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Effect of light-soaking on the hydrogen effusion mechanisms in polymorphous silicon thin film structures



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HIGHLIGHTS

• We study hydrogen effusion due to light-soaking in pm-Si:H PIN and NIP structures.

• Hydrogen diffuses from the bottom layer to the top layer and effuses.

• Hydrogen diffusion process depends on the device structure (PIN or NIP).

• A hydrogen diffusion barrier is generated by defects while trying to effuse on PIN.

• Light-soaking increases the effused hydrogen and is more pronounced on the NIP.

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ABSTRACT

This work describes a study performed on hydrogenated polymorphous silicon PIN and NIP structures, deposited by plasma enhanced chemical vapor deposition (PECVD). We study hydrogen stability and its evolution during exodiffusion experiments in these structures, trying to determine the effect of light-soaking pre-treatments on the hydrogen effusion mechanisms. We examined the structural and optical properties of PIN and NIP structures after exodiffusion experiments. These were analyzed in terms of the different hydrogen diffusion processes comparing light-soaked samples and samples without light-soaking. Also, an analysis of both structures was done in order to determine which one is less affected by light-soaking. From the simultaneous measurement of temperature and hydrogen desorption, we propose a model for long range diffusion of hydrogen rearrangement on the film and microstructure changes which result in a shift on the exodiffusion peaks. Both structures were compared and are discussed in terms of the hydrogen diffusion barrier is generated by light-induced defects, which is expected to cause less degradation of its optoelectronic properties under illumination, and a more stable device during operation along with an enhanced functional performance of this type of structure.

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1. Introduction

Hydrogenated polymorphous silicon (pm-Si:H), which consists of silicon nanocrystals embedded in an amorphous silicon (a-Si:H) matrix, is a material suitable for solar cell devices. The latter is because it exhibits better electronic transport and stability properties compared to those of the conventional a-Si:H [1–3]. Most of

http://dx.doi.org/10.1016/j.matchemphys.2015.07.045 0254-0584/© 2015 Elsevier B.V. All rights reserved. the improved properties related to these materials are a consequence of the hydrogen ability to passivate defects and impurities in the Si network, removing or reducing the intra-bandgap levels [4]. Reversible Staebler–Wronski effect is one of the most significant phenomena that affects the stability and performance of amorphous silicon thin films solar cells [5]. This light induced degradation causes changes in structural and electronic properties of the material [5–9]. Bonded hydrogen atoms may break during light exposure and diffuse through the amorphous network generating an increase of the dangling bonds. On the other hand, diffusing hydrogen atoms may be trapped by the dangling bonds generated previously or may exodiffuse leading to a film richer in

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defects [10]. The trapping probability of free hydrogen atoms increases with temperature due to a higher dangling bonds formation and it is more pronounced closer to the film surface, which shortens the diffusion length [10]. Hydrogen diffusion is considered a key process in this kind of materials [6–8,10,11]. In general, the proposed models in literature for explaining the Stabler–Wronski effect are: weak bond breaking without hydrogen involvement, Si-Si breaking with involvement of hydrogen, charge transfer between charged states and bridge bonded hydrogen in interstitial site [12]. Hydrogen is also known to passivate Si-SiO₂ interfaces and grain boundaries [4]. As a consequence H passivation has become a crucial point of study in microelectronics and photovoltaic materials. A good passivation of the solar cell structure is necessary to improve the cell performance. An important technique to study the hydrogen diffusion is exodiffusion measurements where hydrogen diffusion occurs due to an external energy excitation. Hydrogen moves through all the film generating several reactions during its motion and when it reaches the films surface it exodiffuses. The hydrogen exodiffusion depends on the film structure, hydrogen bonding environment and hydrogen concentration. Recently, studies determined that pm-Si:H thin films and devices present better stability under illumination than conventional a-Si:H [13]. However, an excessive incorporation of weak Si-H bonds into the pm-Si:H film could affect the degradation rate of the device for photovoltaic applications and, moreover, the device conversion efficiency [3,13]. During annealing, exodiffusion of hydrogen takes place and it activates the defects and impurities as recombination centers [4]. At moderate temperatures (100–300 °C), a slow diffusion was observed while at higher temperatures (400-800 °C) fast hydrogen interstitial diffusion is present [14,15]. During diffusion, hydrogen interacts with defects, dangling bonds and electrically active dopant atoms [7,14,18], which in turn could improve the device performance via defects passivation. Hydrogen diffusion is more complex than the diffusion of electrically active dopants [15] but the activation energy is lower. Diffusion in this material is thought to be similar to that of a-Si:H, mediated by the dangling bonds and thermally activated [16–18]. In the low concentration region H diffusion has an activation energy of 2.7 eV and is independent of the H concentration [16], whereas for the higher hydrogen concentration region the diffusion process becomes more complex, decreasing the diffusivity [16,19]. Hydrogen diffusion activation energies for a-Si:H range from 0.5 to 2.7 eV [8], so the incident photon energy of a halogen lamp would be sufficient to enhance (generate) atomic or molecular hydrogen diffusion by breaking atomic bonds and also creating electron-hole pairs [8]. It has been determined that for a-Si:H with diffusion activation energies of 1.8 eV, diffusion coefficients of 9.5×10^{-2} and 9.6×10^{-1} cm²/s are observed, while for energies of 2.7 eV the values are between 2.2 \times 10⁴ and 5.4 \times 10⁴ cm²/s [16,20,21]. The activation energy of free-H diffusion was estimated to be less than 0.5 eV. Therefore, several models related to hydrogen motion have been proposed to explain the light degradation effect in a-Si:H [7,10]. It is considered that hydrogen moves through bond switching between a Si-H bond and a Si dangling bond and/or H exchange between Si–H bonds [10]. Since the H diffusion process depends mostly on the dangling bond density [17], this suggests that hydrogen movement involves the process of bond switching between a Si–H bond and a dangling bond.

During hydrogen diffusion pm-Si:H can be crystallized due to subsurface reactions of bond breaking taking place as part of the exodiffusion process [22–24]. For annealing temperatures greater than 300–400 °C hydrogen in a-Si:H becomes unstable and exodiffuses, which is expected on pm-Si:H as well [1,11]. Thus, it is crucial to understand the hydrogen diffusion process in pm-Si:H and its effects on the structural and optoelectronic properties of this material. In previous works, hydrogen profiles on silicon have shown that p-type and n-type materials have different hydrogen concentration regions. For p-type high, intermediate and low concentration regions are present while for n-type a high concentration near the surface and a low concentration region are present [15.25]. In a PIN type substrate solar cell structure the incident light enters through the p-type layer. Here the absorption of the glasstransmitted UV part of the solar spectrum is expected along with a larger chance of generating dangling bond defects which reduce the cell performance as well [5]. Trying to determine the effect of previous light-soaking on the hydrogen effusion mechanisms in pm-Si:H thin film structures, in this work we examined the optical properties of PIN and NIP structures after exodiffusion experiments. Their structural and optical properties were analyzed in terms of the different hydrogen diffusion processes for samples exposed previously to light-soaking and samples without light-soaking. A comparison of both structures was done in order to determine which one is less affected by light-soaking. Also, we propose a model for long range diffusion of hydrogen in both structures which explains the effects of the light-soaking treatment and the differences between both structures.

2. Material and methods

Pm-Si:H thin films structures were grown in a conventional PECVD system with parallel plates, activated by a RF signal of 13.56 MHz [13,26,27]. The depositions were done with a chamber pressure of 3.3 Torr, substrate temperature of 275 °C and a RF power of 30 W. Hydrogen (H₂) and silane (SiH₄) were used as precursor gases with mass flow rates of 200 and 40 sccm, respectively. Doped layers were grown using conditions similar to previous works [22]. Doping is obtained by adding 5 sccm of a doping gas (trimethylboron for p-doping and phosphine for n-doping) diluted at 2% in hydrogen. To avoid any oxidation effect, the films were grown in a single process without breaking vacuum. The p and n type layers were around 20–30 nm of thickness, while the intrinsic layer was ~200 nm thick. The parameters for both structures were maintained constant and only the order of deposition of the layers was varied. Thus, the structures were deposited on corning glass #7059 in both substrate and superstrate configurations. Analyzed samples description is summarized in Table 1. The light-soaking treatment was performed during 1 h in a homemade equipment under AM1.5 spectrum and the temperature was controlled at 25 °C by heat dissipation and exposition was through the glass side. Thermal annealing during exodiffusion experiments was done with an incremental ramp of 10 °C/min starting at room temperature and up to 650 °C for hydrogen evolution studies in all the pm-Si:H structures. These experiments were performed in a vacuum chamber with a controlled heating system which increases the temperature and the effusing hydrogen was detected with a mass spectrometer. Table 1 shows the NIP and PIN structures studied in this work and their thicknesses. The different states described in Table 1 are as follows: the "exodiffusion" state refers to a structure after the exodiffusion experiment, and the "exodiffusion after lightsoaking" state refers to a structure that was exposed to lightsoaking and immediately after an exodiffusion experiment was

Table 1	
Description and thickness of the PIN and NIP structures used in this wor	rk.

Sample	Name	Total thickness (nm)	Description
D04086 D04086-LS D04087 D04087-LS	PIN PIN-LS NIP NIP-LS	243.7 248.3	Exodiffusion Exodiffusion after light-soaking Exodiffusion Exodiffusion after light-soaking

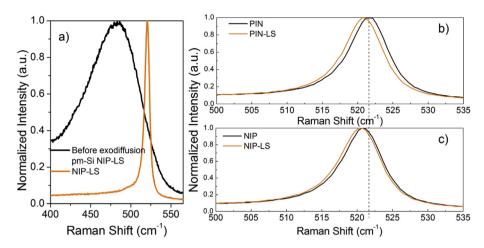


Fig. 1. a) Representative Raman spectra of the NIP-LS structure before (black line) and after (orange line) the exodiffusion experiment. Raman spectra of b) PIN and c) NIP structures after exodiffusion showing a shift of the principal peak with light-soaking. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

conducted. Optical measurements were conducted through the film side for all samples. Spectroscopic Ellipsometry (SE) measurements were performed using a HORIBA Jobin-Yvon UVISEL in order to determine the films thickness, surface roughness and optical properties like the absorption coefficient (α) and the optical gap (Eg). A Tauc-Lorentz model was selected to obtain the optical and morphological model parameters of the films. The Tauc-Lorentz model parameters C and A are related to the film disorder and compactness, respectively [28]. Raman spectroscopy was used to investigate the nanostructure of the film via a Jobin-Yvon Horiba triple monochromator T6400 micro-Raman spectrometer in backscattering configuration equipped with a He-Ne laser (632 nm) excitation. All the measurements were performed at room temperature in open air. The samples were irradiated at a power of 2 mW. The spectrometer is equipped with a CCD detector and a confocal microscope with a $10\times$ objective. The spot was unfocused resulting in a spot size of ~0.6 mm in diameter and a photon flux of ~2 kW/m². The measuring range and selected integration time were 400–600 cm⁻¹ and 1 min, respectively. The Raman spectra analysis of the different phonon modes between 400 and 550 cm⁻¹ is known to be related to the amorphous phase (480 cm^{-1}) , an intermediate phase $(500-517 \text{ cm}^{-1})$ related to small size nanocrystals and grain boundaries, and a crystalline phase (520 cm^{-1}) [29–31].

3. Results and discussion

Fig. 1a shows representative Raman spectra of one structure before and after the exodiffusion experiment. This evidences the crystallization of the pm-Si:H structures resulting from the annealing performed for the exodiffusion experiments. A change of the crystalline fraction in pm-Si:H structures from around 7% to values near 80% is observed. Fig. 1b and c shows the Raman spectra for all the pm-Si:H structures after exodiffusion. The spectra show a principal peak centered around 520 cm⁻¹ associated to the silicon transverse optical mode, which is attributed to the crystalline phase. As observed in Fig. 1b and c, the PIN structure presents more shift of the peak compared to the others as well as a more pronounced effect due to the light-soaking pre-treatment. For both structures a peak shift toward lower wavenumbers caused by lightsoaking is observed. This shift could indicate an increase of the interfaces with a higher contribution of the 500 cm⁻¹ band. Thus, a first proposal is that there is a difference on the way that lightsoaking affects or modifies the structure in dependence of the architectural arrangement.

To understand the micro-structural changes on PIN and NIP structures due to light-soaking SE measurements were done. Fig. 2 shows a comparison of the real (\mathcal{E}_1) and imaginary (\mathcal{E}_2) parts of the pseudo-dielectric function for the PIN and NIP and the effect of light-soaking. First, it can be appreciated that for the PIN structures the imaginary part of the dielectric function presents a double peak in 3.3 and 4.2 eV that could be associated to a crystalline phase present in the films that is not observed on the NIP structures [32]. The NIP structures SE spectra is similar to an amorphous material with some small crystallization, despite a major crystallization observed on the Raman results but maybe related to an increase of interfaces, defects, and additionally a surface damage. The crystallization presented in the PIN structures there is not an evident change with light-soaking.

To determine the changes of the optoelectronic properties a Tauc-Lorentz model was selected as the spectroscopic ellipsometry model to describe the structures. Some of the parameters that resulted from the model are presented in Table 2. The optical gap obtained is higher for the NIP structure (1.7 eV) than for the PIN structure (1.5 eV). Light-soaking has almost no effect on the optical

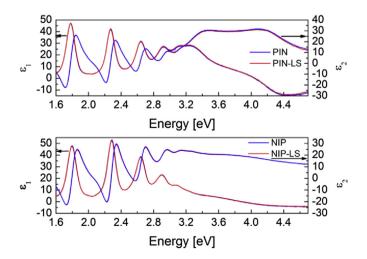


Fig. 2. Comparison of the PIN and NIP ellipsometry spectra after exodiffusion.

 Table 2

 Spectroscopic ellipsometry parameters using a Tauc-Lorentz model to describe the structures.

	PIN	PIN LS	NIP	NIP LS
Eg	1.54 ± 0.02	1.56 ± 0.02	1.74 ± 0.03	1.73 ± 0.02
Α	97.29 ± 2.99	100.28 ± 2.65	96.49 ± 3.91	99.25 ± 3.25
С	0.58 ± 0.02	0.61 ± 0.02	0.40 ± 0.02	0.47 ± 0.02

gap, as shown for both structures. However, the Tauc-Lorentz model parameter C shows an increase for both structures indicating a less ordered structure due to light-soaking. The latter can be related to a surface damage associated with dangling bonds caused by bond breaking during illumination. Moreover, the parameter A related to the film compactness increases due to lightsoaking for both structures indicating a more dense film. This is an indication that light-soaking generates atomic hydrogen diffusion and a rearrangement of the film, diminishing hydrogen in weak bond positions, even before exodiffusion experiments. The ellipsometry measurements were done after the exodiffusion experiments generating a change on the optical band gap of the structures from the as grown samples. There are several reasons why the PIN structure presents a lower optical band gap: the films crystalline fraction, the doping concentration and type of the top layer, the top film disorder, and the hydrogen content of the top film. Raman and SE have shown a higher crystallization degree of the PIN structure after exodiffusion shifting the optical gap from the conventional values of a-Si:H to values closer to µc-Si. On the other hand, it is shown on Table 2 that the PIN presents a higher degree of disorder, associated to the variation of the crystallographic directions. The latter, in turn affects the optical gap of the material decreasing its value.

The refractive index and extinction coefficient obtained from the Tauc-Lorentz model describing the PIN and NIP structures are plotted in Fig. 3a and b, respectively. A small decrease of n and k with light-soaking is observed for the NIP structure in Fig. 3b, indicated by arrows. This could be associated to a change in the film microstructure and hydrogen bonding as a consequence of light-soaking. Hydrogen diffusion during light-soaking generates a decrease on the absorption properties of the films (mostly on the NIP). Comparing both structures an important difference of n and k could be observed as a consequence of the layer order in the structure. For energies above 3.5 eV the decay of n and k is more

pronounced for the NIP structure. The latter is related to the top layer material (n or p) of the structure as a consequence of the response for these incident photon energies. As shown on Fig. 3, the decrease of k for NIP with respect to PIN means that the incident photons on the NIP structure are less susceptible to be absorbed; as a consequence the absorption coefficient should decrease and a larger optical gap is observed (Table 2). Compared to a p-type layer, the hydrogen concentration is higher, moreover, our considerations for the SE model may average the optical properties on the whole structure, therefore the optical gap of the NIP should be greater than that of the PIN structure. For both structures, light-soaking has almost no effect on the optical gap as observed on Table 2. This result should be taken into account due to its importance in the optoelectronic properties for solar cell devices.

Exodiffusion results are shown in Fig. 4. As the temperature is increased we can observe the total hydrogen effusion (molecular and atomic) from the different structures. This experiment allows the analysis of hydrogen stability and its evolution during annealing. The total hydrogen that is effusing from the PIN and NIP structures is very different. A smaller quantity of hydrogen is effusing from the PIN structure compared to the NIP effusion spectra. From these measurements we propose a model for long range diffusion of hydrogen through the whole device. It is proposed that hydrogen diffuses from the bottom layer to the top layer and effuses. This suggests that hydrogen on the PIN and NIP structures diffuses from the bottom-up and it accumulates on the surface film in order to exodiffuse. Several diffusion barriers may be present during the process (interfaces, grains, surface, damaged surface). Therefore, exodiffused hydrogen should not be exactly the same because there is an environment dependent interaction of hydrogen with the different layers during diffusion. Thus, the hydrogen diffusion process depends on the device structure and its effusion is observed to be greater on NIP structures (Fig. 4). This indicates that the n layer on the PIN structure may work as a barrier for diffusing atomic hydrogen. The p layer presents higher diffusivity with respect to the n layer that generates a very small resistance to the hydrogen diffusion and apparently all hydrogen that enters to this layer has a good chance to effuse from the sample [23]. The thermal energy supplied during the exodiffusion experiment is not enough for the total hydrogen to come out from the sample, remaining tightly bonded hydrogen instead. In particular for the NIP structure, this thermal energy would not be enough to overcome the diffusion barrier proposed in our model and that

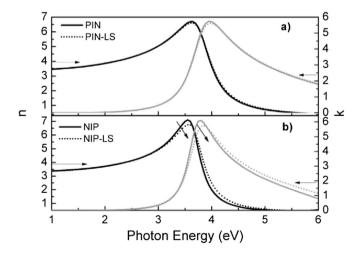


Fig. 3. Refractive index and extinction coefficient for the Tauc-Lorentz model describing (a) PIN and (b) NIP structures.

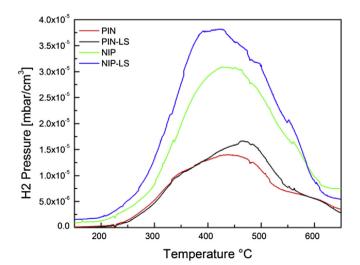


Fig. 4. Exodiffusion spectra for PIN and NIP structures as grown and light-soaked.

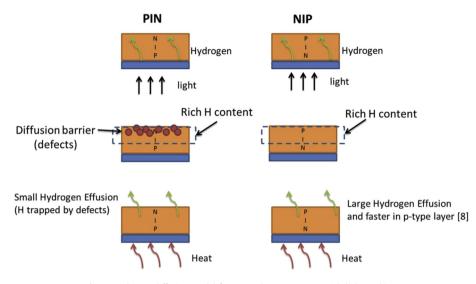


Fig. 5. Hydrogen diffusion model for PIN and NIP structures with light-soaking.

would explain the difference on Fig. 4. Second, it can be appreciated that light-soaking generates an increase of the amount of hydrogen that is effusing from both structures during annealing. The light-soaking effect is more pronounced on the NIP structure than in the PIN where the increase is just observed for temperatures between 400 °C and 570 °C. In contrast, for the NIP structure the effect is observed almost for the whole spectrum. All these effects can be explained with the graphical model shown in Fig. 5.

Thus, hydrogen in the PIN structure has more difficulty to effuse during light-soaking and remains within the structure, resulting in a delay on the degradation process, while for the NIP structure it is totally the opposite and the degradation should be faster. It is well known that hydrogen diffuses faster in p-type hydrogenated amorphous silicon [23]. Apparently, the n layer of the PIN structure works as a barrier for the diffusing hydrogen during light-soaking (Fig. 5). As a consequence of this, the trapping probability of hydrogen diffusing to the surface of the n-type film is increased. Also, the higher hydrogen concentration region, which in this case is the near surface of the n-type layer, is well known to have less hydrogen diffusivity decreasing the hydrogen exodiffusion [16,19]. For both structures, light-soaking has almost no effect on the optical gap as observed on Table 2.

In order to have a precise explanation of all this, previous studies of the i-layer material alone have been done by our group and other groups [33]. Hydrogen diffusion changes the hydrogen bonding environment, hydrogen stability and, furthermore, the film structure as is the case for the a-Si:H [29]. First we have to notice that hydrogen within the structure diffuses to the top layer during annealing where it may be trapped by defects (dangling bonds) or exodiffuse. During light-soaking the hydrogen starts to diffuse to the defects of the top layer or even exodiffuse. Hydrogen coming from the breaking of Si-H bonds is trapped into voids during lightsoaking and it can be released in the lattice during annealing [35]. It has been observed that during exodiffusion experiments a much larger amount of hydrogen is released from the NIP structure. Previous works have proposed that the n layer of the PIN is more damaged during light-soaking generating more dangling bonds, which are in turn, high absorption sites for the diffusing hydrogen during annealing [34]. Further experiments are necessary to analyze the n layer hydrogen concentration in order to justify the proposed model. In addition, F. Kail et al. showed that hydrogen diffuses faster in boron-doped (p-type) hydrogenated amorphous silicon [23]. The large amount of hydrogen diffusing trough the structure could induce crystallization during its motion explaining the structural difference between the PIN and NIP structures [29]. From the evolution of Raman before and after light-soaking, we show that this long range motion of hydrogen causes a rearrangement and affects the crystallization on the film, maybe related to an increase of the interfaces and to the dangling bonds density which also explains the difference of the optical gap between structures. As a result of the light-soaking, a shift and an increase on the exodiffusion peaks is observed, greater in NIP, which explains the degradation process in pm-Si:H thin films solar cells. Previous works have demonstrated that PIN solar cells of pm-Si:H compared to the a-Si:H have better light induced stability on the device efficiency [13,27]. On the other hand, pm-Si:H PIN substrate and superstrate configurations have been studied in order to determine the solar cells efficiency change due to light exposition; indicating that substrate configuration have lower light induced degradation [36]. All of these are in good accordance with the results shown in this work.

4. Conclusions

Hydrogen in hydrogenated polymorphous silicon thin film structure diffuses from the bottom layer to the top layer and effuses. We have demonstrated that the hydrogen diffusion process depends on the device structure PIN or NIP. Weakly bonded hydrogen migrates to tight bonding positions with light-soaking pre-treatment. For the PIN structure a hydrogen diffusion barrier is generated by defects passivated by the hydrogen that is trying to effuse during annealing. The n layer of the PIN structure is more damaged during light-soaking generating a greater amount of dangling bonds, which constitute absorption sites for the diffusing hydrogen during annealing. Light-soaking generates an increase of the amount of hydrogen that effuses from both structures and it is more pronounced on the NIP structure.

Conflicts of interest

The authors declare no conflict of interest.

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