Excellent Response and Recovery Time of Photo-Detectors Based on nc-Si:H Films Grown by Using Hot Wire Method

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In present study, we have synthesized highly crystalline hydrogenated nanocrystalline silicon (nc-Si:H) thin films by hot wire method. The obtained thin films were characterized by using low angle-XRD, Raman spectroscopy, non-contact atomic force microscopy (NC-AFM) and UV-Visible spectroscopy. The low angle-XRD analysis revealed that the obtained nc-Si:H thin films are polycrystalline and have preferred orientation along (111) direction. The formation of nc-Si:H films was further confirmed by Raman spectroscopy analysis. The UV-Visible spectroscopy analysis showed that the synthesized films had a sharp absorption edge in the visible region and had a direct band gap of ~ 1.94 eV. Finally, nc-Si:H based photo-detector has been prepared at optimized process parameters which showed an excellent response time (1.79 s) and recovery time (1.71 s) along with responsivity ($\sim 9.8 \times 10^{-8}$ A/W), detectivity ($\sim 5.5 \times 10^4$ Jones) and quantum efficiency ($\sim 27.37 \times 10^{-6}$ %). The obtained results demonstrate a significant step towards nc-Si:H based photo-detector for broad band photo-detection applications.

Keywords: Hot wire method; nc-Si:H; Low angle XRD; Raman spectroscopy; Photo-detector

Received 10 April 2019, **Accepted** 5 August 2019

DOI: 10.30919/esmm5f236

1. Introduction

Photo-detectors or photo-sensors are essential in opto-electronic devices for a variety of applications, ranging from biomedicine to optical storage.^{1,2} Semiconductor materials from various groups such as group IV elements (Si, Ge), group III-V compounds (GaN, GaAs, InP), and group II-VI compounds (ZnS, ZnO, CdS, CdSe) are most commonly used in photo-detectors. Most of them require a relatively long response time and recovery time and thus limits their practical applications.

The crystalline silicon (c-Si) is the most commonly used in nano-electronics technology due to its enormous availability in the earth crust,³ high mobility and high temperature stability etc.⁴ Furthermore, c-Si has superior intrinsic concentration (10^{10} cm^{-3}) that leads to small leakage current, the most important aspect in nano-electronics device fabrications⁵ and maximum solid solubility of dopants ($\sim 10^{21} \text{ cm}^{-3}$) so that one can change the carrier type and concentration in a very large range for optimum operation of the devices.⁶ However, it fails for device applications in opto-electronics due to its indirect band gap. The nano-scale Si (nc-Si) materials with size dependent properties are potential candidates for photonic devices.⁷ The material has been successfully employed in solar cells,⁸ transistors,⁹ sensors,¹⁰ field emission,¹¹ water splitting,¹² super-capacitors,¹³ waveguides,¹⁴ coatings,¹⁵ photo-detection¹⁶ etc. There are several methods/approaches reported in literature for synthesis of device quality nc-Si thin films such as pulsed laser

deposition (PLD),¹⁷ atomic layer deposition (ALD),¹⁸ hot wire chemical vapour deposition (HW-CVD) or simply hot wire method,¹⁹ plasma enhanced chemical vapour deposition (PE-CVD),²⁰ sputtering²¹ etc. The inherent indirect band gap of c-Si (~ 1.1 eV) limits its ability to convert light below its band edge into photocurrent. The photocurrent can be generated only when the photon energy is slightly higher than the Schottky barrier height. In addition, the undesirable near infrared (NIR) photo-response due to small band gap causes additional optical noise but when coupled with optical filter technique increase structural complexity and cost dramatically.²²

*Knight et al.*²³ proposed a distinguished concept of using an active antenna for photo-detection well below the band edge of the semiconductor material. Using filtered cathodic vacuum arc (FCVA) technique *Tripathi et al.*²⁴ developed phosphorous doped hydrogenated amorphous silicon carbide (P doped a-SiC:H) thin film photo detectors at room temperature having fast response and recovery time. Later, using the same technique *Kesarwani et al.*²⁵ synthesized phosphorus doped amorphous/nanocrystalline silicon thin film photo detectors in the presence of hydrogen gas having good fast response and recovery time. Recently, *Tripathi and others*²⁶ reported nc-Si:H based photo-detector having fast response time (4.92 s) and recovery time (4.06 s) using filtered cathodic vacuum arc technique at room temperature. Due to quantum confinement the nano-sized Si based photodetectors are not only detect photons below its band edge but also provide a great potential for on-chip silicon photonics. With this motivation an attempt has been made to prepare device quality nc-Si:H films and to fabricate photo-detector device using it by hot wire method. The photo-detector shows excellent response time (1.79 s) and recovery time (1.71 s) along with responsivity (9.8×10^{-8} A/W), detectivity (5.5×10^4 Jones) and quantum efficiency (27.37×10^{-6} %). The obtained results open up a new

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opportunity for industrial application of broad band photo-detector based on nc-Si:H thin films.

2. Experimental

2.1 Synthesis of nc-Si:H thin films

Intrinsic nc-Si:H films were deposited on Corning #7059 glass in a locally fabricated hot wire system, details of which have been described elsewhere.²⁷ Films were prepared by using pure silane (SiH_4) as Si source gas and hydrogen (H_2) as dilution gas. The flow rate of SiH_4 and H_2 are kept constant at 2.5 sccm and 50 sccm respectively while filament temperature was kept constant at 1900 °C. The pressure during deposition was kept constant at 0.0667 mbar using manual throttle valve. The substrate temperature was held constant (350 °C) using a thermocouple and temperature controller and the filament-to-substrate distance (d_{s-f}) was fixed at 5 cm. The substrates were cleaned with double distilled water and loaded to the substrate holder. Then the deposition chamber was evacuated to the base pressure less than 1.3332×10^{-6} mbar. Prior to each deposition, the substrate holder and deposition chamber were baked for two hours at 100 °C to remove any water vapours absorbed on the substrates and to reduce the oxygen contamination in the film. After that, the substrate temperature was brought to desired value by appropriately setting the inbuilt thermocouple and temperature controller. The deposition was carried out for desired time and films were allowed to cool to room temperature in vacuum.

2.2 Characterization techniques

The transmittance and reflectance spectra of nc-Si:H films were measured using a JASCO, V-670 UV-Visible spectrophotometer in the range 300-1800 nm. The optical band gap of was deduced using the method followed by Tauc. Raman spectra were recorded with Renishaw microscope in the range 100-700 cm^{-1} . The spectrometer has backscattering geometry for detection of Raman spectrum with the resolution of 1 cm^{-1} . The excitation source was 632.8 nm line of He-Ne laser. The power of the Raman laser was kept to less than 5×10^{-3} W to avoid laser induced crystallization in the films. Low angle x-ray diffraction pattern were obtained by x-ray diffractometer (Bruker D8 Advance, Germany) using Cu K_α line ($\lambda = 1.54056 \text{ \AA}$). The patterns were taken at a grazing angle of 1° . The average crystallite size was estimated using the classical Scherrer's formula. The surface morphology of the films has been investigated using non-contact atomic force microscopy (NC-AFM) (Bruker MultiMode 8 HR, Germany). Thickness of films was determined by profilometer (KLA Tencor, P-16+) and was further confirmed by UV-Visible spectroscopy using the method proposed by Swanepoel.²⁸

3. Results and Discussion

The process parameters play a crucial role in determining the film properties in hot wire method. These parameters affect the film properties in different ways and in order to obtain desired film properties an optimum set of the parameters is to be selected. Although a particular set of optimized process parameters designed for a particular deposition system do not necessarily apply universally, but only serve as a good guideline.

Our group has analysed the influence of various hot wire process parameters on film properties to obtain device quality nc-Si:H films. Therefore, in this article, first we will be discussing the formation of nc-Si:H films at optimized process parameters (see Table 1) of our hot wire reactor. Later, we present the sensor properties of nc-Si:H based photo-detector.

3.1 Low angle XRD analysis

In order to confirm the formation of nc-Si:H film by hot wire method, low angle-XRD and Raman spectroscopy have been used. Fig. 1 displays the low angle-XRD pattern of Si:H thin film deposited under the optimum deposition parameters by using hot wire method. As seen from the figure, low angle-XRD pattern show three distinct peaks at 2θ 28.4°, 46.9° and 55.8° corresponding to (111), (220) and (311) crystal orientations of nc-Si:H, respectively. These results are consistent with the results reported in the literature^{29, 30} and support to the formation of nc-Si:H films by hot wire method. The dominant peak is (111), indicating that the crystallites in the film have preferential orientation in (111) direction.

Fig. 1 Low angle-XRD pattern of nc-Si:H deposited using hot wire method at optimized process parameters.

Table 1 Optimized process parameters to obtain device quality nc-Si:H films.

Process Parameter	Process Value
Filament Temperature (T_{fil})	1900 °C
Deposition Pressure (P_{dep})	0.0667 mbar
Substrate Temperature (T_{sub})	350 °C
Filament to Substrate Distance (d_{s-f})	5 cm
Deposition time (t)	20 Min

The average crystallite size ($d_{x\text{-ray}}$) of Si crystallites was deduced using Debye-Scherrer's formula,³¹

$$d_{x\text{-ray}} = \frac{0.9 \lambda}{\beta \cos \theta_B} \quad (1)$$

where, λ is the wavelength of diffracted radiation, θ_B is the Bragg angle and β is the line width (FWHM) in radians. The calculated value of crystallite size was found ~ 5 nm. These results are in consistent with the nc-Si:H films prepared by hot wire method at different filament temperatures by Gogoi *et al.*³²

3.2 Raman spectroscopy analysis

Raman spectroscopy is a very powerful and non-destructive characterization method used to investigate the structure of materials because it gives a fast and simple way to determine the phase of the material, whether it is amorphous, crystalline or nanocrystalline. Thus, to confirm the formation of nc-Si:H, Raman spectroscopy was used. Fig. 2 show Raman spectrum of Si:H film deposited by hot wire method at optimized process parameters. The Raman spectra show a satellite shoulder centred $\sim 514.17 \text{ cm}^{-1}$, associated with the Transverse Optic (TO) phonon peak originating from nanocrystalline phase.³³

To estimate the volume fraction of crystallites (X_{Raman}) and crystallite size (d_{Raman}), the Raman was de-convoluted into three peaks (Two Gaussian and One Lorentzian peak) with a quadratic base line using Levenberg-Marquardt method,³⁴ a crystalline peak ($\sim 520 \text{ cm}^{-1}$), an amorphous peak ($\sim 480 \text{ cm}^{-1}$) and an intermediate peak, ($\sim 510 \text{ cm}^{-1}$). The typical de-convoluted Raman spectrum is shown in the inset of the Fig. 1(c). The crystalline volume fraction (X_{Raman}) is then calculated by using the equation,³⁵

$$X_{\text{Raman}} = \frac{I_c + I_m}{I_c + I_m + \sigma I_a} \quad (2)$$

where σ is the ratio of Raman cross-section for the amorphous phase to the crystalline phase and is taken as 1.0,³⁶ I_c is the integrated intensity of the crystalline peak centered around 520 cm^{-1} , I_m is the integrated intensity of the intermediate phase or grain boundaries centered between $500\text{--}510 \text{ cm}^{-1}$ and I_a is the integrated intensity of the broad amorphous peak centered at 480 cm^{-1} .

The crystallite size (d_{Raman}) is then given by,³⁷

$$d_{\text{Raman}} = 2\pi \sqrt{\frac{\beta}{\Delta\omega}} \quad (3)$$

Fig. 2 Room temperature Raman spectrum of nc-Si:H deposited using hot wire method at optimized process parameters. The inset shows de-convoluted Raman spectrum for the estimation of volume fraction of crystallites and crystallite size.

where $\Delta\omega$ is the peak shift compared to c-Si peak located $\sim 520 \text{ cm}^{-1}$ and $\beta = 2.0 \text{ cm}^{-1}\text{nm}^2$.³⁸ The volume fraction of crystallite, X_{Raman} and crystallite size, d_{Raman} for the synthesized nc-Si:H film was found 75 % and 4.16 nm respectively. Similar results were obtained by Chong and his group³⁹ for nc-Si:H films deposited at different substrate temperatures by hot wire CVD. The results obtained in the present study are consistent with low angle XRD analysis which further confirm the formation of nc-Si:H film by hot wire method.

3.3 UV-Visible spectroscopy analysis

Fig. 3 shows the optical properties of nc-Si:H prepared using hot wire method at optimized process parameters. Fig. 3(a) display the transmittance spectra of nc-Si:H prepared using hot wire method in the range 300–1800 nm. As seen from the transmission spectra a sharp absorption edge has been observed in visible region, indicating good degree of crystallinity and low defect density near the band edge. Furthermore, the nc-Si:H has maximum transmission in IR region. The presence of interferences fringes in film transmittance spectra indicates that the film has smooth surface morphology. Atomic force microscopy analysis further supports this (discussed later).

The absorption coefficient (α) was estimated from transmission (T) and reflection (R) spectra in the wavelength range of 200–900 nm. The optical absorption coefficient (α) is calculated for each wavelength using the relation.

$$\alpha = \frac{1}{d} \ln \left(\frac{T}{1-R} \right) \quad (4)$$

Fig. 3 a) UV-Visible transmission spectra spectrum of nc-Si:H deposited using hot wire method at optimized process parameters and **b)** Plot of $(\alpha h\nu)^{1/2}$ versus photon energy ($h\nu$) (Tauc) to calculate the optical band gap.

where, d is the thickness of film. The optical band gap (E_{opt}) of the film was then calculated from the dependence of the absorption coefficient (α) on the photon energy ($h\nu$) taking into account for the semiconductor. For direct allowed transition, absorption coefficient (α) and band gap can be expressed as,⁴⁰

$$(\alpha h\nu)^n = B(h\nu - E_g) \quad (5)$$

where, B is an energy independent constant and n is a constant which determines type of optical transition ($n = 1/2$ for direct transition and $n = 2$ for indirect transition).⁴¹ To determine the band gap, $(\alpha h\nu)^{1/2}$ is plotted against energy $E (= h\nu)$ (Tauc plot). A straight line fitted through the data points. A straight line fitting program was developed to fit the data in a straight line. Then the line is extrapolated and the intercept of the extrapolated line on the energy axis gives the band gap. The inset of Fig. 3(b) depicts typical Tauc plot used to estimate the optical band gap (E_{Tauc}) of nc-Si:H. The calculated value of optical band gap of nc-Si:H film was found ~ 1.94 eV. The value of optical band gap matches with recently reported values for nc-Si:H films measured by spectroscopic ellipsometry (SE).⁴²

3.4 Atomic force microscopy (AFM) analysis

To study the morphology of nc-Si:H film, atomic force microscopy (AFM) measurements were performed in non-contact mode on the films

deposited on corning glass at optimized process parameters. Fig. 4 shows the surface morphology of the nc-Si:H on $2\ \mu\text{m} \times 2\ \mu\text{m}$ area of the films.

It is clearly observed from the AFM image that the film is very uniform throughout the sample. The film consists of well resolved, large number of nearly spherical and uniform nano-crystallites each having an individual identity with its size in the range of ~ 20 -25 nm. The root mean square (RMS) surface roughness of nc-Si:H was estimated from AFM image using Scanning Probe Image Processor (SPIP) software. The RMS surface roughness values of the films obtained from AFM analysis was found 39 nm. *Gope et al.*⁴³ also reported similar average crystalline size (~ 35.5 nm) and RMS surface roughness (~ 11.32 nm) for nc-Si:H films deposited at low process pressure (1.3332 mbar) using very high frequency plasma enhanced chemical vapor deposition (VHF PECVD) process.

3.5: Photo-detector properties

The photo-detector is fabricated by directly depositing nc-Si:H on corning glass using the optimized process parameters listed in Table 1. The thickness of nc-Si:H layer is ~ 900 nm. Using metal mask silver (Ag) source and drain electrodes were deposited on nc-Si:H by vacuum evaporation technique. The schematic of nc-Si:H photo-detector device fabricated using optimized process parameters of hot wire method is shown in Fig. 5.

Fig. 4 NC-AFM image of nc-Si:H film deposited by hot wire method at optimized process parameters.

Fig. 5 Schematic of nc-Si:H photo-detector fabricated using hot wire method at optimized process parameters.

The performance of nc-Si:H photo-detector was evaluated using Keithley 2612 source meter connected to PC through GPIB 488 A interface. For white light illumination PEC-L01 portable solar simulator was used which provide wide range of spectrum from Ultraviolet (UV) to near infrared (IR) region. In the present study, white light having intensity 100 mW/cm^2 have been employed for the photo-detector property measurements. The VIBGYOR filters were used to investigate wavelength dependant photo-detection investigation of nc-Si:H photo-detector.

In many reviews and articles, the working principle of the photo-detector is explained in detail.⁴⁴⁻⁴⁶ Basically, it is a sensor that is capable of converting incident light into electrical signal. Due to the incident light energy, electrons at the semiconductor surface get excited to new energetic conductive levels by leaving a hole behind. This photon excitation leads to generation of exciton (electron-hole pair, EHP). After the application of the electric field, the electron and hole drifts inside the crystal lattice and generate photocurrent. Sensitivity, response and decay time measurements are very important parameters in order to characterize the performance of the photo-detector.

Fig. 6 display current-voltage (I-V) characteristics of nc-Si:H

photo-detector under dark and illumination of different light (VIBGYOR) filters in the range of -1 V to 1 V.

It is clear from the graph that the resistance of nc-Si:H decreases after illumination of light. The photocurrent strongly depends on electron-hole separation and thermal mechanisms.⁴⁷ On absorption of photons the photo-detector material, nc-Si:H create extra charge carriers, electrons and holes which drift in opposite directions towards source-drain metal leads. This reduces the electrical resistance and results in a net increase in the photo-current. These results confirms the semiconducting behaviour of nc-Si:H by hot wire method.

The time dependent current response of nc-Si:H photo-detector for repeated cycles is illustrated in Fig. 7. Fig. 8 shows a single-cycle I-t curve for measuring the response time (τ_{rise}) and recovery time (τ_{decay}) of nc-Si:H photo-detector using white light. Response time is defined as the time required for a photo-detector reaching 90 % of its maximum photocurrent value from its dark current value. Similarly, decay time is defined as the time required for photo-detector reaching 10 % of maximum photocurrent response.⁴⁸ The response and recovery time for nc-Si:H photo-detector under white light were found to be $\sim 1.79 \text{ s}$ and $\sim 1.71 \text{ s}$ respectively.

Fig. 6 I-V characteristics of nc-Si:H photo-detector under dark and under VIBGYOR filters at applied voltage of 1 V.

Fig. 7 Current-time (I-t) characteristics of nc-Si:H photo-detector upon illumination of white light for repeated cycles.

The obtained response and recovery time for nc-Si:H photo-detector are compared with some other photo-detector materials and corresponding fabrication methods are listed in Table 2.

As evident from Table 2 that the nc-Si:H based photo-detector has excellent response and recovery time compared to other photo-detector materials.

The photo-sensitivity of photo-detector (S %) is defined as the ratio between the difference in photo current (I_{photo}) and dark current (I_{dark}) values to the dark current,⁴⁸

$$\text{Sensitivity (S \%)} = \frac{I_{\text{photo}} - I_{\text{Dark}}}{I_{\text{Dark}}} \times 100 \% \quad (6)$$

Fig. 9 shows sensitivity of photo-detector obtained at different colours of the radiation spectra.

As seen from the figure for hot wire deposited nc-Si:H photo-detector has the highest sensitivity (24 %) for the white light and lowest for the green light (12 %).

Photo-responsivity, photo-detectivity and external quantum efficiency measurements are very important parameters in order to describe the performance of photo-detector. The photo-responsivity (R_s) refers to the photo-excited current generated per unit power of incident light on the effective area of the photo-detector and is given as,⁵³

$$R_s = \frac{\Delta I}{\omega \times A} \quad (7)$$

where $\Delta I = (I_{\text{photo}} - I_{\text{dark}})$ is the effective change in the emission current upon visible photon irradiation, ω is the intensity of incident photons (100 mW/cm^2) and A is area of photo-detector (0.25 cm^2). The photo-responsivity of the nc-Si:H for visible illumination is found to be $9.8 \times 10^{-8} \text{ A/W}$.

To evaluate the efficiency of the electron transport and carrier collection we have estimated the probable quantum efficiency (QE) of nc-Si:H photo-detector using the equation,⁵⁴

$$\text{QE (\%)} = \frac{h \times c \times R_s}{e \times \lambda} \times 100 \% \quad (8)$$

where h is a Planck's constant, c is a velocity of light; e is charge of the electron and λ is a wavelength of the incident radiations. The quantum efficiency of nc-Si:H photo-detector was found $27.37 \times 10^{-6} \%$.

For photo-detectors one of the essential parameter is detectivity (D^*), which gives a measure of quality of device. It helps in comparing distinct devices with different area and geometries. It is measured in the unit of Jones and is given by,⁵⁵

$$D^* = \frac{R_s}{(2 e \times J_{\text{Dark}})^{1/2}} \quad (9)$$

Fig. 8 Single-cycle I-t curve for measurement of response and recovery time of nc-Si:H photo-detector.

Fig. 9 Sensitivity of nc-Si:H photo-detector obtained under the illumination of different colours.

Table 2 Response and recovery time of recent photo-detector materials and their fabrication method.

Material	Synthesis method	Response time	Recovery time	Reference
	-	20 s	10.97 s	[49]
MoS ₂	Chemical vapour deposition	1.79 s	1.23 s	[50]
WS ₂	PLD	4.1 s	4.4 s	[51]
	Hot wire method	51 ± 5 s	88 ± 6 s	[27]
SnSe	Chemical vapour deposition	8.2 s	5.3 s	[52]
nc-Si:H	Hot wire method	~ 1.79 s	~ 1.71 s	Present

where, J_{Dark} is the dark current density. We observed the value of detectivity ($D^* \sim 5.5 \times 10^4$ Jones) for nc-Si:H photo-detector. Noise from the dark current, Johnson noise, generation-recombination noise, and thermal fluctuation (flicker noise) limits the detectivity.⁵⁶

Conclusion

We have synthesized hydrogenated nanocrystalline silicon (nc-Si:H) thin films using hot wire method. Formation of nc-Si:H have been confirmed by low angle-XRD and Raman spectroscopy analysis. The low angle-XRD and Raman spectroscopy analysis revealed that nc-Si:H thin films are highly crystalline and crystallites have preferred orientation along (111) direction. Formation of nc-Si:H thin films were also confirmed by non-contact atomic force microscopy which formation of well resolved, large number of nearly spherical and uniform Si nano-crystallites having average size in the range of ~ 20 -25 nm. The UV-Visible spectroscopy analysis showed that the synthesized films have sharp absorption edge in visible region and has direct band gap of 1.94 eV. The photodetector fabricated using nc-Si:H shows excellent response time (1.79 s) and recovery time (1.71 s) along with responsivity ($\sim 9.8 \times 10^{-8}$ A/W), detectivity ($\sim 5.5 \times 10^4$ Jones) and quantum efficiency ($\sim 27.37 \times 10^{-6}$ %). The obtained results reveal potential applications of hot wire deposited nc-Si:H as a photo-detector.

Acknowledgement

Subhash Pandharkar, Shruthi Nair, Ravindra Waykar, Ashish Waghmare and Dhirsing Naik are thankful to the Ministry of New and Renewable Energy (MNRE), Government of India for the financial support under National Renewable Energy Fellowship (NREF) program. Ajinkya Bhorde is thankful to Department of Science and Technology (DST), Government of India for INSPIRE Ph. D. fellowship. Rahul Aher is thankful to Savitribai Phule Pune University, Pune for the award of Bharatratna J. R. D. Tata Gunwant Sanshodhak Shishyavrutti. Haribhau Borate is grateful to University Grant Commission (UGC), New Delhi for providing Teacher Fellowship under faculty development programme. One of the authors Sandesh Jadkar is thankful to University Grants Commission (UPE program), New Delhi and Indo-French Centre for the Promotion of Advanced Research-CEFIPRA, Department of Science and Technology, New Delhi for special financial support.

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