

Design, fabrication and optical characterization of photonic crystal assisted thin film monocrystalline-silicon solar cells

Xianqin Meng,^{1,2,3} Valérie Depauw,⁴ Guillaume Gomard,^{1,2,3} Ounsi El Daif,⁴ Christos Trompoukis,⁴ Emmanuel Drouard,^{1,2} Cécile Jamois,^{1,2,3} Alain Fave,^{1,3} Frédéric Dross,⁴ Ivan Gordon,⁴ and Christian Seassal^{1,2,*}

¹Université de Lyon, Institut des Nanotechnologies de Lyon (INL) UMR 5270 CNRS- INSA-ECL-UCBL, France

²Ecole Centrale de Lyon, 36 Avenue Guy de Collongue, 69134, Ecully, France

³INSA de Lyon, Bat. Blaise Pascal, 7 Avenue Capelle, 69621, Villeurbanne, France

⁴Imec, Kapeldreef 75, 3001 Leuven, Belgium

*Christian.Seassal@ec-lyon.fr

Abstract: In this paper, we present the integration of an absorbing photonic crystal within a monocrystalline silicon thin film photovoltaic stack fabricated without epitaxy. Finite difference time domain optical simulations are performed in order to design one- and two-dimensional photonic crystals to assist crystalline silicon solar cells. The simulations show that the 1D and 2D patterned solar cell stacks would have an increased integrated absorption in the crystalline silicon layer would increase of respectively 38% and 50%, when compared to a similar but unpatterned stack, in the whole wavelength range between 300 nm and 1100 nm. In order to fabricate such patterned stacks, we developed an effective set of processes based on laser holographic lithography, reactive ion etching and inductively coupled plasma etching. Optical measurements performed on the patterned stacks highlight the significant absorption increase achieved in the whole wavelength range of interest, as expected by simulation. Moreover, we show that with this design, the angle of incidence has almost no influence on the absorption for angles as high as around 60°.

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1. Introduction

Thin film crystalline silicon solar cells are one of the promising candidates for low cost photovoltaics. Considering fabrication costs, the thickness of the crystalline silicon layer should be limited as much as possible [1]. Meanwhile, increasing the absorption through light management is essential to achieve reasonable conversion efficiency on such devices. More precisely, light trapping schemes are essential to capture the red and near-infrared portion of the solar spectrum [2, 3]. Photonic Crystals (PCs) have been considered as promising candidates for improving the optical performance of solar cells. In particular, various designs have been proposed [4], including a planar periodic pattern which may act as a complex back reflector [5], or an efficient top anti-reflection coating [6, 7]. We have proposed a concept to produce highly efficient absorbers. It consists in patterning an hydrogenated amorphous silicon (a-Si:H) layer as a planar PC in order to couple incident light into Bloch modes standing over the light line, so as to control the photon lifetimes [8–11]. Based on this concept, PC-assisted a-Si:H solar cells were designed [12], and preliminary experimental results have been obtained [13, 14].

The use of monocrystalline silicon (c-Si) layers, with the advantage of a strongly reduced bulk recombination as compared to polycrystalline and amorphous layers, is a very promising way to build efficient and cost-effective photovoltaic devices. In this context, one of the key technologies is the so called Epifree technology [1], which allows fabricating a one-micron-thin monocrystalline layer on glass without resorting to epitaxial growth.

In that case, the integration of an efficient light trapping structure is fundamental for reaching high cell efficiencies and remains an important technical challenge. In particular, it should be highlighted that the photonic structure cannot be obtained just by a conformal deposition of the absorbing layer (like in the case of a-Si:H) on a pre-patterned wafer. Indeed, c-Si layer are inherently flat and photonic structures like periodic grating or PCs should be directly etched on the c-Si layer [15].

To generate such PC patterns, laser holographic lithography (LHL) is appropriate since it is a potentially high throughput and a maskless process compatible with large surfaces, particularly well-suited to the fabrication of periodic patterns [16–19]. The influence of its main parameters has been investigated in a previous study to produce the desired patterns [13].

In this paper, we report on the design and the fabrication of Epifree c-Si thin-film solar cell stacks integrating 1D and 2D PC square lattice patterns on the front – junction - side. The global design of those cells has been optimized optically by finite difference time domain (FDTD) simulations and is introduced in section 2. Topographical parameters have been scanned to calculate the absorption to provide guidelines for experimental developments, as developed in section 3. After presenting the general fabrication process of the patterned cells, we show the 1D and 2D patterning process of the active layer by combining LHL with reactive ion etching (RIE) and inductively coupled plasma (ICP) etching steps. Finally, in section 4, the optical properties of the fabricated solar cell stacks, including their angular response, are investigated experimentally and compared to the simulation results.

2. Optical design of 1D PC c-Si thin film solar cells

2.1. Structure description

The targeted structure investigated in this work is composed, from the back to the front, of a glass substrate, a 100 nm aluminium (Al) back-contact and reflector, a 20 nm highly-boron-doped c-Si layer acting as back-surface field, a 1 μm thick epitaxy free c-Si active layer overlaid by a 10 nm i/n^+ doped a-Si:H layer and finally a 80 nm thick indium tin oxide (ITO) layer acting as a transparent and conductive electrode. The three latter layers are patterned as a PC (shown in Fig. 1).

Besides, the c-Si active layer has a thickness of only 1 μm , a parameter which strongly impacts the final cost of the solar cell at an industrial scale. The ITO layer acts both as a conductive layer and as an anti-reflection coating (ARC).

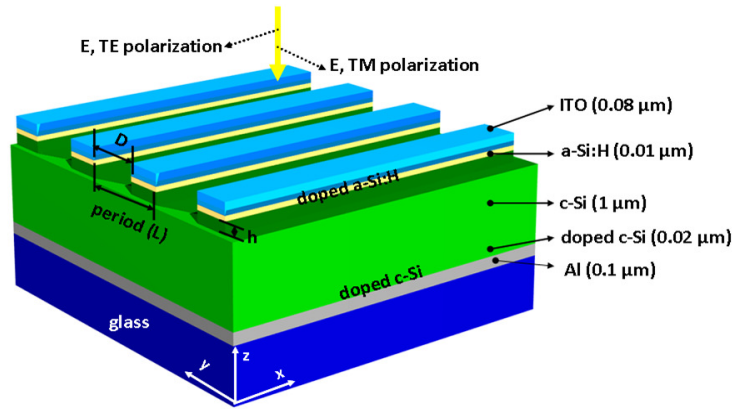


Fig. 1. Complete patterned stack studied, in the case of a 1D PC. Photogenerated carriers originating from the c-Si active layer are collected through the ITO and the Al layers.

In our design, the bottom electrode is an Al layer that also acts as a reflector. Although Al suffers from a higher parasitic absorption than Ag, it was selected for its lower cost and its compatibility with anodic bonding to glass [1].

Therefore, the incident light is expected to be preferentially absorbed in the active layer of c-Si, after assistance by the front anti-reflection layer of ITO and the back reflector layer of Al. In a fully operational solar cell, the photogenerated carriers will be collected by the bottom Al and the top ITO layers.

2.2. Simulation methodology

The FDTD method is used to solve Maxwell's equations in complex geometries [20] and is available into commercial packages [21]. It is suitable to analyze the wave propagation in thin film solar cells including wavelength or sub-wavelength scale patterns [22]. In this work, it has been used to calculate the absorption of complete patterned solar cells, as well as the specific absorption in the active c-Si layer. The optical performance of the structures will be indicated by their integrated absorption, a percentage defined by the ratio between the absorbed optical power over the incident optical power, and integrated over the whole 300-1100 nm spectral range. We consider an illumination at normal incidence, and an AM1.5G solar spectrum intensity distribution. These boundaries roughly and respectively correspond to the lowest wavelength of the solar spectrum and the c-Si gap. The refractive indexes of the materials, ITO, a-Si:H and c-Si, are all from ellipsometric measurement. In addition, the angular dependence is simulated by using CAMFR [23], which is based on the Rigorous Coupled Wave Analysis (RCWA) method [24] under non-normal incidence.

2.3. Absorption enhancement mechanisms in a 1D and 2D PC patterned c-Si layer

In order to assess the role of PCs to assist light absorption in c-Si, a first test was based on a single 1 μm thick layer, which is patterned as a 1D PC or a 2D PC with the same period (L), surfacic filling factor ($sff = \text{surface of air} / \text{surface of material}$) and etching depth (h), in order to compare its absorption spectra with the one of a planar c-Si layer. Figure 2 shows that the absorption in the patterned 2D and 1D PC layers is higher than in the planar layer. The integrated absorption averaged on both TE and TM polarizations (See Fig. 1) for the 1D and 2D patterned layer is respectively 48% and 57% of the solar spectrum, between 300 and 1100 nm, under normal incidence, much higher than the 31% value obtained for the planar c-Si layer. The parameters used in this calculation, $L = 0.61 \mu\text{m}$, $sff = 35\%$ and $h = 0.14 \mu\text{m}$ will be further justified in the next section. Moreover, under normal incidence, we observed that the absorption efficiency of a 2D PC layer does not depend on the polarization of the incident

light. In the unpatterned c-Si layer, from about 450 nm to 1100 nm, the clearly visible absorption resonances correspond to Fabry-Perot modes due to the large index contrast between air and the c-Si layer. These resonances can lead to absorption maxima, but the spectral density of modes is limited by the low thickness of the layer, leading to a low integrated absorption over the whole spectral range. Moreover, the large index contrast leading to a high reflection is also responsible for the low absorption ($\sim 50\%$) below 450 nm. Thus, the roles of the PC are twofold:

- To reduce reflection at short wavelengths
- To increase the spectral density of resonant modes at large wavelengths, leading to an increased absorption

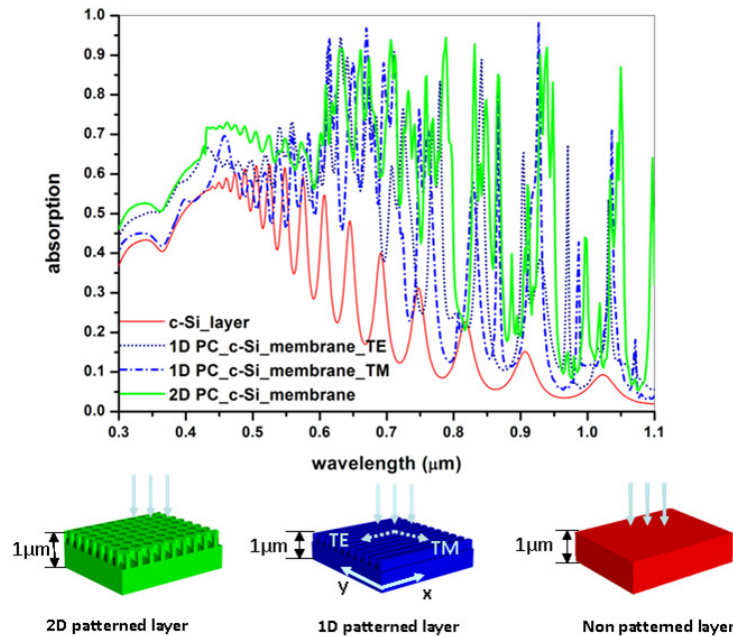


Fig. 2. Simulated absorption spectra of the patterned 2D and 1D PC c-Si layer with the same period, sff and etching depth ($L = 0.61 \mu\text{m}$, $sff = 35\%$ and $h = 0.14 \mu\text{m}$) compared to the one of a planar c-Si layer of the same thickness ($1 \mu\text{m}$).

2.4. Optimized absorption in complete c-Si solar cells

Based on the investigation of the single c-Si layer, a complete solar cell is designed and the values of lattice parameter (L) and sff , defined by D/L for a 1D PC are shown on Fig. 1, achievable using the selected technological processes. More precisely, the configurations of the 1D patterned cell were investigated by scanning simultaneously L between $0.2 \mu\text{m}$ and $0.8 \mu\text{m}$ per steps of $0.01 \mu\text{m}$, and sff between 0.05 and 0.7 per steps of 0.05 . Meanwhile, the range of h (height of c-Si PC) has been scanned from $0.02 \mu\text{m}$ to $1 \mu\text{m}$ per steps of $0.02 \mu\text{m}$. Those calculations were performed with a view to select the set of parameters leading to a maximized absorption in the “photonized” c-Si layer.

The results of these simulations indicate a maximal value for the integrated absorption of about 75% over the whole 1D patterned cell for $L = 0.61 \mu\text{m}$, $sff = 35\%$ and $h = 0.14 \mu\text{m}$. Corresponding absorption spectra are displayed in Fig. 3. The integrated absorption in the Si active layer itself is 44% for the optimized parameters of the 1D patterned stack, which is a relative improvement of 38% of the absorption efficiency compared to the 32% integrated absorption calculated for the unpatterned configuration at normal incidence. Meanwhile, the

absorption is quite robust when varying the topographical parameters (L , sff and h), in which 5% variation could lead to 1% integrated absorption floating in the c-Si layer. Moreover, the main absorption losses occur in the Al and ITO layers, which are 15% and 8% respectively.

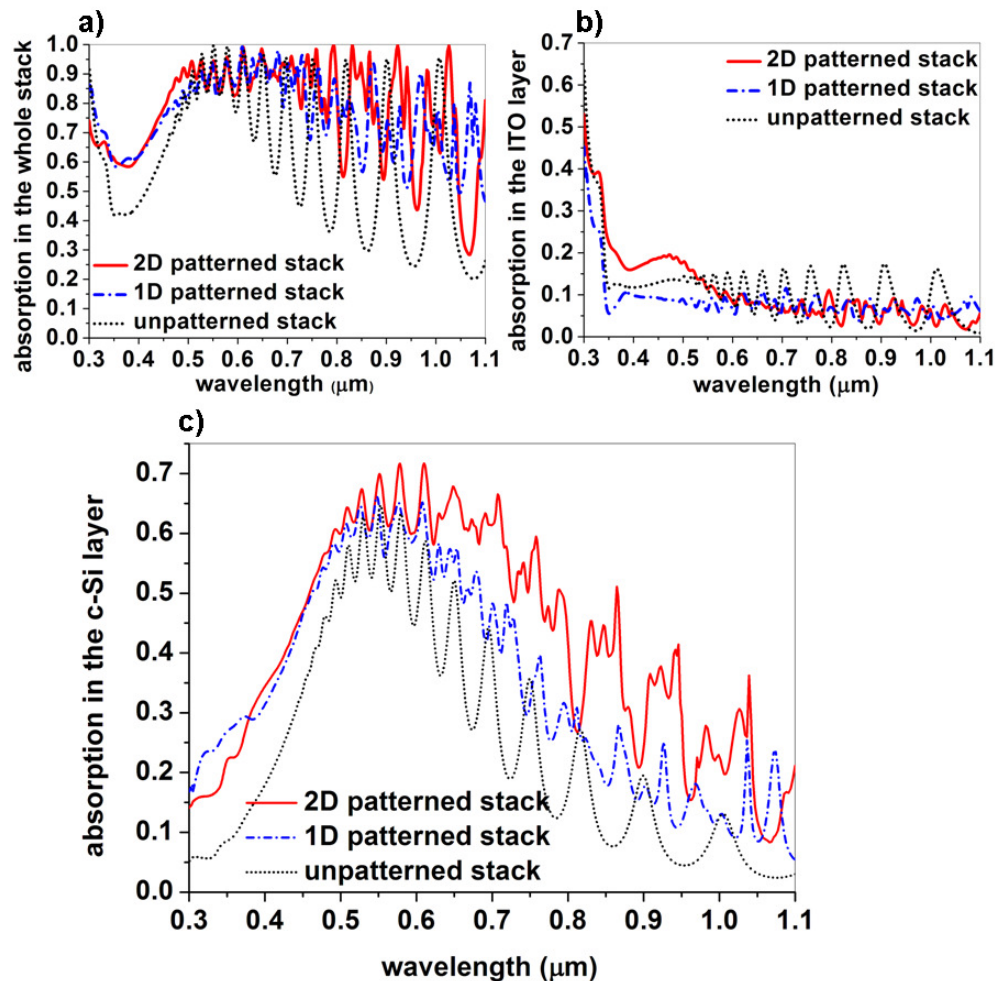


Fig. 3. Simulated absorption spectra averaged on both TE and TM polarizations in (a) the whole stack, (b) the ITO layer and (c) the c-Si layer for the 1D and 2D PC patterned stacks (L , sff and h set to $0.61\ \mu\text{m}$, 35% and $0.14\ \mu\text{m}$ respectively).

As discussed in section 2.3 in the case of simple c-Si layers, the absorption efficiency of 2D PCs is expected to lead to higher gains than their 1D counterpart. Absorption spectra simulated for 2D patterns, keeping the same lattice parameter, sff and etching depth, are also displayed in Fig. 3. From the comparison of the absorption spectra both in the whole stack (Fig. 3(a)) and in only the c-Si layer (Fig. 3(c)), a clear increase appears in the case of the 2D PC structure. However, light confinement in ITO is more important in the 2D case than in the 1D patterns; this leads to a higher useless absorption below 550nm, as shown in Fig. 3(b), and finally a lightly reduced absorption in the active layer below 400nm. At longer wavelengths, the incident photons are mainly lost by absorption in the Al layer. Finally, the 2D patterned stack leads to absorption of 48% of the incident light in the sole c-Si itself, a theoretical improvement of 9% of the absolute absorption efficiency compared to the 1D patterned stack under normal incidence. Due to technical limitations in terms of simulation,

including memory requirements and simulation time, we only considered the case of a square lattice 2D PC, with the same lattice parameter, sff and etching depth as in the case of the 1D PC.

It can be noted that for wavelengths larger than the 610 nm period, the absorption spectra of the PC structures exhibit a complex shape, including peaks which are not as regularly spaced as the Fabry-Perot resonances of the unpatterned structures. These peaks are attributed to the Bloch mode resonance of the PCs. These additional peaks clearly contribute to the absorption enhancement in the long wavelength range.

3. Fabrication of the PC patterned Epifree stack

In order to confirm the positive impact of the Photonic Crystal on an Epifree solar cell stack, we fabricated preliminary demonstrators devoted to absorption efficiency assessment. The fabricated structures include c-Si, Al and ITO layers, which are necessary to achieve a solar cell device. Compared to the structure discussed in section 2, only the 10 nm a-Si:H layer was not deposited, with a view to simplify the process, and since its optical role is negligible. As a starting point, the Epifree stack is constituted of glass/Al(1 μ m)/c-Si(1 μ m), see Fig. 4(a). A 0.1 μ m thick SiO₂ layer is deposited by plasma-enhanced chemical vapour deposition (PECVD) on the top of this stack to act as a hard mask during the etching of the c-Si layer, and is further removed. This stack is then patterned using LHL, combined with RIE and ICP techniques. LHL consists in irradiating a photosensitive resist using an interference pattern [13–15] (Fig. 4(b)). We implemented this generic process using a laser source at 266 nm and a NEB-22 negative tone Chemically Amplified Resist (CAR), spun after a hexamethyldisilazane (HMDS) adhesion promoter treatment. The use of negative resist is well-suited to the generation of slits or holes in the resist, as in the case of, respectively, the 1D and 2D PC designs. The selected LHL laser wavelength enables the patterning of structures with lattice parameters around 0.6 μ m. The process parameters can be tuned in order to reach the targeted parameters of the PC (lattice parameter L , sff and h). After LHL, development, and a descum steps [13, 25], the patterns are transferred into the SiO₂ hard mask (See Fig. 4(c)) using RIE process with a 16 sccm CHF₃ flow, at 15 mTorr, and with a power of 100 W for 900s. The c-Si layer is finally etched through the SiO₂ mask by ICP using 50 sccm for Cl₂, with an ICP power of 500 W and a RF power of 100 W, for 200s (see Fig. 4(d)). As a result of this combination of RIE and ICP, vertical sidewalls are obtained, as shown on the SEM profile views, Fig. 5(b) and 5(d). Then, the resist and SiO₂ are removed again by RIE (Fig. 4(e)). As shown in Fig. 5(a) and 5(c), the lines and holes are patterned with a reasonable regularity and roughness. The achieved period of 1D and 2D stacks are in the targeted range of 0.55-0.65 μ m; the sff is just in the range corresponding to the optimal configuration (35%-60%), and etching depth is just over the optimized 0.1-0.14 μ m range. A 75 nm thick ITO layer is finally deposited on top of the samples by sputtering (see Fig. 4(f)).

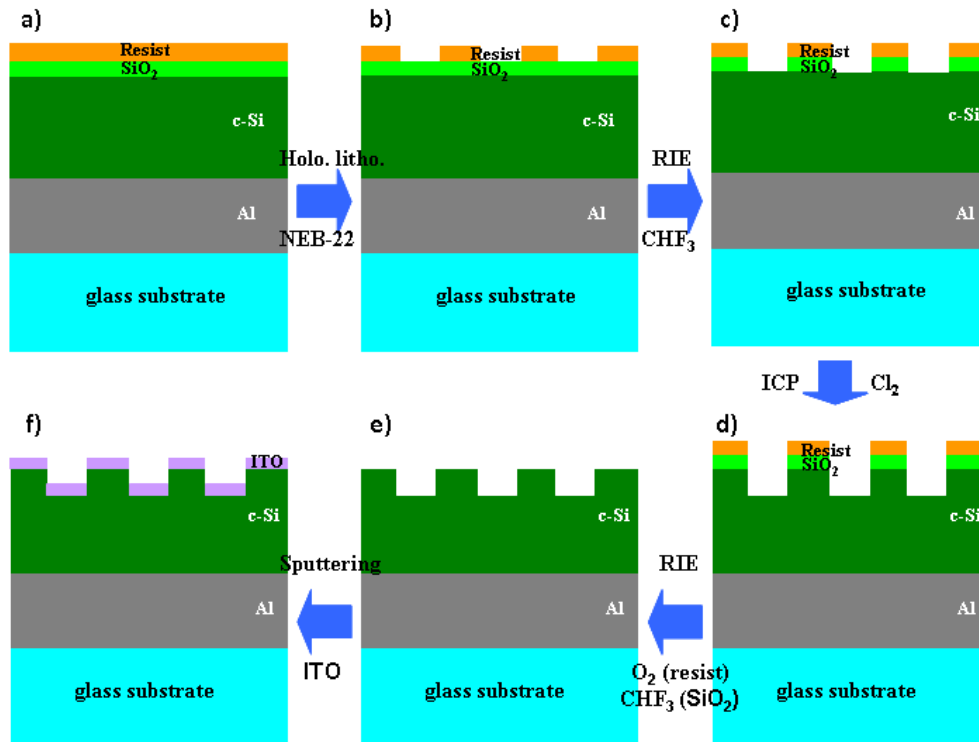


Fig. 4. Main steps of the processes enabling to generate the patterned stack as a 1D and 2D PC through LHL (a, b), RIE(c, e) and ICP (d) combined with (f) final sputtering.

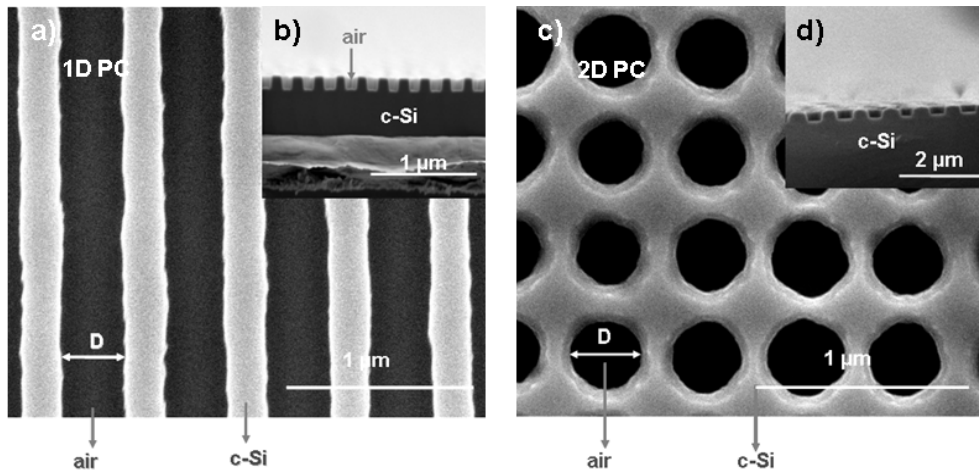


Fig. 5. Top (a), (c) and profile (b), (d) view of a c-Si layer patterned as a 1D network ($L \approx 0.55 \mu\text{m}$, $D \approx 0.28 \mu\text{m}$) and a 2D network of round holes ($L \approx 0.55 \mu\text{m}$, $D \approx 0.35 \mu\text{m}$) into a 0.18–0.25 μm depth. Pictures obtained by SEM after removing the hard mask layer and before ITO deposition.

4. Characterization of PC patterned c-Si thin film solar cell stacks

In order to determine the optical properties of the patterned stack presented in Fig. 5(a) and 5(b), absorption measurements were performed using an integrating sphere. In these experiments, the sample is illuminated by unpolarized light, so as to perform reflectance (R)

and transmittance (T) measurements with a $\sim 1 \text{ mm}^2$ spot size and with an 8° angle of incidence, as imposed by the integrating sphere measurement. The absorption $A(\lambda) = 1 - R(\lambda) - T(\lambda)$ is then simply derived. The absorption of the whole patterned stack was measured between 300 nm and 1100 nm with a 10 nm step. The absorption spectrum of the unpatterned stack was also measured as a reference. The corresponding spectra of 1D and 2D patterned stacks are simulated by FDTD. Measured and computed spectra are reported in Fig. 6(a) and 6(b). The stacks which are really fabricated have been considered and calculated for that purpose. Due to an uncertain determination of the geometrical parameters, and the inhomogeneity of the *sff* and measurement step length, as well as the roughness of the patterned cells, calculated and measured absorption spectra exhibit slightly different features. However, strong similarities are noticeable and the same general trends can be deduced both in 1D and 2D patterned stacks. As expected, the measured and simulated absorption spectra corresponding to patterned stacks display significantly higher values than for the unpatterned case. Moreover, the measured integrated absorption in the 2D and the 1D patterned stacks are 79% and 71% respectively, which are higher than the 50% reference.

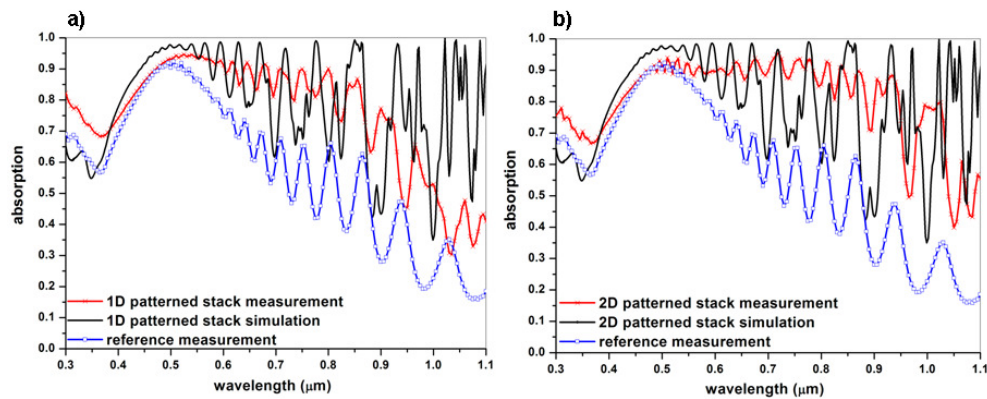


Fig. 6. Comparison of the measured and simulated absorption spectra for the (a) 1D and (b) 2D patterned stacks.

In addition, the optical impact of a 75 nm thick ITO layer was analyzed for both the 1D and 2D patterned stacks. The measured absorption spectra are displayed in Fig. 7.

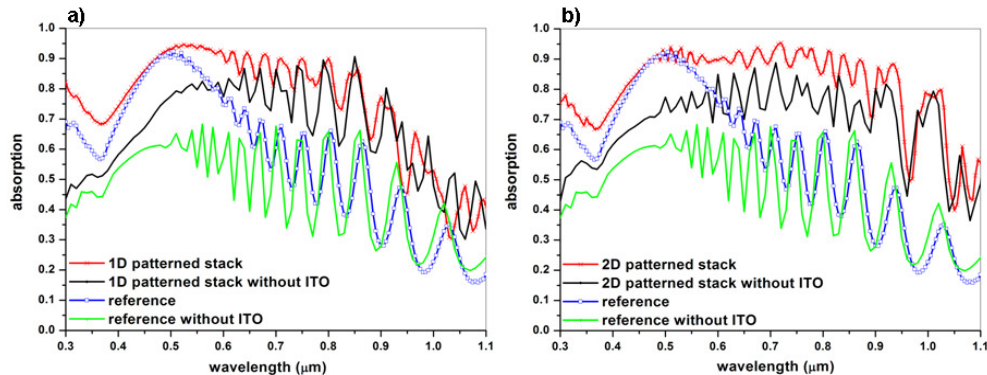


Fig. 7. The measured absorption spectra comparison between the (a) 1D and (b) 2D patterned and the unpatterned stacks with and without the front ITO layer.

Figure 7 shows that samples with ITO, either patterned or unpatterned, have a higher absorption than the ones without ITO. Given its thickness of 75 nm, the ARC effect takes place around a resonance wavelength of $\sim 550\text{-}600 \text{ nm}$. It indicates that the ITO layer fulfils

its role of being an anti-reflector layer in the short wavelength range. It should also be mentioned that below 350nm, a substantial part of the incident light is absorbed in the ITO layer. This was already predicted in the simulated spectra displayed in Fig. 3.

Additionally, the influence of the angle of incidence (α) on the integrated absorption of the 1D and 2D patterned stacks was investigated and compared to the reference stack, as displayed in Fig. 8(a). This corresponds to structures patterned with optimised parameters, including the 0.075 μm thick ITO layer. The integrated absorption is compared by varying α in simulations [0°-86°] and in measurements [6°-46°] and [6°-26°] for patterned 1D and 2D stack, respectively. The angle of incidence range is reduced in the measurements due to the measurement setup and sample size limitations. The corresponding absorption spectra at $\alpha = 26^\circ$, both simulated and measured, are displayed in Fig. 8(b) and 8(c), in the case of 1D and 2D patterned stacks, respectively.

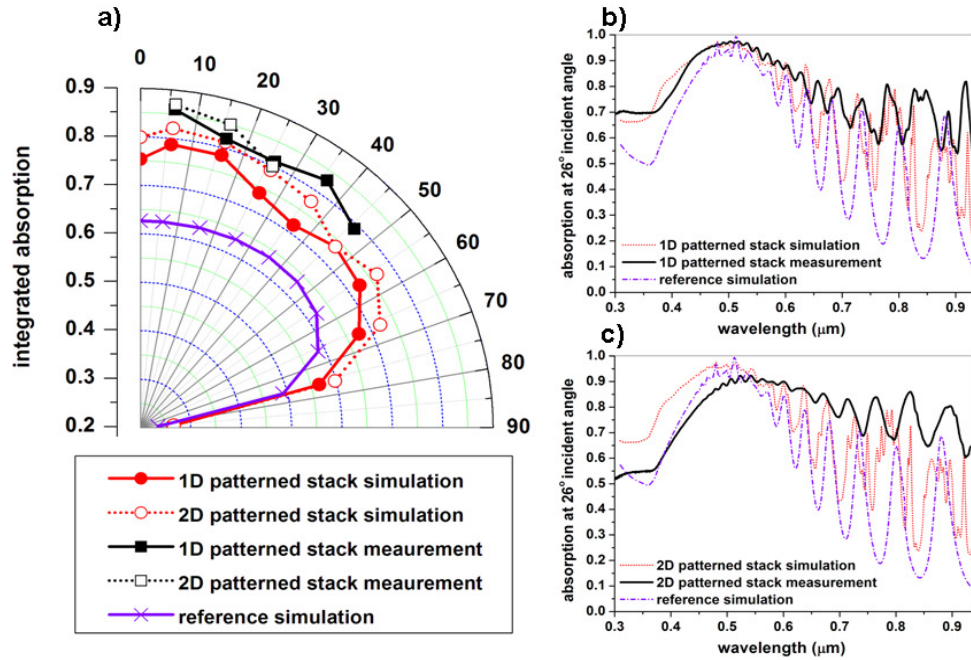


Fig. 8. (a) the incident angle dependence comparison between simulation and measurement of the 1D and 2D patterned stacks, as well as a reference stack, the absorption spectra comparison at 26° incident angle in (b) 1D patterned stack and (c) 2D patterned stack.

Figure 8 first illustrates that patterned samples exhibit a higher integrated absorption at any angle of incidence. Moreover, although optical resonances are used to increase the absorption efficiency, the behaviour is little dependent on the angle of incidence. At $\alpha = 66^\circ$, only a 8% and a 6% decrease in absolute value is observed compared to the value at $\alpha = 6^\circ$ for the simulated 1D and 2D patterned stack, respectively. Besides, optical measurements reveal that there is a 7% decrease of the integrated absorption at $\alpha = 46^\circ$ for the 1D patterned stacks and a 2% decrease of the integrated absorption at $\alpha = 26^\circ$ for the 2D patterned ones. Such behaviour is of particular importance for solar cells without solar tracker. Figure 8(b) and 8(c) illustrate that the shape of the absorption spectra are extremely similar to those corresponding to normal incidence. However, it should be highlight that the simulated integrated absorption is higher at around 10° than at normal incidence. This is explained by the possibility to couple incident light into resonances with a higher symmetry diversity at oblique incidence than at normal incidence [12]. Finally, it appears that the absorption is always lower in the theoretical case than when determined by measurements. Possible

reasons for this quantitative discrepancy are the differences between the designed and the fabricated structures, for example the inhomogeneous lattice parameters, the etched profile slopes and roughness.

5. Conclusion

Thin-film c-Si photovoltaic solar cells stacks integrating 1D or 2D PCs have been designed and optimized. As a result, we demonstrated that with proper nano-patterning parameters, a PC-assisted micrometer-thick structure enables an absorption increase in the c-Si active layer up to 38% for the 1D and up to 50% for the 2D patterning, when compared to the unpatterned stack, from 300 nm to 1100 nm, taking into account the AM1.5G solar irradiance. Looking into the details of the absorption spectra, we show that there is both an effect of decreased reflectivity and increased photon absorption due to the coupling of incident light into Photonic Crystal Bloch modes. The absorption enhancement due to the PC structure integrated on Epifree c-Si was assessed and analyzed on dedicated samples including the ITO and Al layers, which will further be used as electrodes for the solar cells. A process based on laser holographic lithography, RIE and ICP etching, has been developed to enable the generation of such 1D and 2D PCs on surfaces of a few cm². Both the patterned and unpatterned reference cell structures were measured and compared to spectra calculated by FDTD. Despite some discrepancies due to inaccuracies in geometrical parameters, measured and calculated absorption spectra exhibit a very similar behaviour. Additionally, the optical characteristics of the fabricated samples are quite stable with regards to the angle of incidence.

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