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New Numerical Simulation of the Optical Behavior of Nanocrystalline Silicon Thin Films

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Abstract

The absorption enhancement observed on the CPM spectra of nanotextured silicon thin films should be attributed to light scattering effects. A detailed numerical simulation based on Monte Carlo method is presented to calculate the absorption spectra of hydrogenated nanocrystalline silicon films according to the CPM setup. The calculated spectra of apparent optical absorption coefficient depend mainly on the characteristic parameters of nano-Si:H thin film, denoted crystalline/amorphous fraction which favors bulk light scattering phenomena and the film thickness which leads to the light scattering at the rough surface.
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1. Introduction

Hydrogenated nanocrystalline silicon (nano-Si:H) is an heterogeneous material. It consists of an ordered nanocrystallites of spherical form with size varies from 2 to 20 nm embedded in amorphous matrix [1]. These nanomaterials deposed by plasma enhanced chemical vapor deposition (PECVD) technique at low temperature (<300°C) [2] constitute an important class with some of their properties distinctly different from either amorphous or large grain materials or single crystals. Moreover, they present a low metastability during their prolonged exposure to the light, compared with hydrogenated amorphous silicon (a-Si:H) films [3], which is very promising to the photovoltaic applications [4]. The important key for the success of the nano-Si:H layers as a PV absorbent material is its enhanced absorption compared to the monocrystalline silicon (c-Si), mainly in weak energies range. The main reason of this optical behavior is due to its particular structure which gives place to the bulk and/or surface light scattering phenomena [5,6,7]. Indeed, due the waveguiding properties of silicon layer, the photon trapped by the included nanocrystallites undergoes a several scattering events until its light intensity will attenuate, and consequently, it will be absorbed completely within its traveled optical path. The constant photocurrent method

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(CPM) measurements of these heterogeneous mediums give access to an "apparent" optical absorption coefficient (α_{app}) affected by scattering effects different from that the true one measured for the homogeneous mediums, i.e., amorphous or monocrystalline silicon [5]. The CPM is well known from the field of amorphous silicon [8,9]. It detects the light absorbed (either directly or after one or more scattering events) in between two metal electrodes used for the photocurrent measurements. In this study, we will try to calculate $\alpha_{app}(E)$ spectra and the total film absorptance influenced by the characteristic parameters of a nanotextured silicon film, especially the crystalline/ amorphous fraction and the film thickness by introducing a numerical simulation based on Monte Carlo method. More precisely, we will discuss the contribution of bulk light scattering which depended mainly on the crystalline /amorphous fraction in the evaluated apparent absorption spectra and in the calculated optical paths of scattered photons. Moreover, we will simulate the influence of light scattering at the rough surface on the apparent absorption curves calculated from CPM data. The surface roughness is related mainly to the variation of the film thickness. For that, we must take into account in our simulation the diffusive aspect of the characterized material.

We must note that the influence of these parameters on the absorption spectra of nano-Si:H thin films is treated theoretically by Poruba [5]. They introduced the scalar scattering theory for a random rough surfaces in which they calculated numerically $\alpha_{app}(E)$, σ_{rms} and d_f by successive iteration. Finally, they compared their results with experiment effectuated in there laboratory.

However, we tried to study these parameters by another method based on _random walk_ theory where we have introduced the random number to calculate the optical path and the differents other parameters. We have used only the Beer's law of optical absorption coefficient to realize this simulation.

2. Simulation method

In the following, let us consider a monochromatic light beam incident perpendicularly on a thin nano-Si:H film with a typical thickness d_f deposited in between two coplanar electrodes with width W and distance D [8]. Furthermore, we assume that the nano-Si:H film is composed of a similar square lattices set equidistant of a distance L. Each plan contains nanocrystallites of identical size which constitute the scatterer sites in the sample as is shown in Fig. 1.

However, the homogeneous matter in the film represented by no-scatterer sites. We shall note that the distribution of the nanocrystallites on each plan is purely random but their number remains always fixed. Second, we suppose that the incident photon falls randomly on the site of the first plan. If this site is no-scatterer, the photon penetrates in the sample and passes in the second plan on the site located just below the first. If the second site is also no-scatterer, the photon passes in the third plan and so on, until it crosses completely the film thickness. Consequently, the total film absorptance is equivalent to the true one given by the Beer's law [10]:

$$A_{true} = (1 - R)(1 - exp(-\alpha_{true} . d_f)) \qquad (1)$$

Where α_{true} is the true optical absorption coefficient in the film and R is the reflectance.

If we use this simple approach (Beer's law) for bulk and/or surface light scattering in the films, i.e., neglecting the changes in the spectral dependence of reflection $R(E)$ comparing with changes in α_{true} over several orders of magnitude, the attenuation of the specular beam will be given by:

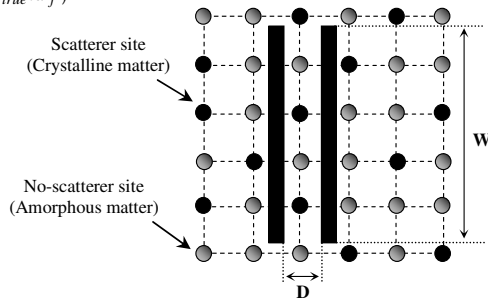$$A_{true} \approx 1 - exp(-\alpha_{true} . d_f) \qquad (2)$$

Fig. 1. Square lattice of a nanocrystalline silicon layer configured according to the CPM setup.
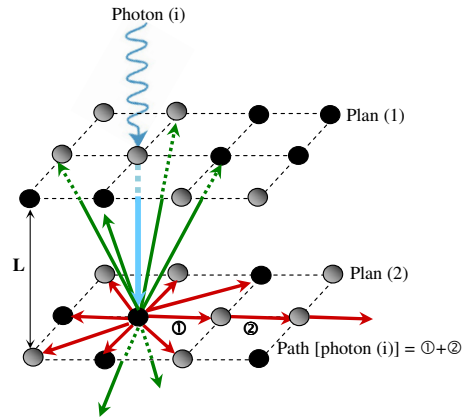
Fig. 2. Scheme of the scattering direction probabilities which the scattered photon (i) can travel them. We present here some directions in order to not condense the scheme. The red allows indicate light scattering on the plan and the green one for the light scattering in space between two plans. A simple example of calculation of the optical path probably traveled by the photon (i) is mentioned on the scheme.

However, if during its crossing the sample, the photon from the specular beam falls onto a *scatterer site* belongs to one of the sample plans, it will scatter randomly on the plan and in the space in all the directions, i.e., isotropic scattering (Fig. 2), until its intensity will be attenuated (lower than 10^{-6} of its initial intensity) and it will be absorbed. If it is not absorbed, it will be scattered again and the same considerations are valid.

Therefore, the film absorptance for the scattered photon will be:

$$A_{sc} \approx 1 - exp\left(-\alpha_{true}.Path\left(i\right)\right) \qquad (3)$$

where *Path* (*i*) represents the mean free path traveled by the scattered photon (*i*).

However, the total film absorptance of scattering nano-Si:H samples is defined as:

$$A_{tot} \approx 1 - exp\left(-\alpha_{app}.d_f\right) \qquad (4)$$

where α_{app} is the apparent optical absorption coefficient in the film measured by CPM.

On the other hand, CPM uses the photocurrent (I_{ph}) proportional to the amount of light absorbed in the film. Thus, taking into account the contribution of the scattered light to the CPM signal, I_{ph} will be composed of two components:

$$I_{ph} = I_{true} + I_{sc} \qquad (5)$$

I_{true} is proportional to A_{true} represents the photocurrent excited by the photons absorbed directly from the specular beam, i.e., without any scattering.

$$I_{true} \approx N_1 A_{true} \approx N_1\left(1 - exp(-\alpha_{true}.d_f)\right) \qquad (6)$$

N_1 is the number of no-scattered photons.

The component I_{sc} represents the contribution of the photons absorbed after one or more scattering events. It's given by:

$$I_{sc} \approx \sum_{i=1}^{N_2} \left[1 - exp\left(-\alpha_{true}.Path(i)\right)\right] \qquad (7)$$

Here N_2 represents the number of scattered photons.

From the formulas (5), (6) and (7), we can write:

$$N\left[1 - exp\left(-\alpha_{app}.d_f\right)\right] \approx N_1\left[1 - exp\left(-\alpha_{true}.d_f\right)\right] + \sum_{i=1}^{N_2}\left[1 - exp\left(-\alpha_{true}.Path(i)\right)\right] \qquad (8)$$

Consequently, α_{app} values can be calculated easily from the formula (8).

Nevertheless, film transmittance is expressed as:

$$T_f \approx -\frac{1}{N} exp(-\alpha_{true} \cdot d_f)$$ (9)

Since our simulation is based on Monte Carlo method, we must assure the reproducibility of the numerical results. For that, we propose the following steps.

2.1. Choice of the sample

The sample of silicon that will simulate its optical apparent absorption has considered at the first time amorphous, i.e., it does not contain nanocrystallites in the bulk or at the surface. The film is deposited in between two electrodes with width W=2.2 mm and distance D=0.03 mm [5]. $\alpha_{true}(E)$ spectrum of the layer has deduced from CPM measurements [5]. After that, we simulate the $\alpha_{app}(E)$ spectra for different proposed values of crystalline/amorphous fraction and film thickness and for several electrodes geometries. It's however important to note that we have simulated the influence of the geometrical setup of the electrodes on the evaluated $\alpha_{app}(E)$ spectra of nano-Si:H thin films by using the same sampling parameters given in this paper.

2.2. Choice of the sampling parameters

The reproducibility method consists to determine the sampling parameters values (incident photons, contained sites, lattices or subdivisions) for which the calculated results keep constant values according to their variation. After a several numerical iterations, we have concluded that the numerical results (transmittance, film absorptance, $\alpha_{app}(E)$, mean optical path,…) remain constant and become reproducible for a number of sites higher than 1000 sites in each lattice and for 1000 incident photons as well as for a distance inter-plan L = 0.2 µm which corresponds to a 10 lattices.

Fig. 3 gives the reproducibility of the total film absorptance according to the variation of the number of sites given in one plan. One can see that A_{tot} values will stabilize from a number of sites higher than 1800. Fig. 4 shows that the calculated transmittance values keep a constant percentage for a number of incident photons higher than 1000.
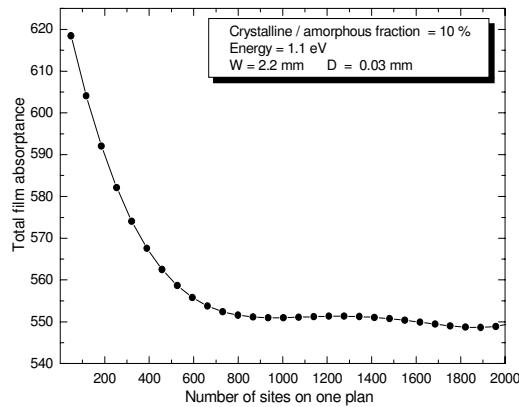

Fig. 3. Total film absorptance calculated according to the variation of the number of sites in one plan of nano-Si:H layer in weak energies range.
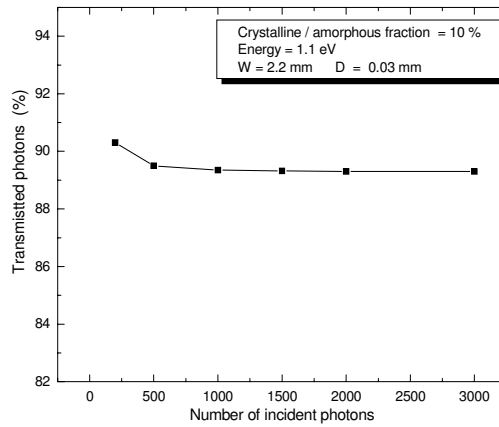
Fig. 4. Reproducibility o f the transmittance according to the variation of the number of photons incident on nano-Si:H layer in weak energies range.

The same observations were obtained in Fig. 5 where the gap between α_{app} spectra become more narrow (in 0.8-1.4 eV spectral region) for a distance inter-plan L equals to 0.2 µm.
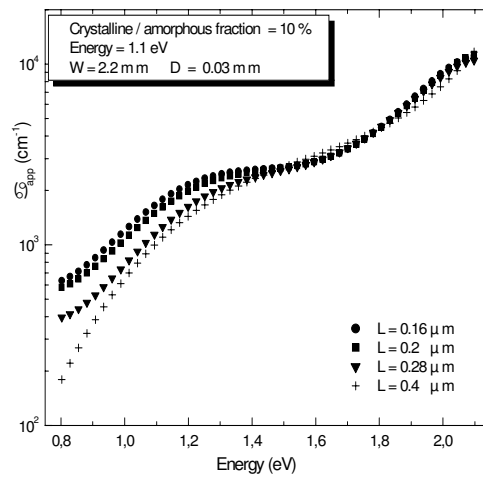

Fig. 5. Reproducibility of $\alpha_{app}(E)$ spectra calculated according to the variation of the distance inter-plan L of a sample of nano-Si:H layer in weak energies range**.**

3. Results and comparison

3.1. Influence of crystalline/amorphous fraction

Fig. 6 shows a significant improvement of the total film absorptance values with the increase of crystalline/amorphous fraction percentage, especially in infrared region. This enhancement is due to the correlation of the mean free path traveled by the scattered photon with the nanocrystallites fraction as is shown in Fig. 7.
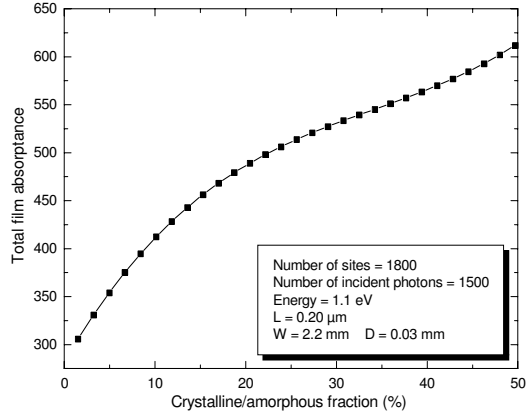
Fig. 6. Influence of the crystalline/amorphous fraction on the calculated total film absorptance curve.
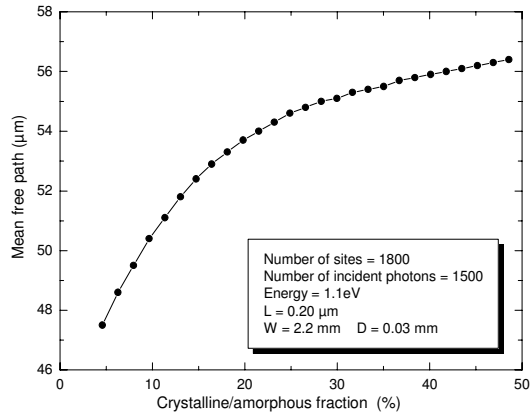
Fig. 7. Influence of the crystalline/amorphous fraction on the calculated mean free path of a scattered photon.

The calculated apparent absorption coefficient spectra $\alpha_{app}(E)$ for different crystalline/amorphous fractions for thin nano-Si:H are shown in Fig. 8. The evaluated spectral dependence of the true absorption $\alpha_{true}(E)$ is shown as the basic experimental result, i.e., deduced for crystalline/amorphous fraction equals practically 0 %.

Results presented in Fig. 8 show that $\alpha_{app}(E)$ values increase with the nanocrystallites number included in the bulk and/or at the film surface, especially in the 0.8-1.2 eV spectral region. This is completely in agreement with the experiment [11,12,13].
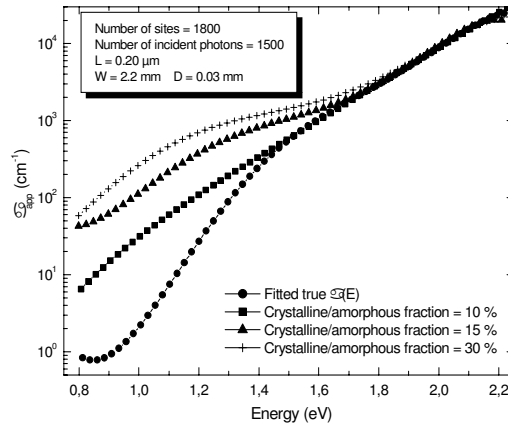

Fig. 8. Calculated $\alpha_{app}(E)$ spectra of nanocrystalline silicon layer with $d_f = 2\mu m$ for a several amorphous/crystalline fraction. The fitted spectral dependence of $\alpha_{true}(E)$ deduced from CPM measurements [5] is shown for comparison.

3.2. Influence of film thickness

The dependence of the calculated $\alpha_{app}(E)$ spectra on the film thickness is clearly shown in Fig. 9, mainly in the 0.8-1.1 eV spectral region.
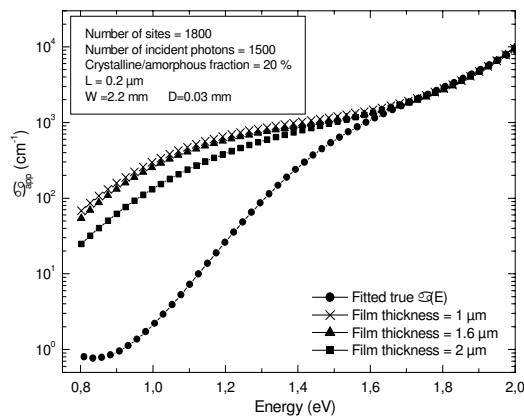

Fig. 9. Calculated spectral dependence of $\alpha_{app}(E)$ of a nanocrystalline silicon layer as a function of the film thickness. The fitted spectral dependence of $\alpha_{true}(E)$ deduced from CPM measurements [5] is shown for comparison.
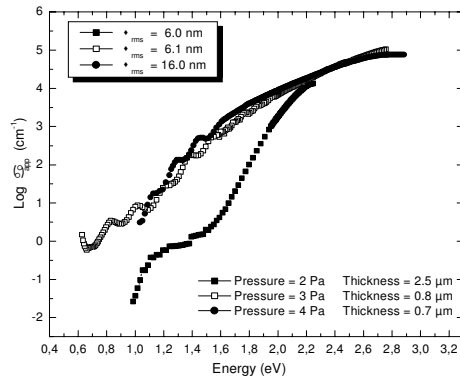
Fig. 10. Apparent optical absorption spectra of nanotextured samples deposited by RF magnetron sputtering method at 150 °C at different pressure deduced from CPM measurements.

The analysis of this figure shows that α_{app} values are inversely proportional to d_f. This result is in agreement with that found in experiments [5,14,15] and in Fig. 10. The samples were deposited by RF magnetron sputtering of a silicon target, under different pressure (2, 3 and 4 Pa) with substrate temperature 150°C. This process enables to deposit the silicon thin films on all types of substrates. The layers results in a nanotextured (rough). The measured typical root mean square surface roughness (σ_{rms}) varies between 6 and 16 nm. We note that the CPM spectra of samples deposited at 3 Pa and 4 Pa are completely different from that of sample deposited at 2 Pa. Moreover, the α_{app} (E) spectrum measured for 2 Pa presents an aspect like that of standard amorphous silicon. However, the aspects of 3 Pa and 4 Pa spectra demonstrate the nanocrystalline character of our samples with a high optical absorption values in all the spectral range. The nanocrystalline signature of the CPM spectra is due to the big nanocrystallites with size close to the film thickness (these results are confirmed as well with Raman and Ellipsometry Measurements).

4. Discussions

In this paragraph, we will discuss and interpret the different results founded previously.

4.1. Crystalline/amorphous fraction

The important presence of the nanocrystallites with small size in silicon layer, increases the probability that the incident photon from the specular beam will be confined within the bulk of the film by total internal reflections and will remain trapped until it is either absorbed or scattered again. However, the optical path of the scattered photon increases also with the increase of the internal reflections events, i.e., the number of included nanocrystallites (Fig. 7). Note that, in weak energies range (below 1.1 eV), the optical absorption is rather low and it has to be increased because of the longer optical path in which the scattered photon will be absorbed.

4.2. Film thickness

Results in Figs. 9 and 10 for nanotextured-Si:H samples demonstrate the influence of the film thickness on the calculated α_{app} (E) spectra. As a special study, we discuss here the surface light scattering effects by varying this parameter. The presence of big nanocrystallites (aggregate composed of small nanocrystallites) with size comparable with the film thickness leads to an important roughness of the film surface. This dependence was confirmed in Figure 10, where we can see that σ_{rms} values increase with the decrease of d_f. However, we shall note that the decrease of film thickness neglect the presence of small nanocrystallites in the bulk and consequently, to a negligible bulk light scattering. Therefore, surface light scattering will be the dominant phenomenon. Hence, the enhanced optical absorption is mainly due to the light scattering only at the rough surface. However, the photons

scattered from the specular beam by the rough surface will be absorbed within the area between the electrodes. And thus, it will contribute to the CPM signal. On the other hand, in Fig. 10, the $\alpha_{app}(E)$ spectra subsist also towards the weak energies with optical absorption attributed to broken bonds due to the amorphous fraction in the films. This would point to the hypothesis that the material contains a high crystalline fraction with an amorphous silicon tissue located in the grain boundaries.

5. Conclusion

We have reported in this paper a Monte Carlo simulation in order to study the influence of different parameters of a heterogeneous material (especially nanocrystalline silicon thin films) on the evolution of its optical absorption mainly in the infrared spectral region. The results issues from this simulation point to the conclusion that the absorbed scattered light which will contribute to the CPM absorption enhancement is affected by the number of the included nanocrystallites and by the rough surface. It's also depended on the geometry of interelectrodes region of the CPM setup. Although the studied phenomena are purely random and complex, we tried to simulate the apparent optical absorption spectrum of a nanotextured-Si:H layer in order to obtain the optimal parameters of an efficient absorbent thin-film used mainly in photovoltaic applications.

Acknowledgements

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