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CSVT deposited kesterite-Cu_2ZnSnS_4 thin films based solar cells: Effect of the film thickness on the device performance

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ABSTRACT

Cu_2ZnSnS_4 (CZTS) thin films were successfully grown by the close-Space-Vapor Deposition (CSVT) process with the objective of making high quality thin films based solar cells. A simulation is performed to relate the characteristics of the CZTS thin film based photovoltaic solar cell with the material parameters to improve its performance. The numerical model uses the n-CdS/p-CZTS solar cell structure and is based on solving the continuity equation with conditions at the appropriate limits to determine the contributions of the different parts of the cell and then investigate the internal quantum efficiency as function the CZTS absorber layer thickness.

Key words: Cu_2ZnSnS_4, thickness, absorber layer, internal quantum efficiency

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INTRODUCTION

In recent years, the renewable energy sector continues to produce great interest in the minds of researchers; in particular, photovoltaic cell technology continues to grow due to the development of new manufacturing techniques and improvements in the efficiency conversion of solar cells. Nowadays, the solar cell market is still dominated by crystalline silicon which occupies about 80% of the market [1]. The high cost of these solar cells is mainly due to the large use of the material and the manufacturing processes that use complicated and very expensive techniques. Making the production processes simple and cheaper or manufacturing solar cells that consume less material could reduce this cost. One of the alternatives is the thin film solar cell because it consumes less material. The best CdTe and CIGS solar cells reached, at laboratory scale, efficiencies of 22.1% and 22.6%, respectively [2]. However, these thin films based devices contain rare and toxic chemical elements (In, Te, Cd, Ga, Se), which limit the relevance of these two technologies. Faced with these difficulties, measures will have to be taken to counteract the scarcity and the toxicity issues in order to produce low-cost solar cells that respect the environment. Use of the quaternary material made of copper, zinc, tin and sulfur **(**Cu_2ZnSnS_4**)** known as CZTS is one such measure.

With a direct and tunable gap energy of about 1.5 eV [3, 4] and a high absorption coefficient greater than 10^4 cm^{-1} for photon energies over 1eV [5, 6], CZTS is an ideal candidate to be used as an absorber layer in thin film solar cells. The constituent elements of this material namely copper, zinc, tin and sulfur are still available in the earth's crust and are safe for the environment [1]. Several physical and chemical deposition techniques have been used to fabricate CZTS-based solar cells such as sputtering [7, 8, 9, 10, 11], thermal evaporation [12, 13, 14], pulsed laser deposition (PLD) [15, 16, 17], sol gel [18, 19, 20], spray pyrolysis [21, 22, 23, 24], electrodeposition [25, 26, 27], Close Spaced Vapor Transport (CSVT) [28, 29].. The efficiency conversion of this cell has been improved from 0.66% in 1996 [1] to 12.6% by the International Business Machines laboratories in 2014 [10]. Despite the development of different manufacturing techniques and efficiency improvements, our understanding of this type of cell remains relatively limited compared to CIGS and CdTe solar cells [1, 2]. In order to fabricate solar cells with high efficiencies, a thorough study of the synthesis and fabrication mechanism is required as well as an understanding of the solar cell performance. This work focuses on the study of the impact of the absorber layer thickness of the CZTS thin film deposited by CSVT following a mathematical model, the physical and geometrical parameters of the different layers of a CZTS-based thin film photovoltaic solar cell in order to improve the performance of this cell.

EXPERIMENTAL DETAILS AND CALCULATION MODELS OF THE CELL

Experimental Details

The CZTSS thin films used were deposited by the Close Spaced Vapor Transport (CSVT) method [28, 29]. Its main objective is to realize a CZTS compound devoid of any secondary phase in order to improve the conversion efficiencies of CZTS-based photovoltaic cells. For this purpose, the bulk material was first synthesized as an ingot by slow and programmed cooling of a melt obtained from pure elements. The characterizations carried out on the massive material show that it is a single-phase Cu2ZnSnS4 compound, of quasi-stoichiometric composition, in the këwhile structure.

The Cu_2ZnSnS_4 (CZTS) thin films were grown on soda-lime glass substrates by the simple and low-cost Close Spaced Vapor Transport (CSVT) method. Experimental details have been described elsewhere [28, 29]. Films characterizations exhibited the kesterite structure of CZTS layers with a (112) plane preferred orientation [28, 29]. Furthermore, they highlighted a p-type conductivity and proved that control over film growth temperature is a critical factor for obtaining good structural, optical, chemical and electrical CZTS layers. The CZTS films used in this study were deposited with substrate temperature controlled in range from 460 °C – 540 °C [28, 29].

CALCULATION MODELS

To reduce the surface phenomena that often lead to recombination centers, a window layer is often applied to the front surface (illuminated side). This allows to move the surface away from the active area (absorber layer). The lattice parameters being very close, few dislocations or defects exist at the interface between the two materials, resulting in a decrease of the recombination rate at the interface.

The cell structure considered in this study which is schematically shown in figure 1, consists of the following materials: an n-ZnO thin deposit as the window layer, an n-CdS thin film as the buffer layer and a p-CZTS film acting as the absorber layer. Figure 1 illustrates also the three main regions of the ZnO/CdS(n)/CZTS(p)/Mo heterojunction and their respective dimensions. The recombination profile and carrier transport are studied in the one dimension model using the Poisson equation and continuity equations for electrons and holes.

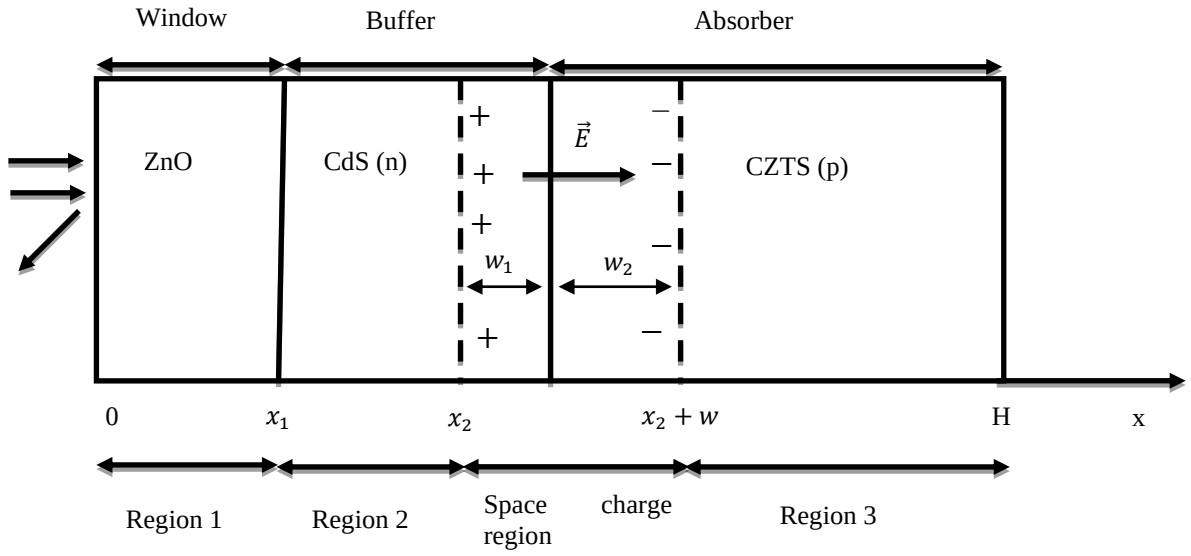

Figure 1: Dimensions and different regions of the ZnO/CdS(n)/CZTS(p)/Mo heterojunction

The transport of the majority charge carriers at the back metal-semiconductor interface is described by the thermionic emission. The transport of minority carriers is described by their surface recombination rates (S_p). Recombination at deep bulk levels and their occupancy are treated according to the Shockley-Read-Hall (SRH) mechanism [31].

Solving the continuity equation combined with the Poisson's equation and the current density equation allows us to calculate the current density in each of the three regions.

For this we considering the generation phenomenon represented by the rate G(λ, x) given by :

$$G(\lambda, x) = \alpha(\lambda)F(\lambda)\big(1 - R(\lambda)\big)e^{-\alpha(\lambda)x} \quad (1)$$

where R(λ) is the fraction of photons reflected from the front surface.

In region 1 (window layer),$0 \leq x \leq x_1$, where the minority carriers are holes, the continuity equation is written:

$$\frac{d^2\Delta p_1}{dx^2} - \frac{\Delta p_1}{L_{p_1}^2} = \frac{-\alpha_1 F(1-R)e^{-\alpha_1 x}}{D_{p_1}} \quad (2)$$

With $L_{p_1}^2 = D_{p_1}\tau_{n_1}$ (3)

and the boundary conditions defined as follows:

$$D_{p_1}\left(\frac{d\Delta p_1}{dx}\right) = S_{p_1}\Delta p_1 \ \ for\ x = 0 \quad (4)$$

$$\Delta n_1 = 0 \ \ for\ x = x_1 \quad (5)$$

The expression of the photocurrent is given by:

$$J_{p_1} = qD_{p_1}\left.\frac{d\Delta p_1}{dx}\right|_{x=x_1}$$

Or

$$J_{p_1} = \frac{q\alpha_1 F(1-R)L_{p_1}}{(\alpha_1^2 {L_{p_1}}^2 - 1)}\left\{\frac{\left(\frac{S_{p_1}L_{p_1}}{D_{p_1}} + \alpha_1 L_{p_1}\right) - e^{-\alpha_1 x_1}\left[\frac{S_{p_1}L_{p_1}}{D_{p_1}}cosh\left(\frac{x_1}{L_{p_1}}\right) + sinh\left(\frac{x_1}{L_{p_1}}\right)\right]}{\frac{S_{p_1}L_{p_1}}{D_{p_1}}sinh\left(\frac{x_1}{L_{p_1}}\right) + cosh\left(\frac{x_1}{L_{p_1}}\right)} - \alpha_1 L_{p_1}e^{-\alpha_1 x_1}\right\} \quad (6)$$

Where L_{p_1} and D_{p_1} are the diffusion length and the diffusion coefficient of holes, respectively S_{p_1} being the hole recombination rate at the ZnO front surface.

In region 2 (buffer layer), $x_1 \le x \le x_2$, the photocurrent is also a hole current, which results from the contribution of regions 1 and 2 taking into account the interface effect characterized by a recombination rate at the interface noted S_{p_2}. The continuity equation is given by:

$$\frac{d^2\Delta p_2}{dx^2} - \frac{\Delta p_2}{{L_{p_2}}^2} = \frac{-\alpha_2 F(1-R)e^{-\alpha_1 x_1}e^{-\alpha_2(x-x_1)}}{D_{p_2}} \quad (7)$$

With ${L_{p_2}}^2 = D_{p_2}\tau_{p_2}$ (8)

and boundary conditions expressed as:

$$D_{p_2}\frac{d\Delta p_2}{dx} = S_{p_2}\Delta p + D_{p_1}\frac{d\Delta p_1}{dx} \ \ for\ x = x_1 \quad (9)$$

$$\Delta n_2 = 0 \ \ for\ x = x_2 \quad (10)$$

The photocurrent obtained by $J_{p_2} = qD_{p_2}\left.\frac{d\Delta p_2}{dx}\right|_{x=x_2}$ has the following expression:

$$J_{p_2} = -\frac{q\alpha_2 F(1-R)L_{p_2}e^{-\alpha_1 x_1}}{\left(\alpha_2{}^2 {L_{p_2}}^2 - 1\right)}$$
$$\left\{\frac{\left(\frac{S_{p_2}L_{p_2}}{D_{p_2}} + \alpha_2 L_{p_2}\right) - e^{-\alpha_2(x_2-x_1)}\left[\frac{S_{p_2}L_{p_2}}{D_{p_2}}cosh\left(\frac{x_2-x_1}{L_{p_2}}\right) + sinh\left(\frac{x_2-x_1}{L_{p_2}}\right)\right]}{\frac{S_{p_2}L_{p_2}}{D_{p_2}}sinh\left(\frac{x_2-x_1}{L_{p_2}}\right) + cosh\left(\frac{x_2-x_1}{L_{p_2}}\right)} - \alpha_2 L_{p_2}e^{-\alpha_2(x_2-x_1)}\right\} +$$
$$\frac{J_{p_1}(x_1)}{\frac{S_{p_2}L_{p_2}}{D_{p_2}}sinh\left(\frac{x_2-x_1}{L_{p_2}}\right) + cosh\left(\frac{x_2-x_1}{L_{p_2}}\right)} \quad (11)$$

In the depletion zone (space charge region), $x_2 \le x \le x_2 + w$, charges recombination is neglected. It is assumed that the transit time of free carriers in this zone is much less than their lowest lifetimes because of the strong electric field there [50]. They will not have time to recombine and then are all collected. The expression of the electron photocurrent is given by:

$$J_{ZCE} = qF(1-R)e^{-\alpha_1 x_1}e^{-\alpha_2(x_2-x_1)}[e^{-\alpha_2 w_1} - 1]$$
$$+qF(1-R)e^{-\alpha_1 x_1}e^{-\alpha_2[(x_2+w_1)-x_1]}[e^{-\alpha_3 w_2} - 1] \quad (12)$$

In region 3 (absorber layer), $x_2 + w \leq x \leq H$, where the minority carriers are electrons, the continuity equation is written:

$$\frac{d^2\Delta n_3}{dx^2} - \frac{\Delta n_3}{L_{n_3}^2} = -\frac{\alpha_3}{D_{n_3}} F(1-R) e^{-\alpha_1 x_1} e^{-\alpha_2[(x_2+w_1)-x_1]} e^{\alpha_3(x_2+w_1)} e^{-\alpha_3 x} \quad (13)$$

With $L_{n_3}^2 = D_{n_3}\tau_{n_3}$ (14)

The boundary conditions are written as follows:

$$D_{n_3} \frac{d\Delta n_3}{dx} = -S_{n_3}\Delta n_3 \ \ for\ x = H \quad (15)$$

$$\Delta p_3 = 0 \ \ for\ x = x_2 + w \quad (16)$$

The electron photocurrent given by $J_{n_3} = -q\, D_{n_3} \frac{d\Delta n_3}{dx}\Big|_{x=x_2+w}$, is the expressed by:

$$J_{n_3} = \frac{q\alpha_3 L_{n_3} F(1-R) e^{[(\alpha_2-\alpha_1)x_1]}\, e^{[(\alpha_3-\alpha_2)(x_2+w_1)]}}{\left(\alpha_3^2 L_{n_3}^2 - 1\right)} \times \left\{ \frac{\left(\alpha_3 L_{n_3} - \frac{S_{n_3}L_{n_3}}{D_{n_3}}\right) e^{-\alpha_3 H} + e^{-\alpha_3(x_2+w)} \left\{ \frac{S_{n_3}L_{n_3}}{D_{n_3}} cosh\left[\frac{H-(x_2+w)}{L_{n_3}}\right] + sinh\left[\frac{H-(x_2+w)}{L_{n_3}}\right] \right\}}{\frac{S_{n_3}L_{n_3}}{D_{n_3}} sinh\left[\frac{H-(x_2+w)}{L_{n_3}}\right] + cosh\left[\frac{H-(x_2+w)}{L_{n_3}}\right]} - \alpha_3 L_{n_3} e^{-\alpha_3(x_2+w)} \right\} \quad (17)$$

The total photocurrent density which is the sum of each of the above calculated current densities is then given by:

$$\mathrm{J_{ph}}(\lambda) = J_{p_1} + J_{p_2} + J_{ZCE} + J_{n_3} \quad (18)$$

For this analytical study, we developed the collection efficiency model for thin-film solar cells and established its mathematical expression. The developed model is applied to determine the internal quantum efficiency which is the ratio of the number of charge carriers collected by the photovoltaic device to the number of photons of a given wavelength or energy on the device [32]. The internal quantum efficiencies are given by:

$$\eta = \frac{\mathrm{J_{ph}}}{\mathrm{qF(1-R)}} \quad (19)$$

$$\eta_1 = \eta(CdS) = \frac{J_{p_2}}{\mathrm{qF(1-R)}} \quad (20)$$

$$\eta_2 = \eta(ZCE) = \frac{J_{ZCE}}{\mathrm{qF(1-R)}} \quad (21)$$

$$\eta_3 = \eta(CZTS) = \frac{J_{n_3}}{\mathrm{qF(1-R)}} \quad (22)$$

Where η is the total internal quantum efficiency, η_1 the emitter or buffer zone (CdS) internal quantum efficiency, η_3 the base internal quantum efficiency (CZTS), and η_2 the space charge zone internal quantum efficiency.

RESULTS AND DISCUSSIONS

In this study a theoretical model is developed to investigate the effects of a CSVT deposited CZTS absorber layer thickness on the internal quantum efficiency of the CZTS-based solar cell.

Figures 2 highlights the contribution of each part of the cell, in the internal quantum efficiency as a function of the photon energy for various thicknesses of the CZTS layer.

We notice that the contribution of the bulk (n-CdS) layer, corresponding to the volume absorptions is less important than those of the bulk absorber layer (p-CZTS) and of the space charge region. The CdS layer thickness being very low, the greatest part of its thickness is located in the space charge layer.

In the range of higher energies (far from the absorption threshold or the gap of the CZTS), the absorption of incident photons is superficial, they are almost absorbed by the front. The surface phenomena become decisive, leading to the fall of the efficiency observed in the different curves.

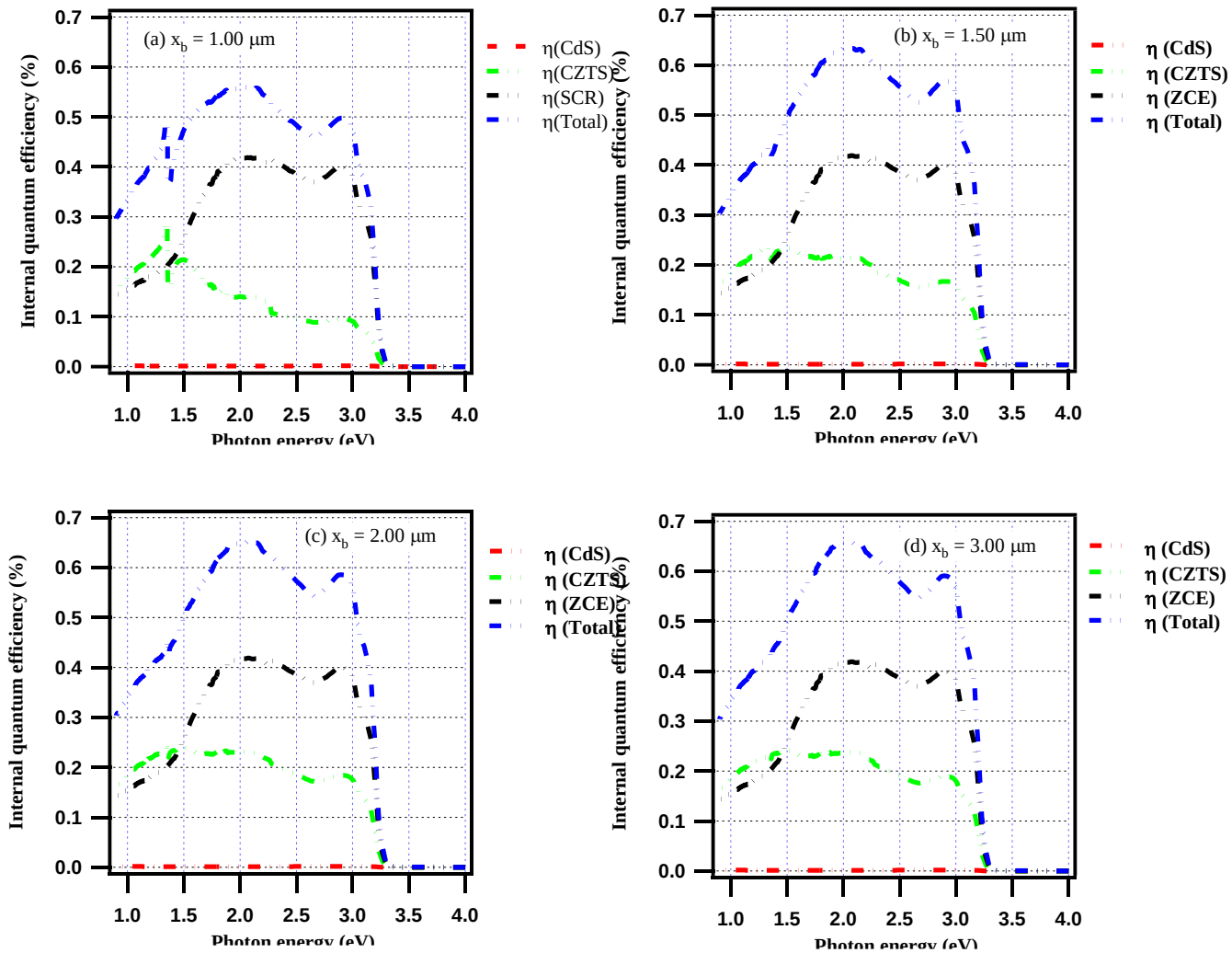

Figure 2: Contribution of the different parts of the n-CdS/p-CZTSS cell

One can note that the contribution of the space charge region does not really change when increasing the base layer. The variation in the quantum efficiency as a function of the thickness of the base layer originates essentially from the contribution of the CZTS bulk layer as can be seen in figure 3.

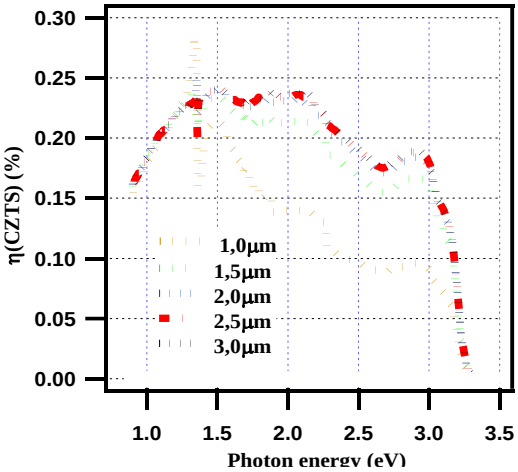

Figure 3: Contribution of the CZTS layer in the internal quantum efficiency as function of photon energy: effect of the thickness.

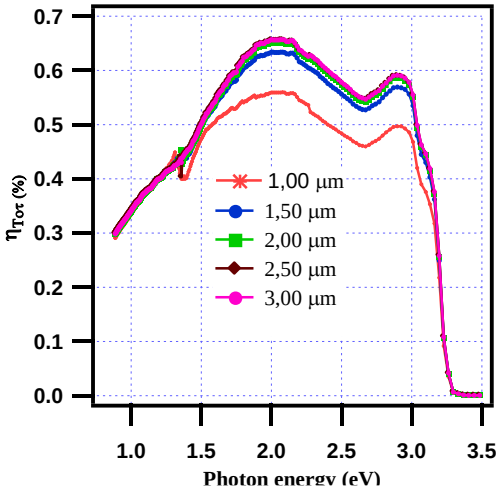

Figure 4: Effect of the thickness in the total internal quantum efficiency of the CdS/CZTS based solar cell.

The absorber layer is the most important component of the solar cell, where incident photons are absorbed and excess carriers are generated. The absorption of photons in a semiconductor material follows the Beer-Lambert law. Therefore, as the thickness of the absorber layer increases, most long wavelength photons (low energy photons) are absorbed, which generates a greater number of excess carriers. This leads to a higher internal quantum efficiency and thus an increase in cell efficiency. In the present study, the effect of absorber layer thickness on the internal quantum efficiency of the cell is simulated by changing the thickness of CZTS from 1 to 3 µm (Figure 3 and 4). The efficiency of the solar cell initially increases with increasing CZTS layer thickness and almost saturates at higher values. This saturation of the efficiency is due to an increased probability of SRH recombination (due to the finite carrier diffusion length) with increasing absorber thickness.

On this graph the internal quantum efficiency drops slightly near 2.4 eV corresponding to the absorption of CdS. We also notice that the signal does not depend any more on the thickness of the absorbing layer xb beyond 2.4 eV, showing that all the incident photons are absorbed by the CdS and the space charge zone (CdS/CZTS). A second drastic drop in efficiency is observed beyond 3 eV, which corresponds to the absorption of photons by the ZnO (Front).

Figure 5 shows the three dimension representation of the internal quantum efficiency of the CdS/CZTS based solar cell. Three-dimensional representation allows to visualize simultaneously the evolution of the internal quantum efficiency as a function of the photon energy and another considered parameter such as in this case, the thickness of the absorber layer.

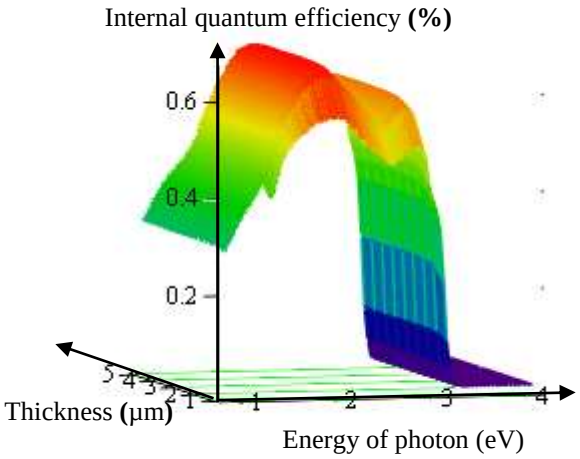

Figure 5: Evolution of internal quantum efficiency as function of photon energy: effect of absorber layer thickness (CZTS)

CONCLUSION

In this work, we studied the internal quantum efficiency of CZTS-based solar cells. We have developed a theoretical model for two layers. We performed a theoretical simulation on all the spectral responses studied by

varying the physical parameters such as the thickness of the absorber layer in order to improve the device performance. We have shown that the internal quantum efficiency increases with the thickness of the absorber layer.

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