Studies on optical signal due to oxygen effect on hydrogenated amorphous/crystalline silicon thin films

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Abstract

We have studied the effects of oxygen on hydrogenated amorphous/crystalline silicon films in terms of their structural and optical properties. Different “hydrogenated silicon oxide” (SiO:H) and “silicon” (Si:H) films are fabricated between microcrystalline and amorphous transition region. X-ray diffraction, Raman, FTIR and UV–VIS emission spectrometry have been used to characterize different films. A comparison of the results with those of different types of films like “hydrogenated amorphous silicon oxide” (a-SiO:H), “hydrogenated amorphous silicon” (a-Si:H) and “microcrystalline silicon” (μc-Si:H) films reveal their superiority as an excellent substance for solar cell. X-ray diffraction, FTIR, and Raman spectral analysis show that difference of the H dilution effect has a major effect on the structure of the film and the optical properties. Photoluminescence analysis of amorphous silicon–oxygen and silicon-hydride alloy films has established their efficient application appropriate as Si-based light-emitting devices. A large optical band gap of 1.83 eV and appearance of strong photoluminescence at 2.0 eV validates the applicability of a-SiO:H film as a better alternative for the solar cells.

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1 Introduction

Amorphous silicon is a very well known base material for higher conductivity. Out of different noncrystalline semiconductor materials like: selenium, etc., though they have very good electronic properties, they are proved unsuitable for making of efficient solar cells [1]. After the work of Chittick et.al [2], it was proved that as crystalline silicon, amorphous silicon can also have a high conductivity in the presence of some doping. The first industrial solar cell was reported by doped amorphous silicon on 1976 by Christopher Wronski [3]. Normally, hydrogen atoms are found in amorphous silicon in a considerable fraction. These hydrogen atoms play an important role for the enhancement in the electronic properties of amorphous silicon structure. It is generally known as “hydrogenated amorphous silicon (a-Si:H)” [4]. Photovoltaic (PV) technology based on a-Si:H is exceptional compared with other available PV technologies [5]. The importance of a-Si:H comes into the picture due to its promising optical properties for collecting solar energy. The technology based on a-Si:H is also very easy and low cost in comparison with other available technologies for making crystals such as crystalline Si (c-Si) [6]. Due to the amorphous nature of a-Si, it absorbs sunlight extra powerfully than other crystalline and polycrystalline silicon (c-Si and poly-Si). The selection rules, which weaken the absorption in c-Si an indirect band gap type of semiconductor, which is not applicable to a-Si. “Band gap” of a-Si is considerably larger than c-Si, which reduces optical absorption of c-Si than a-Si [7, 8] of same thickness. To absorb the same energy, a c-Si layer needs to be much thicker. This indicates that solar cell composed from a-Si required less material than from c-Si. a-Si can be made at a very low temperature on inexpensive substrate like glass and so the product can be made through a low-cost process. The thickness of the absorbing layers in a-Si solar cells is less than 1 micron. Another important quality of a-Si selected substrates is that it can be made light weight and flexible. These effects are very significant for numerous applications. A positive temperature coefficient is also observe for output power of a-Si PV products. In area having extra
sunshine, a-Si-based devices observed to possess higher efficiency at higher temperature. The conductivity of a-Si can be modified by the effect of p-type or n-type doping on it. Normally, increased doping indicates the better conductivity due to the higher concentration of carriers. For n-type materials, conductivity can be induced due to mobile electrons, whereas for p-type material, the induced conductivity appears due to mobile positively charged holes. The energy payback time is also observed to be very less in a-Si products as compared to c-Si [9]. For thin-film silicon solar cell, silicon usually is deposited on a glass substrate. There are lots of research works have been reported on hydrogenated µc-Si:H thin-film solar cells [10, 10, 11] till now, research works on a-Si solar cells are very few. Out of differently available hydrogenated amorphous phase silicon, one is hydrogenated amorphous silicon oxide (a-SiO:H), where oxygen is attached with silicon. SiO:H films have wide band gap over Si:H films. The reason of the wide band gap is the presence of oxygen. Presence of oxygen increases the band gap of silicon [12]. Oxygen-rich phase in SiO:H films helpful to increase band gap, whereas the silicon-rich phase in SiO:H films contributes towards higher conductivity [13]. a-SiO:H is an fascinating material, because it can introduce as doped layer leading an increase in the efficiency and hence performance of the solar cell by reducing the recombination losses at interlayers and increasing the built-in potential [14]. Many research works have been reported on optical (optical absorption coefficient, integrated solar irradiance) and electrical (band gap, voltage, efficiency) properties of hydrogenated amorphous silicon solar cells which get affected by its oxygen content [15, 16] in the form of a-SiO:H. In 2015 Hairen Tan et al. [17] have been studied performance of a wide band gap p-type nanocrystalline silicon oxide for high thin-film silicon multi-junction solar cells, In 2016, Simrov et al. [18] Studied the optoelectronic properties and the chemical and structural composition of the doped µc-SiO_{x}:H. In 2016 M. Klingsporn and their research group investigated the chemical, electrical, and optical properties of phosphorous doped nanocrystalline silicon suboxides (nc-SiOx) by using plasma enhanced chemical vapor deposition (PECVD) deposition technique [19]. But very few information about the properties of a-SiO:H has been explored till now.

Generally, PECVD technique is used to deposit hydrogenated silicon thin films by glowing discharge decomposition of silane [20, 21]. This silane gas is diluted with hydrogen and have many significant impact on the characteristics of a-Si:H films [22, 23]. By changing the deposition parameters such as chamber pressure, gas mixture composition, flow rates, RF power density and substrate temperature, one can change structure of the films. Depending on the deposition parameters, amorphous, micro/nano crystalline and procrystalline [24] structures of hydrogenated silicon (Si:H) films can be prepared. These different Si:H structures can be used for enhancing the performance of silicon-based thin-film solar cells.

In the present work, we have synthesized and characterized some doped and intrinsic hydrogenated amorphous and microcrystalline SiO:H and Si:H thin films on glass substrate. We have developed some n- and p-type µc-SiO:H and amorphous thin films having suitable characteristic properties used for the fabrication of amorphous silicon (a-Si) solar cells. The SiO:H and Si:H thin films described in the present study were deposited on glass substrates via PECVD at a plasma frequency of 13.56 MHz (RF). We will try to characterize these films to describe some important aspects of their electrical and optical properties in view of their probable application as solar cells. In order to characterize the doped and intrinsic hydrogenated amorphous and microcrystalline SiO:H and Si:H thin films, XRD analysis, Fourier transform infrared (FTIR) spectroscopy, Raman spectroscopy, UV–visible absorption, Photoluminescence analysis, were. This work is also the first systematic study of the hydrogen dilution and oxygen incorporation effects on the structure of amorphous and microcrystalline silicon films. We have also reported the results in view of comparison of SiO:H with Si:H film.

2 Materials and methods

2.1 Materials

Different types of silicon-based materials used in the present study have been prepared by using “five chamber cluster tool plasma enhanced chemical vapor deposition (PECVD) unit” [25] at a plasma frequency of 13.56 MHz (RF). The power density of the substrate was fixed at around 300 mW cm^{-2}. The same chamber has been used to grow n-doped a-Si:H and µc-Si:H. Phosphine (PH_{3}) (1%) in SiH_{4} and H_{2} gases were used to grow a-Si:H and µc-Si:H films. To grow µc-SiO:H films, CO_{2} gas was employed [25]. Deposition conditions, such as the pressure and volume fraction plays a significantly role in the properties of materials. For both SiO:H and Si:H films, optimized deposition pressure condition is 1 torr and PH_{3} as 0.04. Deposition parameters in the PECVD process, for SiO:H films SiH_{4}/H_{2} as 1:60 and CO_{2}/SiH_{4} as 2 for Si:H films SiH_{4}/H_{2} as 1:45. Since the material properties are thickness dependent. Hence, for the present work, we kept the thickness fixed for all the films at ~600 Å.
2.2 Experimental methods

For measuring X-ray diffraction (XRD) patterns, “Bruker D8 Advance diffractometer has CuKα” as radiation source have been used. Raman spectra have been measured with “Raman spectrometer equipped with a SPEX TRIAX 550 monochromator (model GDL-5015 L, 8 mw)” and a “liquid-nitrogen-cooled charge-coupled device (CCD; Spectrum One with CCD 3000 controller)”. Spectral acquisition time and spectral resolution was 1 min and 2 cm\(^{-1}\), respectively, for the Raman system. FTIR spectra were recorded with a “Perkin-Elmer FTIR spectrophotometer (model Spectrum BX-II source: nichrome glower wire with DTGS detector)”. The spectral resolution of FTIR spectrometer was 2 cm\(^{-1}\) with a range 400–4000 cm\(^{-1}\). UV–VIS absorption spectra were measured with a “Perkin-Elmer absorption spectrophotometer (model Lamda-35, source: tungsten iodide and deuterium) in the 200–1100 nm spectral range with a 0.5 and 1.0 nm slit width. The UV–VIS fluorescence spectra were recorded with “Perkin-Elmer luminescence spectrophotometer (model Fluorescence-55 source: xenon)” in the 200–900 nm spectral range.

3 Results and discussion

We have discussed the properties of a-SiO:H and a-Si:H films, that are placed on the top of glass substrate. Figure 1 shows the XRD spectra of these films as compared with the XRD spectra of crystalline Si wafer and glass. Normally, Si wafer shows five distinct peaks at 21°, 39.3°, 43°, 58° and 69.4° correspond to \(\langle 111 \rangle\), \(\langle 200 \rangle\), \(\langle 220 \rangle\), \(\langle 311 \rangle\) and \(\langle 400 \rangle\) planes of crystalline silicon phases [26], whereas glass shows only a wide, band around 20.8° due to its amorphous phase [27].

In the presence of different a-Si films, both the samples show different characteristics. In a-Si:H film in addition to all silicon crystalline phases a wide peak at 22.1° appears due to the amorphous phase of silica, whereas in a-SiO:H film due to excess oxygen content the crystalline nature of Si disappears totally and amorphous nature dominates with the appearance of only one wide band at 22.2°.

Raman spectra of different doped crystalline and amorphous SiO:H and Si:H films have been taken (Fig. 2). A broadband centered at 490 cm\(^{-1}\) in SiO:H films depicted the amorphous phase of it. Shift from a-SiO:H to \(\mu\text{-Si} \) growth shown by an emergence of thin and sharp band at 528 cm\(^{-1}\). When we compare Raman intensities of the SiO:H films with Si:H films, we observe that with increasing H ratio the comparative intensity of the crystalline peak increases. This clearly indicates the good conformity with the results obtained from X-ray diffraction pattern. The whole Raman spectrum divided into three components as crystalline component, amorphous component...

![Fig. 1 XRD pattern of crystalline silicon, glass, amorphous a-SiO:H and a-Si:H films placed on top of glass](image1)

![Fig. 2 Raman spectra of doped “SiO:H” and “Si:H” films placed on the top of the glass substrate](image2)
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and intermediate component. The peak is observed at 528 cm\(^{-1}\) due to crystalline component, peak at 485 cm\(^{-1}\) due to amorphous component and intermediate component peaked between the ranges 495–520 cm\(^{-1}\) that is associated with bond dilation at grain boundaries. The Raman peak intensity at 528 cm\(^{-1}\) of the doped glasses (\(p\)-\(\mu\text{-SiH}\) and \(i\)-\(\mu\text{-SiH}\)) is quite high as compared to the undoped glass (Fig. 2), whereas the Raman peak intensity at 485 cm\(^{-1}\) (due Si–H stretching) of the undoped glass is highest and disappears with both \(n\)- and \(p\)-type of doping. This vanishing of the Raman peak is due to the absence of Si–H stretching mode in doped glasses.

By using Raman spectral results, the crystalline volume fraction \(X_c\) have been deduced as:

\[
X_c(\%) = \left(\frac{I_c}{I_c + I_{i} + I_{a}}\right) \times 100
\]  

where \(I_c\) is the integrated intensities of the crystalline \(I_i\) is the integrated intensities of the amorphous and \(I_{i}\) is the integrated intensities of the intermediate peaks. The observed result shows that \(X_c\) gradually increases from amorphous to microcrystalline SiO:H films. Also \(X_c\) increases monotonically from 63.5 to 89.3% from \(\mu\text{-SiO:H}\) to \(\mu\text{-Si:H}\) films as the H dilution increases to the \(\mu\text{-Si}\) growth which reflects with the narrow band appearance at 528 cm\(^{-1}\).

Similarly, the grain boundary volume fraction \(X_i\) has been calculated by the equation [28]

\[
X_i(\%) = \left(\frac{I_i}{I_c + I_{i} + I_{a}}\right) \times 100
\]  

It is found that as H dilution increases the \(X_i\) decreases from 35.2 to 29.1%. So an opposite behaviour is observed in grain size and crystalline volume fraction with increase the phase from amorphous to crystalline as well as with increasing H dilution (Table 1).

The bonded hydrogen content and the effects of oxygen atom have been also studied from the FTIR absorption spectra as shown in Fig. 3. Structural characterization of amorphous films is extremely difficult. According to the “random bonding model (RBM)” [29], “unhydrogenated silicon oxide films (SiO\(_x\)) are composed of five basic bonding configurations”, Si \((\text{Si}_{4-n}\text{O}_n)\), \(n = 0–4\) with basic bonding units as: \(\text{Si}_2\text{O}\), \(\text{SiO}\), \(\text{Si}_2\text{O}_3\), \(\text{SiO}_2\).

Table 1 Comparison of Raman, UV–VIS data for SiO:H and Si:H films

| Nature of film | Raman shift (\(\delta\)) in(cm\(^{-1}\)) | Volume fraction from Raman(\%) | Optical band gap (\(\Delta\varepsilon\)) in (eV) |
|---------------|--------------------------------------|-------------------------------|-----------------------------------------|
| \(\mu\text{-Si:H}\) | 528                                   | 89.3                          | 29.11                                   |
| \(a\text{-Si:H}\) | 490                                   | 63.5                          | 32.27                                   |
| \(\mu\text{-SiO:H}\) | 490                                   | 70                             | 35.21                                   |
| \(a\text{-SiO:H}\) | 490                                   | 70                             | 35.21                                   |

According to modified RBM, in hydrogenated films \((\text{a-SiO}_x\text{:H})\) the incorporation of Si–H bonds can be performed by replacing an Si nearest neighbor to the Si site by an H atom in Si. In hydrogenated Si film, Si–H stretching absorption peaks correspond to Si–H stretching is observed at 2337 cm\(^{-1}\) and 2365 cm\(^{-1}\). Due to the attachment of oxygen atom, the peak get shifted to a higher wave number side associated with \(\text{H-Si}(\text{Si}_{3-n}\text{O}_n)\) for \(n = 0, 1, 2, 3\), respectively, with an appearance of new broad peak at 2000 cm\(^{-1}\). Therefore, the Si–H stretching band can be dissolved into three components at 2000, 2337 and 2365 cm\(^{-1}\) due to \(\text{H-Si}(\text{O}_2\text{Si})\), \(\text{H-Si}(\text{Si}_2\text{O}_3)\), \(\text{H-Si}(\text{O}_3)\). Again a new band appears in a-SiO:H films at 1386 cm\(^{-1}\) due to \(\text{Si-O-Si}\) symmetric stretching vibration which is absent in hydrogenated silicon.

The crystalline and grain size, volume fraction behaves differently with the increment of H dilution. It is not obvious that how the optical and electronic properties of the films will change with changing the structure of the films from SiO:H to Si:H. We have carried out UV–VIS absorption and luminescence studies. Figure 4 shows the absorption spectra of both the films.

To calculate the optical band gap, we have been used the tauc model. It uses experimental spectroscopic absorbance from the Fig. 4 we can estimate that band gap (\(\Delta\varepsilon\)) of a-SiO:H film comes out to be 1.83 eV, whereas \(\Delta\varepsilon\) for a-Si:H observed as 1.6 eV which perfectly matches with the conventional literature [30] values. According to the Watanabe model [31] SiO:H belongs to a two phase material having an island of SiO in a matrix of a-Si:H. The two-phase model suggested that the oxygen rich phase is
efficient in optical band gap increment. This is one of the advantages that why a-SiO:H films are widely used as the window layer of the solar cells in research and development level. Also from the available atom probe tomography method [32] a complex three-dimensional model of nc-Si gives a conductive paths for the electrical current in both the direction (lateral and transversal). While an oxygen rich phase suggested a high optical band gap and a low refractive index. By adjusting this microstructure a extensive variety of properties can be achieved for nc-SiOx:H layers.

SiO:H and Si:H films have tunable optical band gap. The red, blue colored emission and their combination mechanism from silicon-based materials have been studied by different researchers [33–37]. Kanashima et al. [38] have been reported dual emission at 3.5 eV (354.24 nm) and 4 eV (309.96 nm) from SiO2 films. But there are very few studies on “a-SiO:H” and “a-Si:H” films. Figure 5 shows different a-SiO:H and a-Si:H films photoluminescence spectra with excitation at 3.68 eV (336.91 nm) at room temperature. a-SiO:H film shows two strong emission peaks at 2.05 eV (604.80 nm) and 2.10 eV (590.40 nm) with two weak shoulders at higher energies at 2.34 eV (529.84 nm) and one at 1.60 eV (774.90 nm) lower energy side, respectively, whereas in a-Si:H film the emission band at 2.05 eV (604.80 nm) disappears with the existence of only one strong emission at 2.09 eV (585 nm).

The two remaining shoulders exists at the same positions as that of a-SiO:H film at higher energies at 2.34 eV (529.84 nm), and 1.60 eV (774.90 nm), respectively. Due to the presence of oxygen content in a-SiO:H film, a redshift in the emission peak at 2.10 eV (590.40 nm) is observed. So the luminescence (visible) properties associated with excitation (3.68 eV or 336.91 nm) from a-SiO:H and a-Si:H films differ with the available oxygen content.

4 Conclusion

We have studied the property of oxygen on the structural and optical properties of the different doped and undoped hydrogenated a-SiO:H and a-Si:H films. This work systematically study of amorphous Si and microcrystalline Si structure of films based on the hydrogen dilution and oxygen incorporation effects on them. Different XRD, Raman and FTIR spectral analysis display that difference of the H dilution effect has a major effect on the structure of the film and the optical properties. A structural change from a-Si to μc-Si is caused by the increasing H ratio. This structural change creates a regular alter of the electronic structures which is observed by the UV–VIS absorption and photoluminescence spectroscopy. We have also observe a very remarkable type of the high energy (cm⁻¹) Raman from μc-Si phase to a-Si phase both in SiO:H and Si:H. Major optical change we observed of higher cm⁻¹ Raman from μc-Si phase such as energy blue
shift and decrement in the intensity and increment in the band width to amorphous phase. The same observation has been verified by the FTIR data. The observable photoluminescence (visible) properties with excitation (3.68 eV) from different hydrogenated Si films changes with oxygen content. The decrease in 2.05 eV emission band intensity observed in a-Si:H is tentatively due to the absence of oxygen. In conclusion, combining XRD, UV–VIS, Raman and IR spectra give us a rich information about the SiO:H and Si:H films which are very crucial to know different optoelectronic properties of μc-Si and a-Si films.

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