Optimization of Si (core)/CZTS/ CZTSe (shell) nanowire array for solar cell

adnen melliti (adnenmelliti@yahoo.fr)
Universite de Tunis Ecole Nationale Superieure d'Ingenieurs de Tunis https://orcid.org/0000-0003-4233-6939

Research Article

Keywords: CZTS, CZTSe, Si, RCWA, solar cell, nanowire, heat losses

Posted Date: March 23rd, 2021

DOI: https://doi.org/10.21203/rs.3.rs-331984/v1

Optimization of Si (core)/CZTS/ CZTSe (shell)

nanowire array for solar cell

Adnen Melliti[1,2]

[1]Université de Carthage, Institut Préparatoire aux Etudes Scientifiques et Techniques, Laboratoire Matèriaux-Molécules et Applications, BP51 La Marsa 2070, Tunisia.

[2]Université de Tunis, Ecole Nationale Supérieure des Ingénieurs de Tunis, 5 Rue Taha Hussein – Montfleury – 1008 Tunis, Tunisia.

Abstract

We present an optical simulation of a solar cell employing core (Si) /shell (CZTS or/and CZTSe) vertically-aligned nanowire array. The method of the simulation is rigorous coupled wave analysis. In the first stage, we studied the case where the shell is composed of only CZTS or CZTSe. A larger absorption of CZTSe led to a larger value of the ideal short circuit current (41 mA/cm^2) in the case of CZTSe solar cell than in the case of CZTS solar cell (24 mA/cm^2). In the second stage, to avoid the heat losses in CZTSe solar cell without reducing the current, we proposed a shell composed of a 3µm of CZTS in the upper part and a 6µm of CZTSe in the lower part. The maximum ideal current value in this structure is almost twice as large as that of a planar solar cell with the same amounts of used materials.

Keywords: CZTS; CZTSe; Si; RCWA; solar cell; nanowire; heat losses

1. Introduction

Thanks to its particular optical properties, like the light scattering and trapping [1-3], nanowires (NWs) configuration is economical than other configurations because solar cell (SC) have relatively less material. Among these solar cells, kesterite solar cells based on n-Si nanowires coated by p-type CZTS are emerging as the most auspicious candidate for scalable SC development [4-5]. Indeed, CZTS has outstanding electrical and optical features as direct optical band gap of 1.5 eV [6] and large absorption coefficient ($10^{4} cm^{-1}$). Furthermore, the p-CZTS-n-Si heterojunction is of high quality and these solar cells are only consisting of earth-abundant and non-toxic constituents.

For a better performance of SC based on coated Si NWs, numerical simulation can play a significant role that can save time and cost of the research community. Especially that the power of the conversion efficiency of solar cell based on Si/CZTS core-shell nanowire array (1-1.3 %) [4-5] is relatively weak compared to the one reported, for example, for CIGS thin film solar cell (22.9 %) [7]. We need, among others, to optimize the geometrical parameters and to look for other material or composition of the shell.

In the present article, we present an optical simulation of a solar cell with core (Si)/shell (CZTS or/and CZTSe) vertically-aligned NWs array. It's made using rigorous coupled wave analysis (RCWA) with Pavel Kwiecien's rcwa-2d code for MATLAB (sourceforge.net/projects/rcwa-2d/), based on [8]. The validation of the used code was made in a previous work [3]. To maximize its value, we calculated the ideal short circuit current density (J_{ph}) as a function of the thickness of the shell and the ratio between the period array and the NWs diameter for different values of NWs height. The maximal value obtained of J_{ph} is almost twice as large as that of a planar solar cell with the same amounts of used materials. According to our best knowledge, an optical simulation of solar cell based on Si nanowires decorated with a thin layer of CZTSe or CZTSe and CZTS has been reported for the first time in the present study.

In section 2 we described the structure of the SC simulated. The first part of section 3 is devoted to the absorption and optimization of the geometrical parameters of SC based on Si NWs coated by CZTS to maximize the ideal short circuit current. The results obtained were compared, in the second part, to that of SC with CZTSe shell. The third part is reserved to study the SC with shell composed of CZTSe and CZTS layers.

2. Device setup

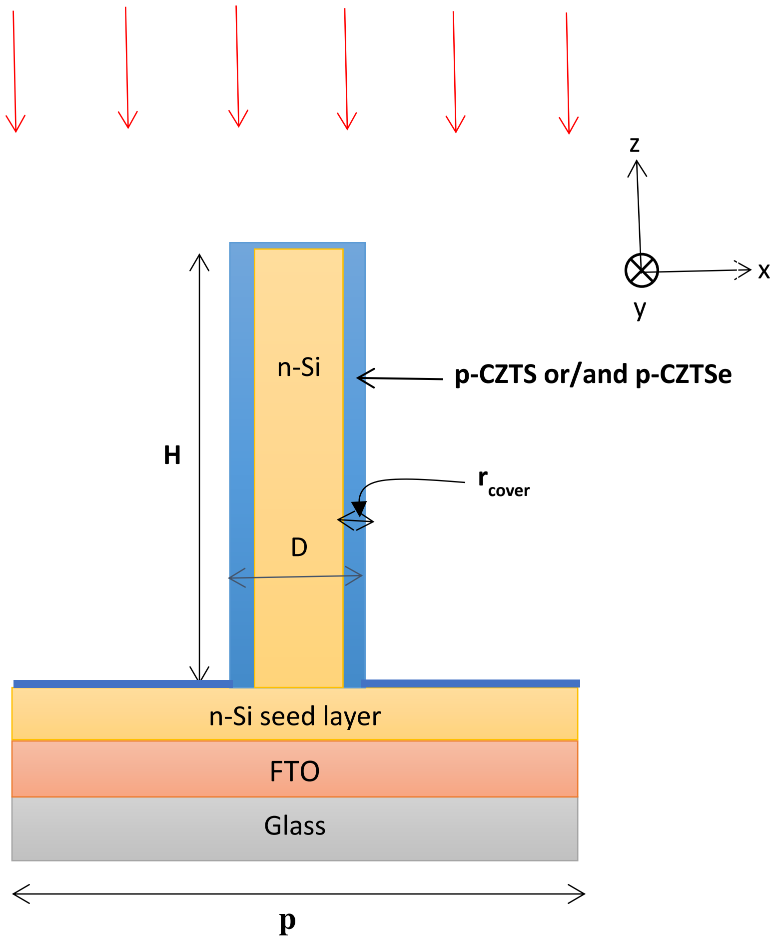
The structure of the solar cell, with square periodic array of NWs, investigated in this article are shown in figure 1.a. The NWs have n-Si core used as a buffer layer and coated by a thin CZTS or/and CZTSe absorber layer. The NWs are deposited on n-Si seed layer of 0.15 μm thickness. The upper surface of the Si-cores are covered by 2 0 nm of CZTS. The Si seed layer is covered by a thin CZTS or CZTSe absorber layer, of 20 nm thick, to connect all the absorber layers (blue cylinders in figure 1.a) to the upper device contact (not shown) which is deposited on this thin layer. The n-Si seed layer is electrically connected to the device contact by a 0.3 μm FTO layer.

A schematic of a planar solar cell (P SC), used to assess the improvement of the investigated solar cells caused by the using of the NWs configuration, is shown in figure 1.b.

Finally, we note that The optical constants of Si, CZTS, CZTSe, and FTO, used in the simulation were taken from references [9-12], and the polarization of the incident light, perpendicular to the substrate surface, is along the axe (Ox).

.

(a)


z
x
y
n-Si
p-CZTS or/and p-CZTSe
H
r cover
D
n-Si seed layer
FTO
Glass
p

(b)

Incident light

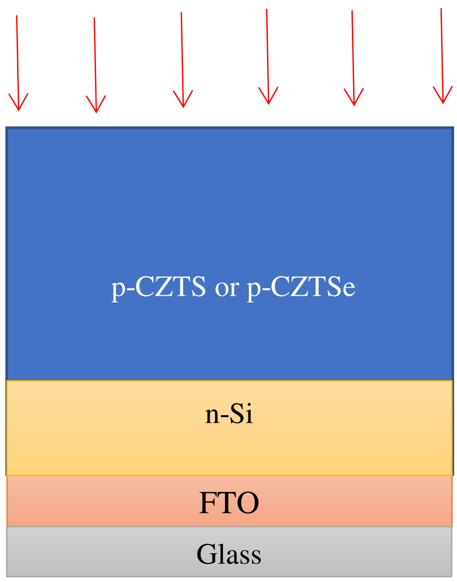


Figure 1: (a) Structure of a period of the investigated solar cells. H, D, p, and r_{cover} denote the height of the NWs, the diameter of the NWs, the array period, and the thickness of the CZTS or CZTSe layer respectively. **(b)** Structure of the planar solar cell based on the the same materials.

3. Results and discussion

3.1 CZTS NWs-solar cell

3.1.1 Absorption

To study the evolution of the absorption of the NWs solar cell with the geometrical parameters, we presented in figure 2 the absorptance spectrum obtained for different values of the thickness of CZTS layer (r_{cover}) and of the ratio between array period p and the diameter D of the coated NWs. The height of the NWs is 5 µm. We remark that the absorption spectra present many resonances and these are more pronounced for larger r_{cover}. On the other hand, we remark that the absorption is strongest for r_{cover} = 80 nm and p/D = 2. To interpret this result, we present in figure 3, the maps of the modulus of the component E_x of the electric field at light wavelength (λ) of 0.5 µm for the values of p/D and r_{cover} used in figure 2. We note that in the case of p/D=2 and r_{cover} = 80 nm, the light is efficiency confined inside the NWs unlike other cases where the intensity of the electric field is important outside the NWs. This is related to the NWs array effect which is strongest when the NWs are closest.

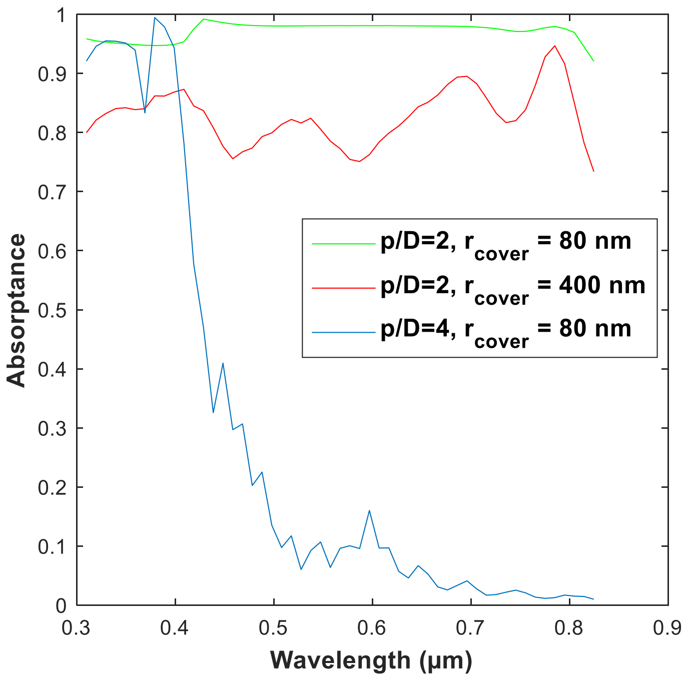
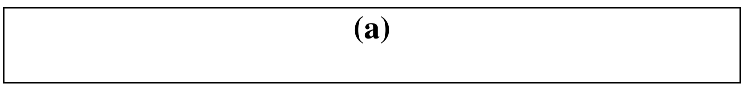
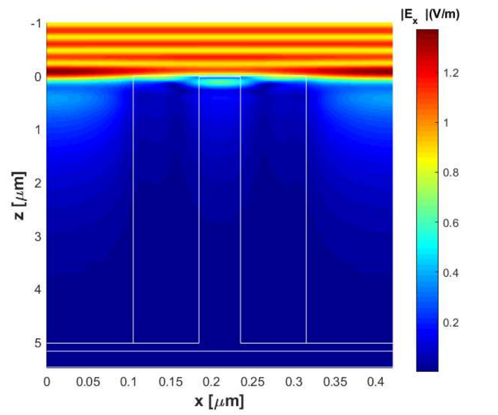
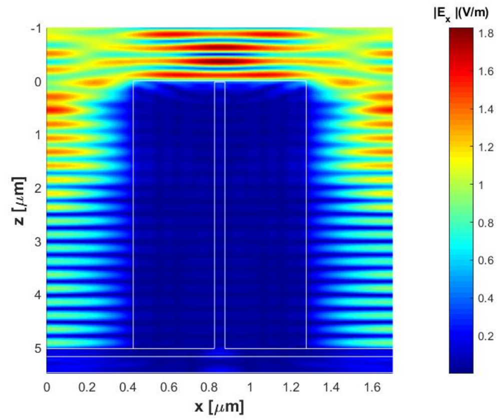


Figure 2: Absorption spectrum of CZTS NWs-SC obtained for different values of the thickness of CZTS layer (r_{cover}) and of the ratio between array period p and the diameter D of the coated NWs. The height of the NWs is 5 µm.





(b)



(c)

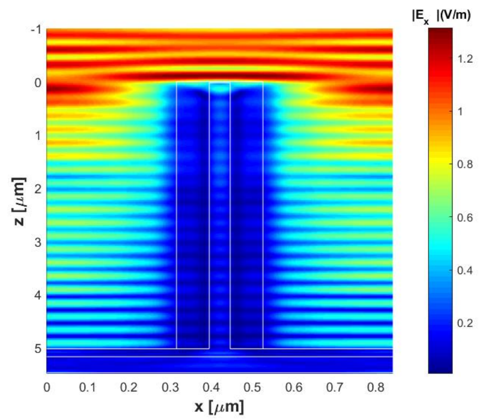


Figure 3: Component E_x of the electric field in CZTS NWs-SC at a light wavelength of 0.5 μm and for the ratio between array period p and the diameter D of the coated NWs (the thickness of CZTS layer) of 2 (80 nm) (a), 2 (400 nm) (b), and 4 (80 nm) (c). The height of the NWs is 5

μm. The white lines are guides for the eyes to show the underlying nanowire structure described in Figure 1.

3.1.2 Optimization of the geometrical parameters

The geometrical parameters (period, the thickness of the absorber layer as well as the NWs height) were optimized to maximize the ideal short circuit current created in the absorber layer assuming that every generated electron–hole pairs are collected. This current is estimated by:

$$J_{ph} = J_{ph}^{w} - J_{ph}^{Si} \quad (1)$$

where J_{ph}^{w} is the ideal short circuit current created in the whole SC and J_{ph}^{Si} is that created in the structure of the SC without the absorber layer. J_{ph}^{w} and J_{ph}^{Si} are given by:

$$J_{ph}^{w(Si)} = \frac{e}{h.c}\int_{300}^{\lambda_g} A^{w(Si)}(\lambda).I(\lambda).\lambda.d\lambda \qquad (2)$$

where $A^{w(Si)}(\lambda)$ is the wavelength-dependent optical absorptance, $I(\lambda)$ denotes the ASTM AM1.5G solar irradiance taken from Ref. [13], e, *h,* and *c* are fundamental physics constants: electron charge, Plank constant, and light celerity, respectively. The integration has been performed every 0.01 μm from 300 nm to the wavelength of the threshold of absorption of the absorber layer ($\lambda_g = 0.826\ \mu m$ for CZTS and 1.239 μm for CZTSe). We chose to begin the integration from 300 nm because the solar power is very weak below this wavelength,

To optimize the geometrical parameters, we calculated J_{ph} as a function of the thickness of the absorber layer (r_{cover}) and the ratio between the array period and the diameter of the coated NWs (p/D) for different values of Si NWs height (H). In figure 4.a, we presented the mapping obtained for H= 3 μm. The maximum value of Jph (23.7 mA/cm^2) is obtained for the optimal values of r_{cover} =82 nm and of p/D= 2.01. To interpret the existence of this maximum, we presented in figure 4.b the evolution with r_{cover} and p/D of the ratio of the absorber volume (volume of CZTS) and the volume of the period ($\frac{V_a}{V_p}$). We remark the absence of a maximum in the obtained mapping. We deduce that the existence of the maximum in Jph mapping is related to NWs effect.

Figure 4c shows the evolution with H of the optimal values of r_{cover} and p/D that give the maximal value of J_{ph} for a given value of H. We note that the optimal value of r_{cover} (82 nm) is independent of H. In contrast, the optimal value of p/D increases from 2 to 3 when H varies from 1 to 5 μm and remains constant for greater value of H. This increase is explained by the fact that with increasing H, we need less absorber material, consequently a greater p/D ratio, to reach the maximum value of Jph.

Figure 4.d shows that the maximal value of Jph saturates at about 24 mA/cm^2 and the enhancement of Jph from H=3μm isn't important (about 1%). Thus, the optimal value of H is 3μm.

In the following, we compare the optimal value of J_{ph} to the one of a P SC (figure 1.b) involving the same amount of materials (CZTS and Si). We obtained a value of 17 mA/cm^2 for J_{ph} created in the CZTS layer (without taking into account the Jph created in Si layer). Then, the optimal value of Jph of the NWs SC (24 mA/cm^2) is greater by about 41% than the one of P SC. This improvement of J_{ph} added to the improvement of the collection of generated minority carriers in CZTS layer, due to the radial geometry of the NWs SC, shows the superiority of the NWs SC.

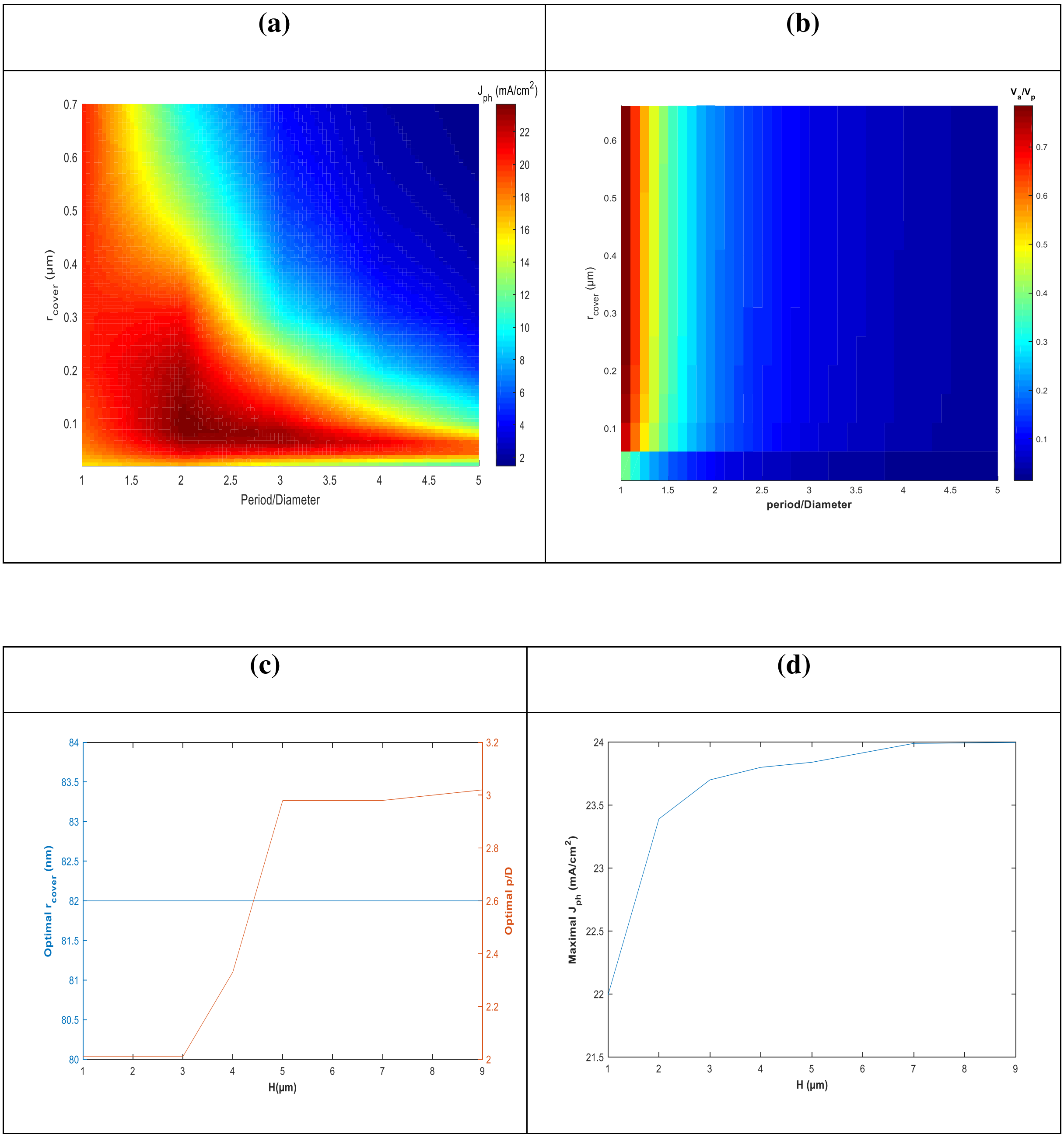


Figure 4: **(a)** Ideal short circuit current density (J_{ph}) computed for H = 3 µm as a function of the thickness of CZTS layer (r_{cover}) and the ratio between the array period p and the diameter D of the coated NWs. The 55 data points map has been linear interpolated on a 182 times larger grid. **(b)** Evolution with r_{cover} and p/D of the ratio of the absorber volume (volume of CZTS) and the volume of the period ($\frac{V_a}{V_p}$). **(c)** The optimal values of the thickness of CZTS layer (r_{cover}) and the ratio between the array period p and the diameter D of the coated NWs as a function of

coated NW height. **(d)** Maximal value of (J_{ph}) obtained for the optimal values of r_{cover} and p/D as a function of the coated NW height.

3.2 CZTSe NWs-solar cell

In this section, we present the results obtained when using CZTSe as absorber instead CZTS. As shown by figure 5, the CZTSe solar cell absorbs very efficiently for wavelength smaller than 1.1 µm. In figure 6, we present the evolution as a function of Si NW height of the optimal values of the thickness of CZTSe layer (r_{cover}) and of p/D and the maximal value of J_{ph} obtained for the optimal values of r_{cover} and p/D. We remark that Jph saturates at about 41 mA/cm^2 and the enhancement of Jph from H = 6µm is small (about 1%). Thus, the optimal value of H is about 6µm. This corresponds to optimal values of r_{cover} and of p/D of 102 nm and 2.98 respectively. We remark that the saturation value of Jph is much larger in the case of CZTSe SC than in the case of CZTS SC. This is explained by the strong absorption of CZTSe for wavelength between 0.826 and 1.236 µm, as shown by figure 5, that isn't the case of CZTS. On the other hand, the NW height of saturation of Jph is larger in the case of CZTSe SC than in the case of CZTS SC. Indeed, the penetration depth increases with wavelength and CZTSe absorbs long wavelengths, longer than 826 nm, that aren't absorbed by CZTS.

On the other hand, the value of J_{ph} of a P SC (figure 1.b) involving the same amount of materials is 21 mA/cm^2 (without taking into account the Jph in Si layer). Then, the maximal value of Jph of the NWs SC (41 mA/cm^2) is greater by about 95% than the one of P SC.

Given previous results, CZTSe NWs SC is more performant than CZTS NWs SC, but in this section we didn't take into account the lost as heat of a part of the energy of generated carriers in CZTSe [14-15]. Indeed, an important part of the photons of sun light have energy larger than CZTSe bandgap. Then, a part of their energy is wasted as heat. Consequently, this process reduces the efficiency of the cell. To avoid this problem, we propose in the following section a graded absorber SC.

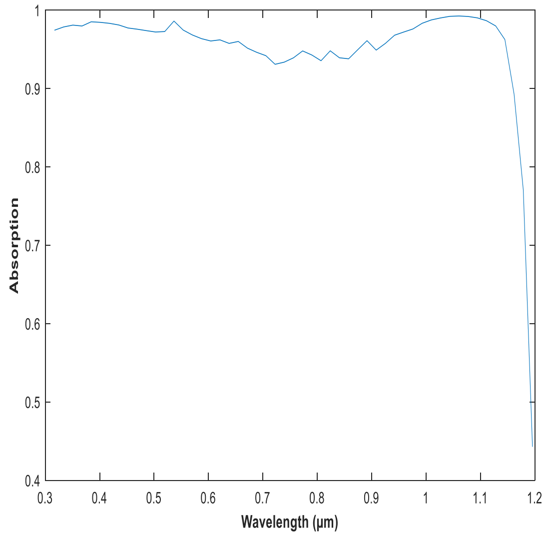
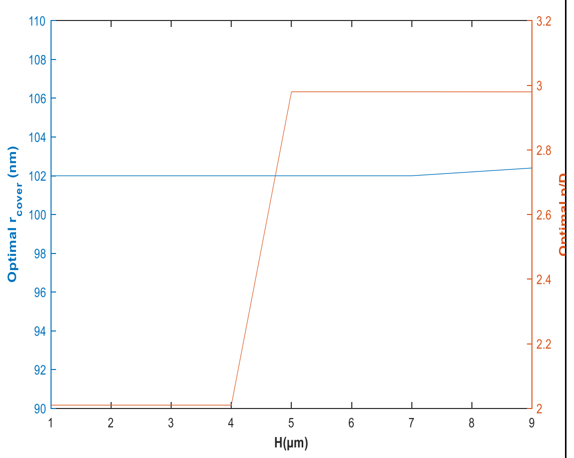


Figure 5: Absorption spectrum of CZTSe NW-SC. The thickness of CZTSe layer, the height of NWs and the ratio between the array period p and the diameter D of the coated NWs are 100 nm, 5 μm and 3 respectively.

(a)



(b)

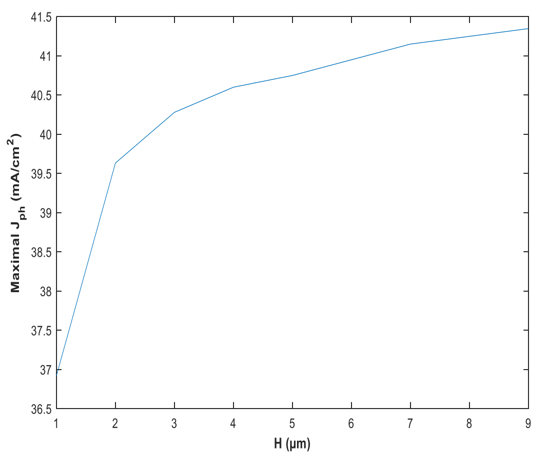


Figure 6: (a) The optimal values of the thickness of CZTSe layer (r_{cover}) and the ratio between the array period p and the diameter D of the coated NWs as a function of NW height. **(b)** Maximal value of (J_{ph}) obtained for the optimal values of r_{cover} and p/D as a function of coated NW height.

3.3 Graded absorber solar cell

In this section we replace the uniform CZTS or CZTSe shell by an upper CZTS shell and a lower CZTSe shell. The height of each shell is the optimal height found in the previous sections. On other words, 3 µm for CZTS shell and 6 µm for CZTSe shell. In this structure, most of photons with larger energy will be absorbed by CZTS layer. So, the amount of loss will be reduced.

In figure 7 we present the absorbance spectrum of the gradient SC obtained for p/D = 3 and r_{cover} = 80 nm. We remark that for wavelength smaller than 1.1 µm, the absorptance is almost 1.

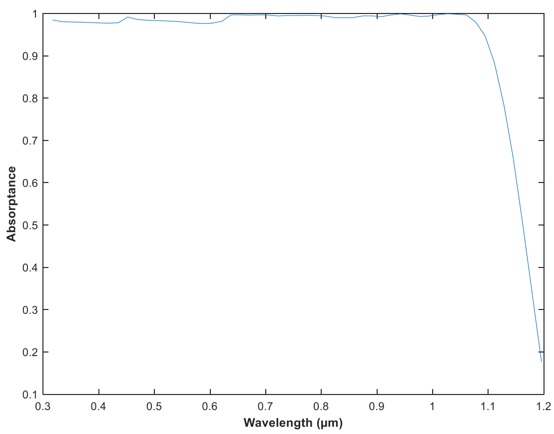
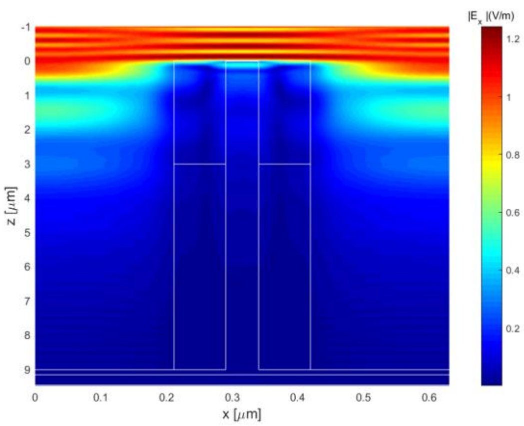


Figure 7: Absorption spectrum of gradient NW-SC. The thickness of the absorber layer and the ratio between the array period p and the diameter D of the coated NWs are 100 nm and 3 respectively.

In figure 8, we present the map of the modulus of the component E_x of the electric field at light wavelengths (λ) of 0.5 and 1 µm and for the values of p/D and r_{cover} of 3 and 80 nm respectively. We note that for wavelength of 0.5 µm, the intensity of the field is very weak outside the NWs. On the other hand, for 1 µm and in the region of CZTS shell, the radiant modes are dominant unlike in the CZTSe shell where the light is efficiency confined inside the NWs. Given the refractive index of CZTS and CZTSe are close, these results are related to the fact that at 1 µm, CZTS is transparent and CZTSe is absorbent that isn't the case at 0.5 µm where CZTS and CZTSe are absorbent.

(a)



(b)

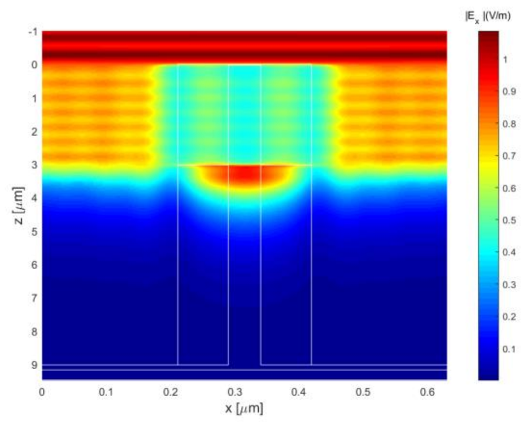


Figure 8: Component E_x of the electric field in gradient NWs-SC at a light wavelength of (a) 0.5 µm and (b) 1µm and for the ratio between the array period p and the diameter D of the

coated NWs (the thickness of absorber layer) of 3 (80 nm). The white lines are guides for the eyes to show the underlying nanowire structure described in Figure 1.

In figure 9 we presented a mapping showing the variation of Jph with r_{cover} and the ratio p/D. We note that the maximal value of the current is 41 mA/cm^2 and it's obtained for r_{cover} =102 nm and p/D= 2.98. This value, as in the case of CZTSe SC, is greater by about 95% than the one of P SC involving the same amount of materials.

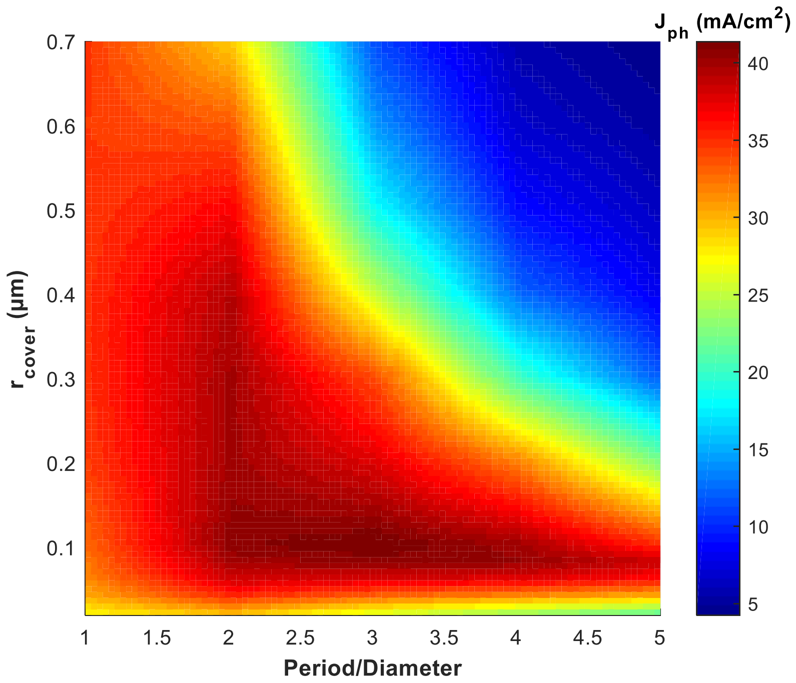


Figure 9: Ideal short circuit current density (J_{ph}) computed for gradient SC as a function of the thickness of absorber layer (r_{cover}) and the ratio between the array period p and the diameter D of the coated NWs. The 55 data points map has been linear interpolated on a 182 times larger grid.

4. conclusion

In this article, we optically simulated a solar cell with core (Si)/shell (CZTS or/and CZTSe) vertically-aligned NWs array to optimize the structural morphology. An optimum value of 41 mA/cm^2 has been found when the NWs are coated by an upper 3 µm-CZTS shell and a lower 6 µm-CZTSe shell. This configuration offers an excellent absorption of the sun light and minimize the heat losing. The optimal shell thickness and ratio of period and NWs diameter are of 102 nm and 2.98 respectively. The obtained value of J_{ph} is almost twice as large as that of a planar solar cell with the same amounts of used materials. We conclude that the investigated SC is a good candidate for the SCs based on semiconductors nanowires.

References

[1] A. Ghosh, R. Thangavel and A. Gupta, Solution-processed Cd free kesterite Cu2ZnSnS4 thin film solar cells with vertically aligned ZnO nanorod arrays, j.jallcoJ. Alloys Compdm. 694 (2017) 394-400.

[2] W. Sun, M. Brozak, J. C. Armstrong, J. Cui, IEEE 39th Photovoltaic Specialists Conference (PVSC) (2013).

[3] A. Melliti, Optical simulation of solar cell based on ZnO/ZTO core–shell nanowire array embedded in CZTS layer, Optical and Quantum Electronics 53, published online 24 January 2021, DOI: 10.1007/s11082-021-02738-w

[4] E. Peksu and H. Karaagac, Characterization of one-step deposited Cu_2ZnSnS_4 thin films derived from a single crystalline powder, Journal of Alloys and Compounds 774 (2019) 1117-1122.

[5] E. Peksu, O. Guller, M. Parlak, M. S. Islam, H. Karaagac, Characterization of one-step deposited Cu_2ZnSnS_4 thin films derived from a single crystalline powder, Physica E 124 (2020) 114382-114394.

[6] T. Ericson, F. Larsson, T. Törndahl, C. Frisk, J. Larsen, V. Kosyak, C. Hägglund, S. Li, and C. Platzer-Björkman, Zinc-Tin-Oxide Buffer Layer and Low Temperature Post Annealing Resulting in a 9.0% Efficient Cd- Free Cu2ZnSnS4 Solar Cell, Sol. RRL (2017) 1700001-1700008.

[7] M.A. Green, Y. Hishikawa, E.D. Dunlop, D.H. Levi, J. Hohl-Ebinger, A.W. Ho-Baillie, Solar cell efficiency tables (version 52). Prog. Photovoltaics Res. Appl. 26 (2018) 427-436.

[8] L. Li, "New formulation of the Fourier modal method for crossed surface-relief gratings," J. Opt. Soc. Am. A 14 (1997) 2758-2767.

[9] E. D. Palik. Handbook Of Optical Constants of Solids Academic Press, Orlando, 1998

[10] N. E. Gorji, Quantitative analysis of the optical losses in CZTS thin-film semiconductors, IEEE Transactions On Nanotechnology 13 (2014) 743-748.

[11] D. Cozza, C. M. Ruiz, D. Duché, S. Giraldo, E. Saucedo, J. J. Simon, and I. Escoubas, Optical modeling and optimizations of $Cu_2ZnSnSe_4$ solar cells using the modified transfer matrix method, Optics express 24 (2016) A1201-A1209.

[12] R. Thomas, T. Mathavan, M.A. Jothirajan, H.H. Somaily, H.Y. Zahran, I.S. Yahia, An effect of lanthanum doping on physical characteristics of FTO thin films coated by nebulizer spray pyrolysis technique, Optical Materials 99 (2020) 109518-109528.

[13] ASTM, Reference Solar Spectral Irradiance: Air Mass 1.5 Spectra, http://rredc.nrel.gov/solar/spectra/am1.5 (2020).

[14] I.M. Dharmadasa, N.D.P.S.R. Kalyanaratne, R. Dharmadasa, Effective harvesting of photons for improvement of solar energy conversion by graded bandgap multilayer solar cells, J. Natl. Sci. Found. Sri Lanka 41 (2013) 73–80.

[15] M. A. Contreras, J.Tuttle, A. Gabor, A. Tennant, K. Ramanathan, S. Asher, A. Franz, J. Keane, L. Wang, R. Noufi, High efficiency graded bandgap thin-film polycrystalline Cu(In,Ga) Se_2-based solar cells, Sol. Energy Mater. Sol. Cells 41–42 (1996) 231–246.

Figures

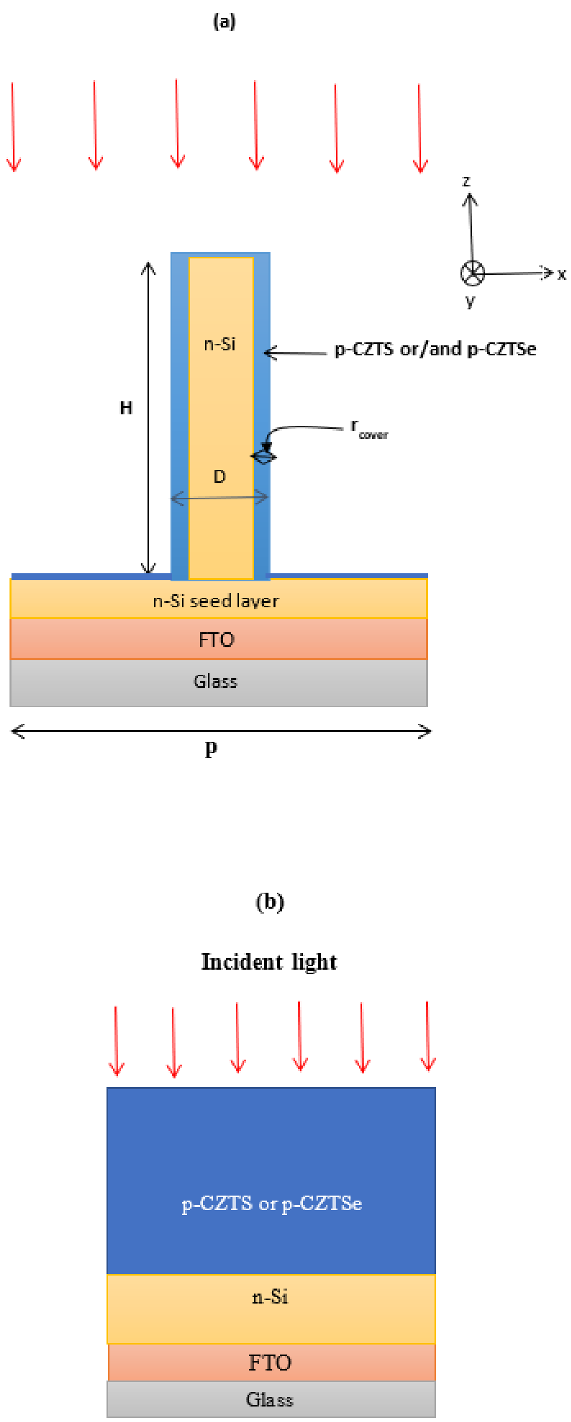


Figure 1

(a) Structure of a period of the investigated solar cells. H, D, p, and rcover denote the height of the NWs, the diameter of the NWs, the array period, and the thickness of the CZTS or CZTSe layer respectively. (b) Structure of the planar solar cell based on the the same materials.

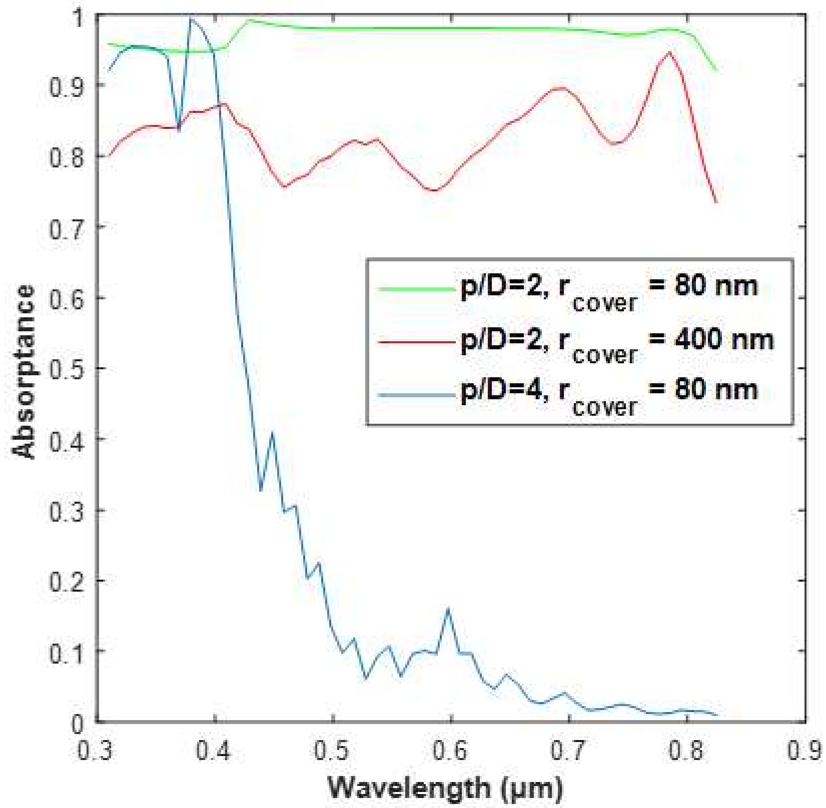


Figure 2

Absorption spectrum of CZTS NWs-SC obtained for different values of the thickness of CZTS layer (rcover) and of the ratio between array period p and the diameter D of the coated NWs. The height of the NWs is 5 μm.

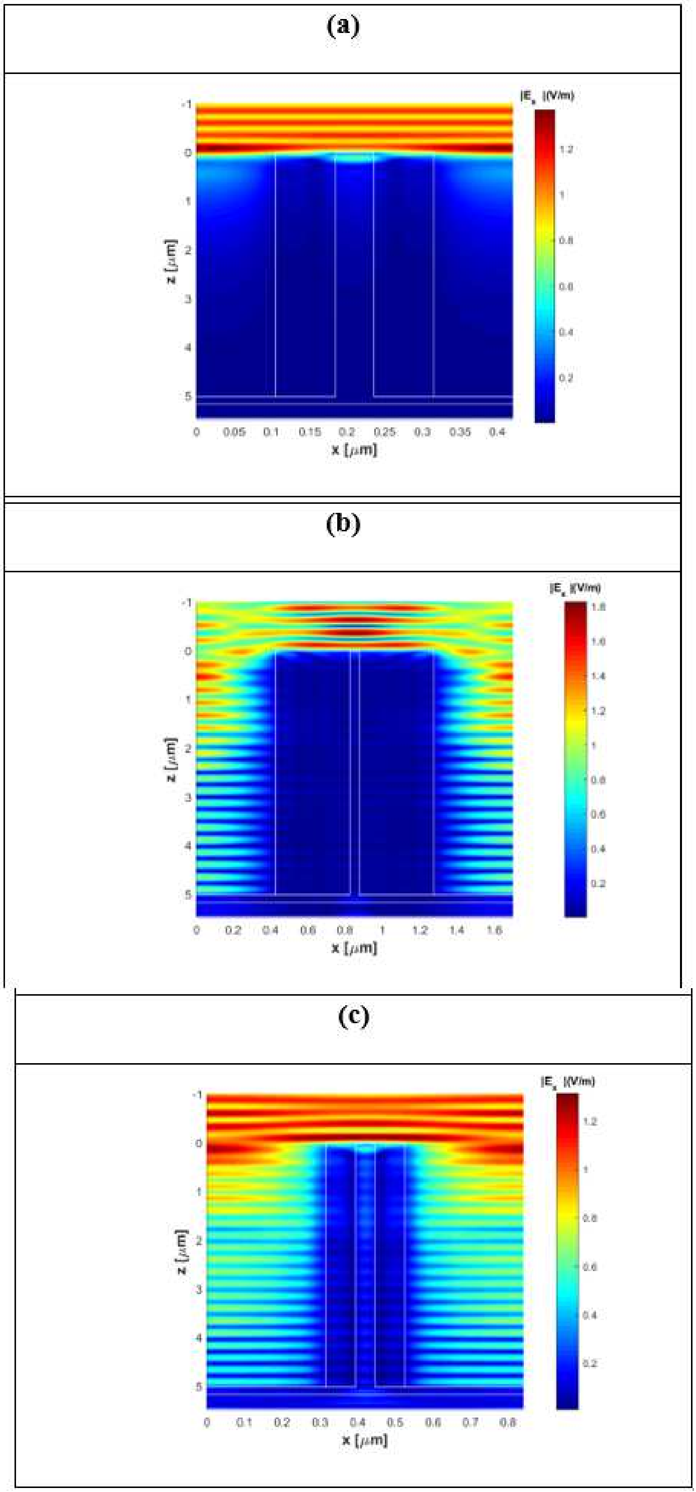


Figure 3

Component Ex of the electric field in CZTS NWs-SC at a light wavelength of 0.5 μm and for the ratio between array period p and the diameter D of the coated NWs (the thickness of CZTS layer) of 2 (80 nm) (a), 2 (400 nm) (b), and 4 (80 nm) (c). The height of the NWs is 5 μm. The white lines are guides for the eyes to show the underlying nanowire structure described in Figure 1.

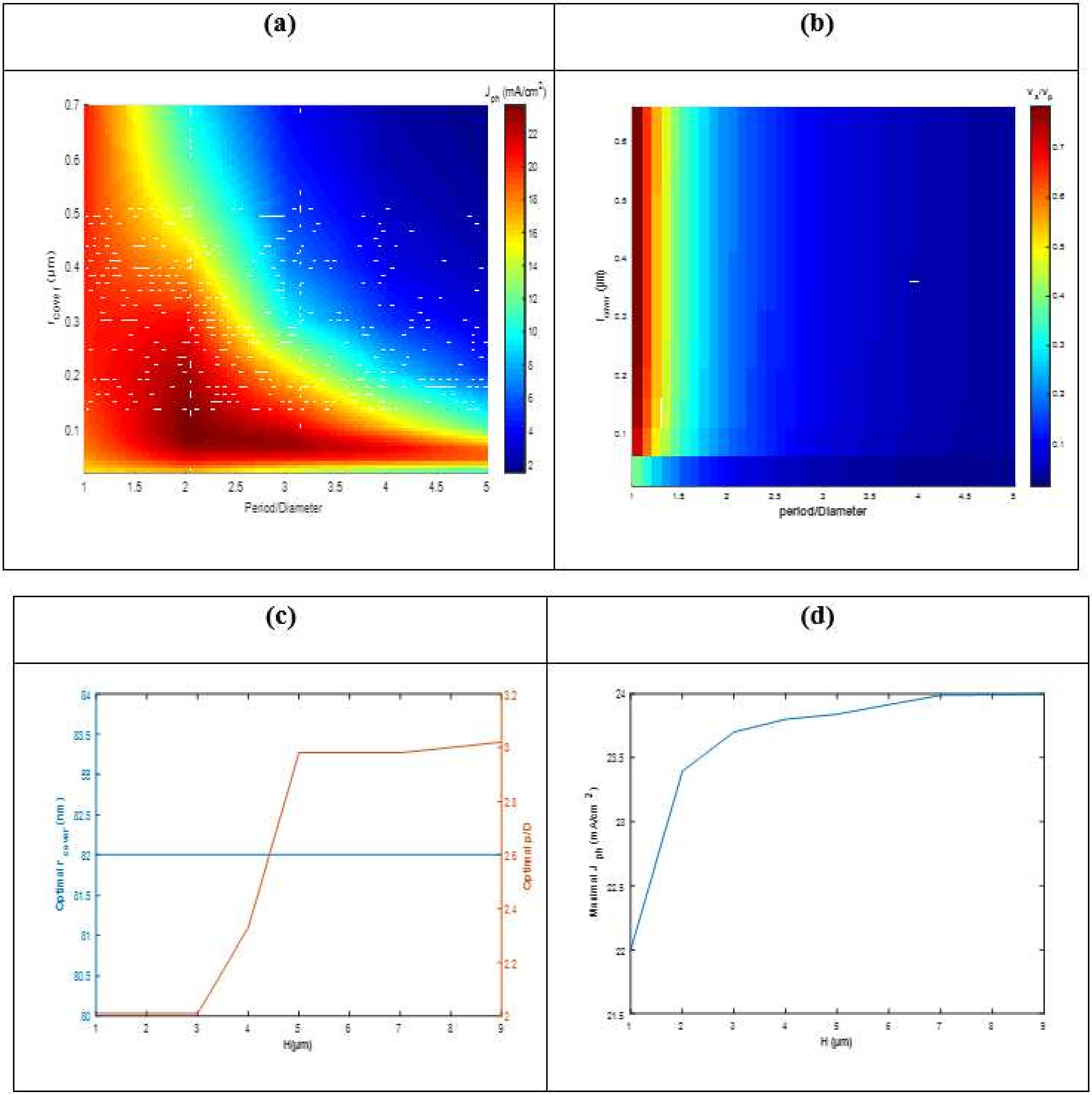


Figure 4

(a) Ideal short circuit current density (Jph) computed for H = 3 µm as a function of the thickness of CZTS layer (rcover) and the ratio between the array period p and the diameter D of the coated NWs. The 55 data points map has been linear interpolated on a 182 times larger grid. (b) Evolution with rcover and p/D of the ratio of the absorber volume (volume of CZTS) and the volume of the period (V_a/V_p). (c) The optimal values of the thickness of CZTS layer (rcover) and the ratio between the array period p and the diameter D of the coated NWs as a function of coated NW height. (d) Maximal value of (Jph) obtained for the optimal values of rcover and p/D as a function of the coated NW height.

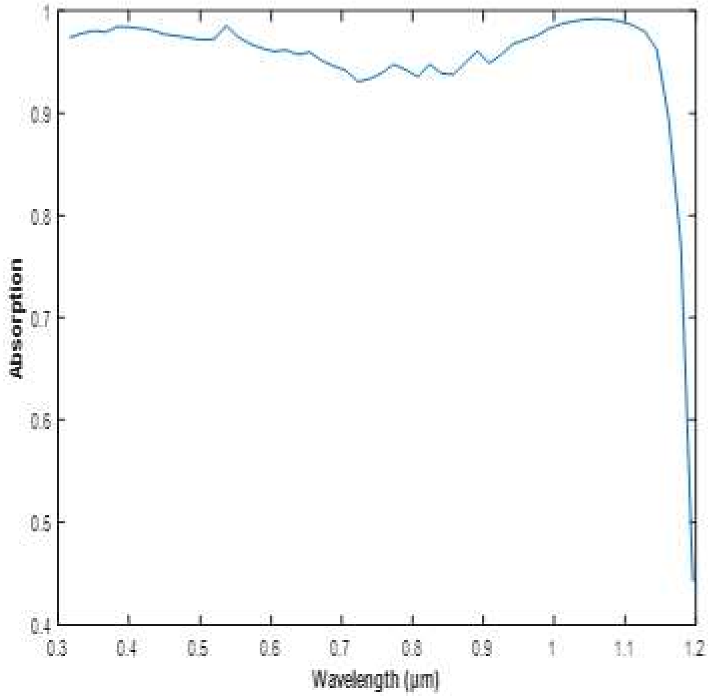


Figure 5

Absorption spectrum of CZTSe NW-SC. The thickness of CZTSe layer, the height of NWs and the ratio between the array period p and the diameter D of the coated NWs are 100 nm, 5 µm and 3 respectively.

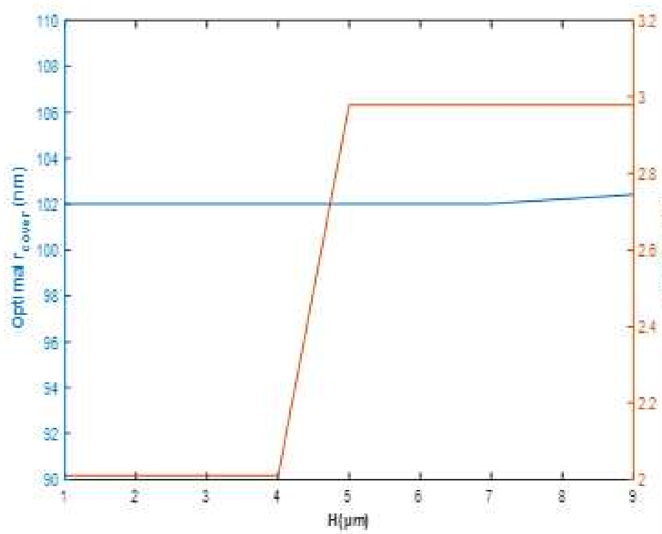
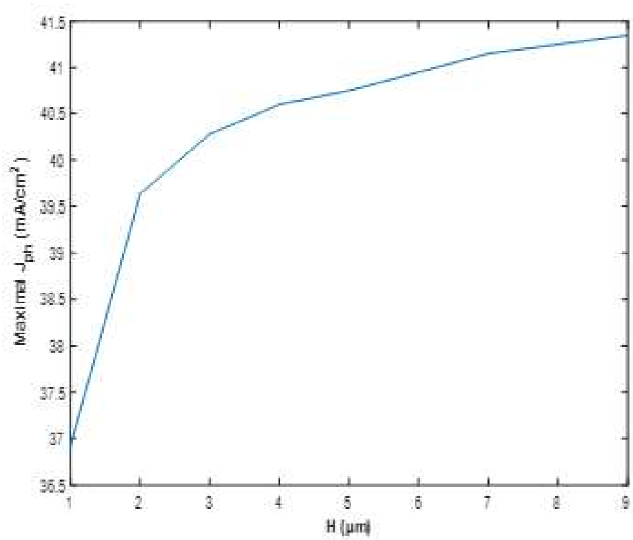
(a)	(b)
	

Figure 6

(a) The optimal values of the thickness of CZTSe layer (rcover) and the ratio between the array period p and the diameter D of the coated NWs as a function of NW height. (b) Maximal value of (Jph) obtained for the optimal values of rcover and p/D as a function of coated NW height.

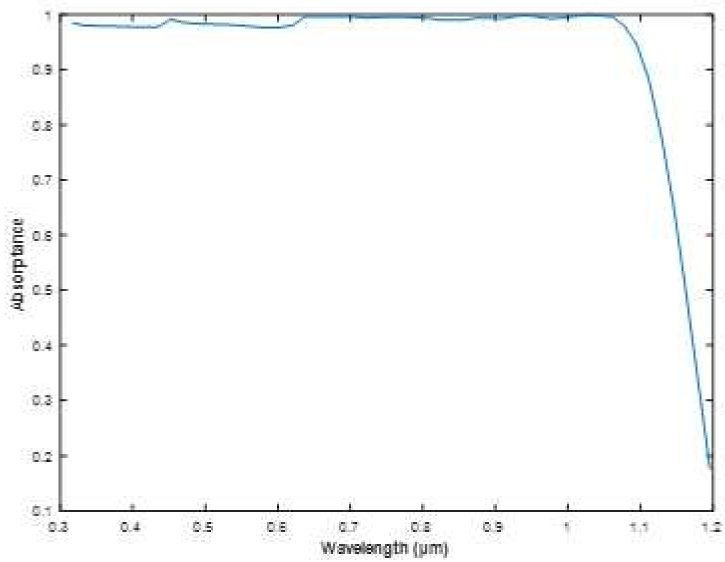


Figure 7

Absorption spectrum of gradient NW-SC. The thickness of the absorber layer and the ratio between the array period p and the diameter D of the coated NWs are 100 nm and 3 respectively.

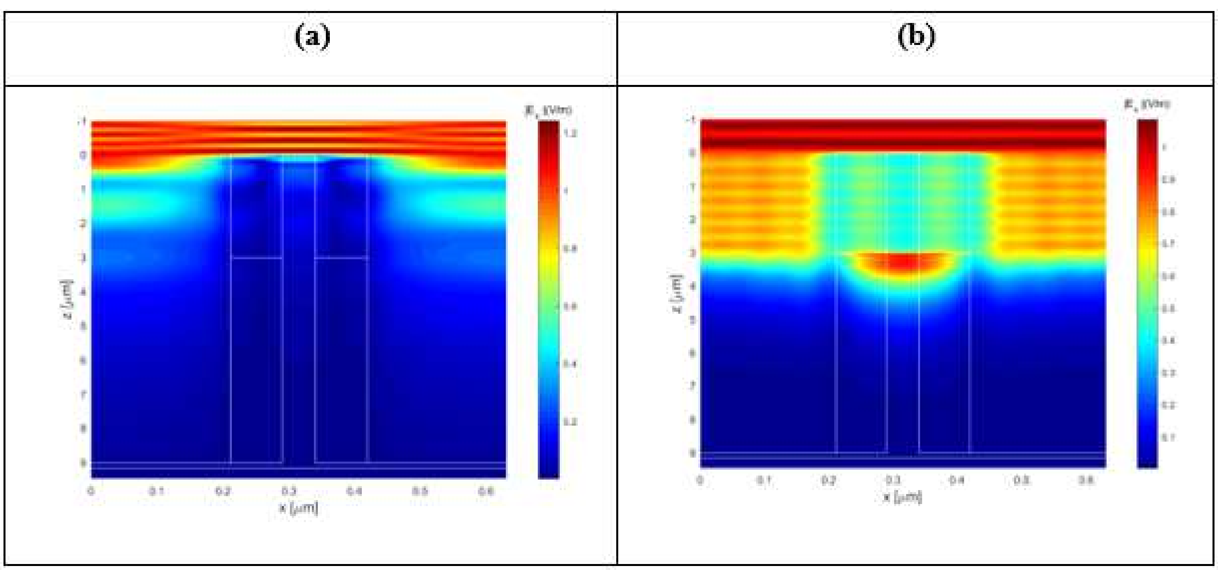


Figure 8

Component Ex of the electric field in gradient NWs-SC at a light wavelength of (a) 0.5 μm and (b) 1μm and for the ratio between the array period p and the diameter D of the coated NWs (the thickness of absorber layer) of 3 (80 nm). The white lines are guides for the eyes to show the underlying nanowire structure described in Figure 1.

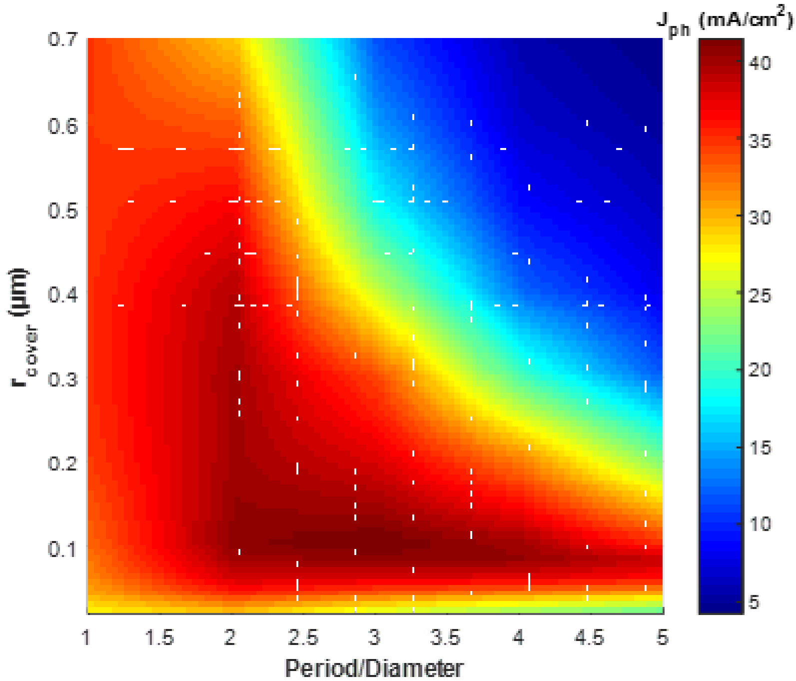


Figure 9

Ideal short circuit current density (Jph) computed for gradient SC as a function of the thickness of absorber layer (rcover) and the ratio between the array period p and the diameter D of the coated NWs. The 55 data points map has been linear interpolated on a 182 times larger grid.