Conference Paper

Raman and FTIR Studies on PECVD Grown Ammonia-Free Amorphous Silicon Nitride Thin Films for Solar Cell Applications

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Ammonia- (NH$_3$-) free, hydrogenated amorphous silicon nitride (a-SiN$_x$:H) thin films have been deposited using silane (SiH$_4$) and nitrogen (N$_2$) as source gases by plasma-enhanced chemical vapour deposition (PECVD). During the experiment, SiH$_4$ flow rate has been kept constant at 5 sccm, whereas N$_2$ flow rate has been varied from 2000 to 1600 sccm. The effect of nitrogen flow on SiN$_x$:H film has been verified using Raman analysis studies. Fourier transform Infrared spectroscopy analysis has been carried out to identify all the possible modes of vibrations such as Si–N, Si–H, and N–H present in the films, and the effect of nitrogen flow on these parameters is correlated. The refractive index of the above-mentioned films has been calculated using UV-VIS spectroscopy measurements by Swanepoel’s method.

1. Introduction

Amorphous silicon nitride (a-SiN$_x$) thin films deposited by PECVD are considered to be one of the most promising materials in semiconductor industry as gate dielectrics, isolation materials, diffusion barriers, and acoustic wave devices on semiconductor substrates [1, 2]. Also a-SiN$_x$ layers are widely used as antireflection coatings, bulk, and surface passivation layers in solar cells applications [3, 4]. The main advantage of a-SiN$_x$ thin films grown by PECVD is the low process temperature and high deposition rate. Low process temperature is a crucial parameter in the fabrication of multocrystalline silicon solar cells, where high-temperature thermal processing may cause degradation of the bulk life time of the charge carriers and thereby decreasing the efficiency of the solar cell [5]. Also the amorphous silicon nitride films grown by PECVD are nonstoichiometric in nature, that is, SiN$_x$ with $x \neq 4/3$ [6]. Adjusting the deposition process parameters can vary the actual composition of the films. Many researchers investigated the structural, optical, and electrical properties of silicon nitride films deposited using silane (SiH$_4$) and ammonia (NH$_3$) as source gases [7–9]. Fewer works has been reported on the deposition of a-SiN$_x$ using SiH$_4$ and N$_2$ as the source gases [10, 11]. The major advantages of using N$_2$ over NH$_3$ are the abundant availability at a cheaper cost than NH$_3$ and nontoxic in nature. So in our present investigations, a-SiN$_x$ thin films are deposited by PECVD using SiH$_4$ and N$_2$ as source gases. Structural properties are investigated using FTIR and Raman spectroscopy analysis and optical properties by UV-VIS spectroscopy analysis.

2. Experimental

Amorphous silicon nitride films have been deposited on corning (Eagle XG) glass using plasma-enhanced chemical vapour deposition technique (Plasmalab System 100, Oxford,
<table>
<thead>
<tr>
<th>Sample number</th>
<th>SiH\textsubscript{4} (sccm)</th>
<th>N\textsubscript{2} (sccm)</th>
<th>Deposition time (min.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiN A</td>
<td>5</td>
<td>2000</td>
<td>12</td>
</tr>
<tr>
<td>SiN B</td>
<td>5</td>
<td>1900</td>
<td>12</td>
</tr>
<tr>
<td>SiN C</td>
<td>5</td>
<td>1800</td>
<td>12</td>
</tr>
<tr>
<td>SiN D</td>
<td>5</td>
<td>1700</td>
<td>12</td>
</tr>
<tr>
<td>SiN E</td>
<td>5</td>
<td>1600</td>
<td>12</td>
</tr>
</tbody>
</table>

Table 2: Modes of vibration of a-SiN\textsubscript{x}:H films.

<table>
<thead>
<tr>
<th>S. no</th>
<th>Si–H\textsubscript{x} stretching</th>
<th>Si–N stretching</th>
<th>N–H wagging</th>
<th>Si–H\textsubscript{x} wagging</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiN A</td>
<td>2046</td>
<td>783</td>
<td>1148</td>
<td>705</td>
</tr>
<tr>
<td>SiN B</td>
<td>2035</td>
<td>785</td>
<td>1151</td>
<td>688</td>
</tr>
<tr>
<td>SiN C</td>
<td>2029</td>
<td>788</td>
<td>1145</td>
<td>670</td>
</tr>
<tr>
<td>SiN D</td>
<td>2031</td>
<td>794</td>
<td>1148</td>
<td>684</td>
</tr>
<tr>
<td>SiN E</td>
<td>2033</td>
<td>800</td>
<td>1160</td>
<td>699</td>
</tr>
</tbody>
</table>

3. Results and Discussions


FTIR analysis is a nondestructive versatile tool in identification of various vibrational modes that occur in the materials and in determining the nature of chemical bonds present in the material. Figure 1 shows the FTIR analysis of various a-SiN\textsubscript{x} films deposited using PECVD at different N\textsubscript{2} concentrations. Generally, in silicon nitride films, three groups of bonds can be observed like Si–N, N–H, and Si–H. In the range from 2000 to 2200 cm\textsuperscript{-1} various vibrational modes like H–Si–Si\textsuperscript{3}, H – Si–H\textsuperscript{2}, H – Si–N\textsuperscript{2}, H – Si–N\textsuperscript{3}, H–Si–H, and H–Si–N\textsuperscript{3} will exist in a-SiN\textsubscript{x} films [12]. Si–N vibrational mode is observed around 780–800 cm\textsuperscript{-1} whereas around 1130–1160 cm\textsuperscript{-1} N–H wagging mode is observed [12]. All the possible modes of vibrations present in the a-SiN\textsubscript{x} films are tabulated in Table 2.

3.2. Raman Spectroscopy Analysis. Raman spectroscopy is a powerful analytical technique used for the analysis of inelastic scattering of light interacting with the material under test. The convoluted Raman spectra of SiN A and SiN D samples are shown in Figures 2(a) and 2(b). The broad peak maxima observed at 482 cm\textsuperscript{-1} in SiN A and 485 cm\textsuperscript{-1} in SiN D is due to the scattering of Si–Si bonds corresponding to the density of states of local TC like phonon modes in amorphous silicon peak [13]. Another peak observed at 400 cm\textsuperscript{-1} in SiN A and 405 cm\textsuperscript{-1} in SiN D corresponding to the scattering on the asymmetric Si–N bond stretching mode in which one of the neighboring Si atoms is removed and one of the central Si atoms is replaced with a nitrogen atom [13].

3.3. Optical Properties. Optical transmission spectra of a-SiN\textsubscript{x}:H films have been recorded in the wavelength range of 300–900 nm. The transmission spectra of SiN A, SiN C, and SiN E are as shown in Figure 3. The refractive index (\(\eta\)) of the deposited films were calculated using Swanepoel’s method [14], which is based on the parabolic fitting procedure of adjacent maximum \(T_M\) and minimum \(T_m\) of the transmission spectra. The refractive index (\(\eta\)) and the absorption coefficient of the film depend on the wavelength, \(\lambda\).

The refractive index (\(\eta\)) of the film has been calculated as a function of wavelength using expression given below which is deduced from the maximum (\(T_M\)) and minimum (\(T_m\)) values
of transmission spectra:

\[ \eta = \left[ N + \left( N^2 - s^2 \right)^{1/2} \right]^{1/2}, \quad (1) \]

where

\[ N = 2s \times \frac{T_M - T_m}{T_M T_m} + \frac{s^2 + 1}{2}, \quad (2) \]

where “s” is the refractive index of the glass substrate, \( S = 1.500 \).

The refractive index values are calculated for SiN A, SiN C, and SiN E using above-mentioned Swanepoel’s method at \( \lambda = 700 \text{ nm} \), and the values are 2.68, 2.78 and 3.01, respectively. These values lie between that of hydrogenated amorphous silicon (\( \eta = 3.75 \)) and stoichiometric SiN\(_4\) (\( \eta = 2.0 \)). Efforts are being made to achieve the value of refractive index close to stoichiometric silicon nitride.

4. Conclusion

Hydrogenated amorphous silicon nitride thin films have been deposited by PECVD technique using SiH\(_4\) and N\(_2\) (instead of NH\(_3\)) as source gases. FTIR analysis has been carried out to identify various vibrational modes present in the deposited films. Raman spectroscopy analysis has been carried out, and peaks around 480 cm\(^{-1}\) and 400 cm\(^{-1}\) are observed, which are mainly due to the Si–Si and Si–N bonds, respectively. Optical transmission spectra are recorded in order to calculate the refractive index of the deposited films. Refractive index values are calculated using Swanepoel’s method and are found to be in between amorphous silicon and stoichiometric silicon nitride.

References


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