PECVD system. In addition, at each T_s, the comparison study was done on the films grown directly under the gas inlet of our showerhead plasma reactor and away from it.

2. Experimental

The nc-Si:H thin films were firstly grown at floating T_s, i.e. the bottom electrode of the PECVD reactor (Fig. 1a) was not actively heated prior to and during the deposition process. It was found that after performing H_2 etching of the substrates just before the film deposition, the electrode temperature raised from RT up to 38 °C. Then, during the deposition process, lasting for 120 min, T_s is raised up to 70 °C without any external heating. This rise in T_s is probably associated with the high RF power density ($\sim$300 mW/cm^2) used in the growth of these films. It causes high-energy ion bombardment of the film surface, which locally heats the substrates so that T_s increases with the deposition time (t_d) [13]. The films obtained at floating T_s peeled off, which is likely to be associated with this temperature gradient that induces a stressful film structure. The time dependence of T_s was recorded in $\sim$3 min intervals and is provided in Fig. 1b. T_s rise is linear with time until 56 °C and has a logarithmic nature above 56 °C. When t_d is extrapolated for 180 min from the established logarithmic time dependence, T_s rises up to $\sim$77 °C. Therefore, the minimum stable T_s available for the film growth at the above specified power density during 100–160 min was chosen to be 80 °C.

To investigate the effect of T_s on the film properties, T_s was scanned from 80 °C to 200 °C with 40 °C intervals, i.e. four sets of films were deposited at 80 °C, 120 °C, 160 °C and 200 °C. Each deposition contained the film sets grown at the center of the PECVD electrode (directly under the gas inlet) and away from the gas showerhead, as it is depicted in Fig. 1a. To identify T_s and the position of the sample at the PECVD electrode during the film deposition process, the short notation is used throughout this work. For example, 'c-200 °C' and 'e-80 °C' notations are attributed to the films grown at the center of the PECVD electrode with $T_s = 200$ °C and at the edge of the PECVD electrode with $T_s = 80$ °C, respectively.

The film deposition rate (DR) was calculated via division of the film thickness (d) by t_d. d was deduced from both mechanical stylus profilometer (AMBIOS-XP2) and the UV–visible transmission spectrum (Perkin Elmer Lambda 2S spectrometer, 200–1100 nm). Refractive index (n), absorption coefficient (α) and d were resolved from UV–visible transmission spectra by the minimization program OPTICHAR [14]. The optical gap (here taken as an energy corresponding to $\alpha = 10^4$ cm^{-1}, E_{04}) and subgap absorption tail (Urbach tail, E_0) were determined from the strong absorption part of the spectrum.

The crystalline nature of the films was analyzed by Rigaku Miniflex X-ray diffractometer having Cu Kα radiation source with a wavelength (λ) of $\sim$1.54 Å, running at 30 kV and 15 mA. X-ray diffraction (XRD) patterns were obtained from the films grown on the glass substrates by scanning a reflection 2θ-angle (θ being the Bragg angle) from 20° to 60° with 0.05° steps and 1°/min scan rate. Since the largest peak intensity was obtained for (111) crystallite orientation, the grain size and relative crystalline volume fractions were estimated from Gaussian-fitted (111) XRD peak [12]. The crystallite grain size (L) was calculated from the Scherrer's formula $L = 0.89\lambda/(\beta \cos \theta)$, where β is the full width at half maximum (FWHM) of the XRD peak, and to compare the relative crystalline volume fraction, the intensity of the XRD peak was normalized by the thickness of each film [12]. Besides, the film crystalline phase was followed by the UV–visible diffuse reflectance spectrum,

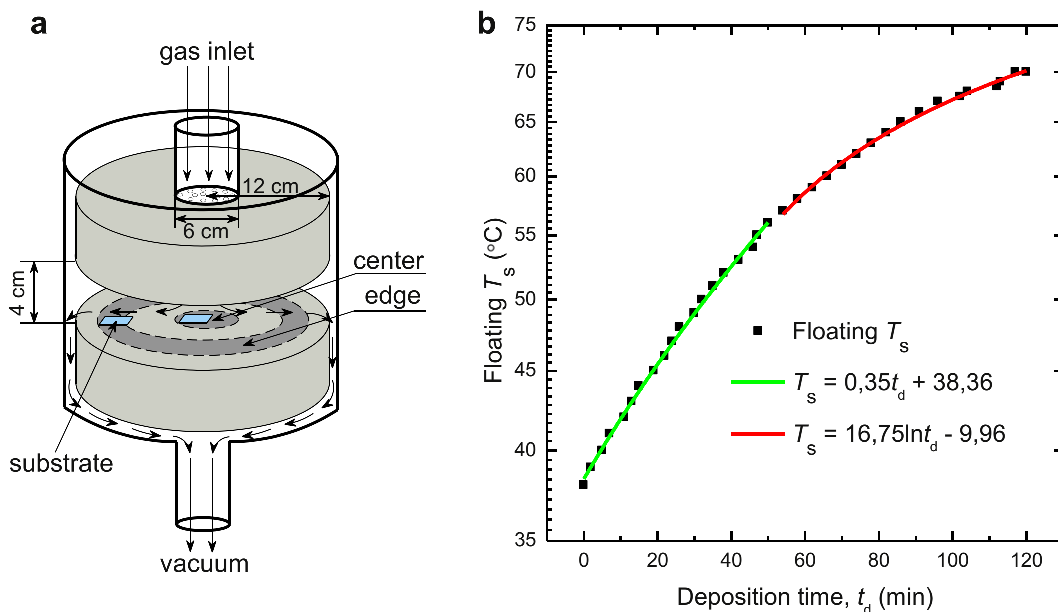

Fig. 1. a) Schematic of the PECVD system used for the nc-Si:H film deposition. The arrows show the gas flow directions. The sets of the substrates are placed at the center and near the edge of the bottom electrode, which is let floating or is heated actively up to the predetermined T_s. b) Floating T_s as a function of the deposition time for nc-Si:H thin films grown at 300 mW/cm^2 RF power density.

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