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Citation for published version (APA):

Document license:
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DOI:
10.1016/j.solmat.2018.11.002

Document status and date:
Published: 01/03/2019

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Enhancement of the photocurrent and efficiency of CdTe solar cells suppressing the front contact reflection using a highly-resistive ZnO buffer layer


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A R T I C L E   I N F O
Keywords:
ZnO
CdTe
Thin film PV
Anti-reflection
ALD
MOCVD

ABSTRACT

We report on the effects of using an atomic layer deposited ZnO transparent buffer layer with > 10^6 Ω cm resistivity on the performance of CdZnS/CdTe solar cells grown by metalorganic chemical vapour deposition (MOCVD). The buffer film thickness is adjusted by optical modelling to suppress the reflection losses at the front contact. A clear improvement, up to 1.8% in conversion efficiency, was obtained in comparison to reference devices without the ZnO buffer layer, thanks to the enhancement of the current density (Jsc) and fill factor (FF). Device spectral response showed improved collection for most of the visible region. Reflectance measurements confirmed that the ZnO film reduced the optical reflectance around the transparent front contact. This effect permitted light management through the front contact leading to an improvement of the Jsc and hence the photovoltaic conversion efficiency. These results are intriguing since the literature on CdTe solar cells did not previously report improvement to the photocurrent and device response through controlling the highly-resistive transparent buffer layer.

1. Introduction

The interest in cadmium telluride (CdTe) thin film solar cells from both academia and industry has risen significantly due to the recent record device efficiencies of 22.1% for cells and 18.6% for modules [1]. CdTe solar cells produced by metalorganic chemical vapour deposition (MOCVD) has also gone through significant improvements in recent years. High performance cells and mini-modules were reported through controlling the highly-resistive ZnO, ZnS absorber layer. Fedorenko et al. studied ~ 100 nm thick ZnO, ZnSe, and ZnS layer combinations were used [6]. In general, they reported performance boost via the shunt resistance (Rsh) and open circuit potential (Voc) parameters with the inclusion of the HRT layer at the front contact. Spectrally-resolved FF measurements indicated enhanced carrier collection in the long wavelength (500–800 nm) region, which was interpreted to emanate from strengthening of the collection field as well as carrier lifetime in the absorber layer. Ferekides et al. studied the performance of CdTe solar cells for which a variety of TCO [indium-tin-oxide (ITO), SnO2:F, and CdIn2O4] and HRT (SnO2, In2O3, and Zn2SnO4) layer combinations were used [6]. In general, they reported performance boost via the shunt resistance (Rsh) and fill factor (FF) and open circuit potential (Voc) parameters with the inclusion of the HRT layer at the front contact. Spectrally-resolved FF measurements indicated enhanced carrier collection in the long wavelength (500–800 nm) region, which was interpreted to emanate from strengthening of the collection field as well as carrier lifetime in the absorber layer. Ferekides et al. studied ~ 100 nm thick ZnO, ZnSe, and ZnS films as the HRT layer for CdS/CdTe devices [7]. Device efficiency was found to be much poorer when using ZnSe or ZnS HRT films while no specific performance gain was observable for the ZnO HRT case. These results were attributed to inefficient doping and formation of recombination centers within the CdTe absorber, in relation to the extreme thinning of the window layer often leads to poor substrate coverage and hence localized TCO-absorber direct junctions which produces a poor junction [5]. However, contradictory results were obtained as to how much the performance and which device parameters are actually improved with the use of a HRT buffer film. Ferekides et al. studied the performance of CdTe solar cells for which a variety of TCO [indium-tin-oxide (ITO), SnO2:F, and CdIn2O4] and HRT (SnO2, In2O3, and Zn2SnO4) layer combinations were used [6]. In general, they reported performance boost via the shunt resistance (Rsh), fill factor (FF) and open circuit potential (Voc) parameters with the inclusion of the HRT layer at the front contact. Spectrally-resolved FF measurements indicated enhanced carrier collection in the long wavelength (500–800 nm) region, which was interpreted to emanate from strengthening of the collection field as well as carrier lifetime in the absorber layer. Ferekides et al. studied ~ 100 nm thick ZnO, ZnSe, and ZnS films as the HRT layer for CdS/CdTe devices [7]. Device efficiency was found to be much poorer when using ZnSe or ZnS HRT films while no specific performance gain was observable for the ZnO HRT case. These results were attributed to inefficient doping and formation of recombination centers within the CdTe absorber, in relation to the extreme thinning of the window layer often leads to poor substrate coverage and hence localized TCO-absorber direct junctions which produces a poor junction [5]. However, contradictory results were obtained as to how much the performance and which device parameters are actually improved with the use of a HRT buffer film. Ferekides et al. studied the performance of CdTe solar cells for which a variety of TCO [indium-tin-oxide (ITO), SnO2:F, and CdIn2O4] and HRT (SnO2, In2O3, and Zn2SnO4) layer combinations were used [6]. In general, they reported performance boost via the shunt resistance (Rsh), fill factor (FF) and open circuit potential (Voc) parameters with the inclusion of the HRT layer at the front contact. Spectrally-resolved FF measurements indicated enhanced carrier collection in the long wavelength (500–800 nm) region, which was interpreted to emanate from strengthening of the collection field as well as carrier lifetime in the absorber layer. Ferekides et al. studied ~ 100 nm thick ZnO, ZnSe, and ZnS films as the HRT layer for CdS/CdTe devices [7]. Device efficiency was found to be much poorer when using ZnSe or ZnS HRT films while no specific performance gain was observable for the ZnO HRT case. These results were attributed to inefficient doping and formation of recombination centers within the CdTe absorber, in relation to the...
effect of strain on the electronic properties of the grain boundary interface states. Williams et al. reported the effect of ZnO HRT films grown by atomic-layer-deposition (ALD) and sputtering on Cu(In,Ga)Se (CIGS) solar cells employing a CdS window layer [8]. Some ZnO films were treated with an oxygen plasma to increase their electrical resistance. It is found that cells with lower resistivity (as-deposited) HRT performed better than those with a high resistivity (plasma-treated) HRT ZnO film.

More recently, through optical modelling Womack et al. showed the possibility of an anti-reflection (AR) behaviour, observable when using a ZnO or SnO2 buffer layer in CdS/CdTe solar cells [9]. The reflection and transmittance in TCO/HRT/CdS/CdTe stacked thin film device structures were calculated using the transfer matrix method, as function of the HRT and CdS window layer thickness. It was found that due to the low absorption in the ZnO layer it was possible to achieve window/ ZnO layer combinations that provide useful broad anti-reflection effects.

In this study we focus on ZnO thin film material deposited by ALD to study its contribution to CdTe solar cells as a HRT buffer layer which could possibly impart an AR property to the transparent front contact. The choice of ALD emanates from its proven ability to provide uniform, pin-hole free and conformal coatings at relatively low growth temperatures [10–12], which is essential for good coverage of the rough TCO layer without degrading its electrical conductivity. Optical modelling was used to aid the experimental work to suppress the reflection in the front contact region and thereby to boost the photocurrent. We observed a clear enhancement in the performance of all devices fabricated with the designed HRT layer due mainly to the optical gain and shunt resistance it provided. To our knowledge, this is the first experimental report which significantly improved the performance of a CdTe solar cell by reducing the reflection loss due to the front contact, using a suitable HRT buffer. Detailed device characterization and simulation results will be presented to explain the origins of the changes observed in device operation.

2. Experimental

Undoped ZnO buffer layers were deposited onto commercial 5.0 × 2.5 cm² boro-alumino-silicate glass coated with a 150–200 nm indium-tin-oxide (ITO) TCO layer (4–8 Ω/sq sheet resistance with 30 nm SiO₂ underlayer) as well as reference oxygenized silicon substrates by ALD at 120 °C. ZnO film thickness was controlled to be in the region of 50 nm, which was predicted to effectively suppress the optical reflection around the ITO film. The Zn and O precursors used were diethyl-zinc and H₂O₂ respectively. The growth process followed was similar to that in Refs. [8,10], which provided good reproducibility and high electrical resistance. Four-point probe measurements carried out on ZnO films deposited on reference substrates showed an electrical resistivity on the order of 1–5 × 10⁶ Ω.cm. Ellipsometry was measured on reference ZnO films to obtain the refractive index (n, k). It is worth noting that no additional AR coating was applied to the glass/air interface.

Thin film Cdₓ₁₋ₓZnₓS/CdTe:As solar cells were deposited using MOCVD in a horizontally configured growth chamber and H₂ carrier gas at atmospheric pressure. In every deposition run, one uncoated and one ZnO coated ITO substrates were placed side-by-side in the chamber, in order to have a direct comparison of the MOCVD film properties and the solar cell performance. Bare ITO substrates were surface-treated with an O₂ plasma prior to MOCVD which improves lateral uniformity. This treatment process was omitted for the ZnO/ITO substrates due to the strong influence of O₂ plasma on ZnO electrical resistance which tends to increase by several orders of magnitude [8]. Instead, ZnO/ITO substrates were kept sealed after the ALD process and only blown with dry N₂ gas before loading them into the MOCVD chamber. The thickness of Cdₓ₁₋ₓZnₓS window layer (referred as CdZnS window hereafter) was varied between 60 and 150 nm with Zn concentration (x) set to 0.7. Extrinsic As doping was used to obtain CdTe with p-type conductivity, with concentration of ~3 × 10¹⁸ atoms/cm² for the bulk of CdTe absorber (2000 nm) and ~1 × 10¹⁹ atoms/cm² for the heavily-doped CdTe back contact layer (250 nm). The CdCl₂ heat treatment for cell activation was carried out after depositing CdCl₂ at 200 °C and annealing the samples under the H₂ ambiant at 420 °C for 10 min. After cooling the substrates to room temperature and rinsing the excess CdCl₂ using deionised water, a post-deposition anneal was also performed in air ambient at 170 °C for 90 min for further cell activation. Further details on MOCVD growth of individual layers as well as device processing can be found in Refs. [3,13].

Solar cells of 0.5 × 0.5 cm² area were finished by mask-deposition of a ~200 nm gold (Au) film on each sample via thermal evaporation. The current density-voltage (J-V) curves were obtained employing an Abet Technologies Ltd. solar simulator in dark and light conditions. The intensity of the lamp output (AM1.5 solar spectrum) was calibrated using a GaAs reference cell. The device spectral response was characterized by measuring the external quantum efficiency (EQE) on a Bentham PV300 quantum efficiency system without a bias. The response of the system was corrected with respect to the output of a reference crystalline Si photodetector. The concentration and depth profiling of the arsenic dopant in the CdTe film were measured by secondary ion-mass spectroscopy (SIMS) using a Cameca IMS-4f instrument. The primary source was Cs⁺ ions operated at 10 keV energy with 20 nA current. The specimen to be analysed was cleaved (to ~1 × 1 cm² size) from the main sample and then etched in 0.2% bromine in methanol solution in order to reduce the surface roughness and thereby improve the depth resolution. An ion-implanted CdTe:As layer served as the calibration specimen.

3. Optical and device simulations

The reflectance data were modelled using The Essential Macleod, an optical modelling software based on the transfer matrix method [14]. The optical light is introduced through the glass substrate in normal direction. The optical constants (n, k) were taken from the library files available with the software, except for the ZnO layer for which these were taken from ellipsometry data. Due to lack of available data, CdS was selected in place of the CdZnS material due to their material and functional similarity. Thickness of the films involved were kept close to their nominal values with some flexibility considered to account for the experimental variations due to deposition and post-processing. Table 1 provides the film thicknesses and the refractive indices (n, k at 510 nm) used in the optical model.

Device structures were modelled using SCAPS, a one-dimensional thin film solar cell simulation software [19] to investigate the effect of ZnO HRT layer on the performance of CdZnS/CdTe solar cells. Material parameters used in these simulations (mostly taken from previous work [3]) are given in Table 2. The carrier density of the CdTe and ZnO layers

<table>
<thead>
<tr>
<th>Layer</th>
<th>Material</th>
<th></th>
<th></th>
<th>Thickness (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Medium</td>
<td>Glass</td>
<td>1.52083</td>
<td>0.00000</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>SiO₂</td>
<td>1.46180</td>
<td>0.00000</td>
<td>30</td>
</tr>
<tr>
<td>2</td>
<td>ITO</td>
<td>1.94000</td>
<td>0.01000</td>
<td>180</td>
</tr>
<tr>
<td>3</td>
<td>ZnO</td>
<td>2.06286</td>
<td>0.00518</td>
<td>0–50</td>
</tr>
<tr>
<td>4</td>
<td>CdS</td>
<td>2.51992</td>
<td>0.02695</td>
<td>110</td>
</tr>
<tr>
<td>5</td>
<td>CdTe</td>
<td>3.11402</td>
<td>0.49164</td>
<td>2000</td>
</tr>
<tr>
<td>Medium</td>
<td>Air</td>
<td>1.00000</td>
<td>0.00000</td>
<td></td>
</tr>
</tbody>
</table>
as well as device parasitic resistances ($R_{sh}$, $R_s$) were taken directly from the experimental J-V data.

4. Results and discussion

Modelling showed that a ZnO film thickness in the region of 50 nm provided the highest AR effect for an ITO layer of ~180 nm thickness. The simulated reflection spectra with and without a ZnO buffer film of 50 nm in the ITO/HRT/CdS/CdTe device structure is given in Fig. 1. It is noticeable that the broad reflection peak at 625 nm observable for the non-HRT case is suppressed, along with some other gains obtained at other wavelengths, for the HRT device. This demonstrates that a 50 nm ZnO buffer layer can introduce AR to the ITO substrate used in our experiments.

Table 2

<table>
<thead>
<tr>
<th>Parameter</th>
<th>ITO</th>
<th>ZnO</th>
<th>CdZnS</th>
<th>CdTe:As</th>
<th>CdTe: As+</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness (nm)</td>
<td>200</td>
<td>50</td>
<td>150</td>
<td>2000</td>
<td>250</td>
</tr>
<tr>
<td>Bandgap (eV)</td>
<td>3.72</td>
<td>3.37</td>
<td>2.90</td>
<td>2.45</td>
<td>1.435</td>
</tr>
<tr>
<td>Electron affinity (eV)</td>
<td>4.50</td>
<td>4.00</td>
<td>4.26</td>
<td>4.28</td>
<td>4.28</td>
</tr>
<tr>
<td>Dielectric permittivity (relative)</td>
<td>9.4</td>
<td>9.0</td>
<td>9.3</td>
<td>9.4</td>
<td>9.4</td>
</tr>
<tr>
<td>CB density of states (cm$^{-3}$)</td>
<td>$4 \times 10^{19}$</td>
<td>$1.8 \times 10^{19}$</td>
<td>$2.1 \times 10^{18}$</td>
<td>$1.5 \times 10^{18}$</td>
<td>$8 \times 10^{17}$</td>
</tr>
<tr>
<td>VB density of states (cm$^{-3}$)</td>
<td>$1 \times 10^{18}$</td>
<td>$2.4 \times 10^{18}$</td>
<td>$1.7 \times 10^{19}$</td>
<td>$1.8 \times 10^{19}$</td>
<td>$1.8 \times 10^{19}$</td>
</tr>
<tr>
<td>Electron mobility (cm$^2$/Vs)</td>
<td>30</td>
<td>25</td>
<td>70</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>Hole mobility (cm$^2$/Vs)</td>
<td>5</td>
<td>25</td>
<td>20</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>Shallow donor density (cm$^{-3}$)</td>
<td>$1 \times 10^{21}$</td>
<td>$1 \times 10^{19}$</td>
<td>$1.2 \times 10^{18}$</td>
<td>$1 \times 10^{16}$</td>
<td>$2 \times 10^{16}$</td>
</tr>
<tr>
<td>Shallow acceptor density (cm$^{-3}$)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

![Fig. 1. Calculated reflection spectra of a CdTe solar cell structure with and without a 50 nm ZnO HRT layer. Reflection is suppressed at most visible wavelengths with the HRT film, suggesting more light will be transmitted to the absorber.](image1)

![Fig. 2. EQE spectra of CdZnS/CdTe devices with different CdZnS thickness without (a) and with (b) the ZnO HRT layer. The $J_{sc}$ values calculated via spectral integration are also given for each spectrum. Absorption edges due to ZnO, CdZnS, and CdTe are indicated in (b).](image2)
As predicted from optical simulations, increased transmit-tance is likely to result from reduced re-flectance losses around the front contact. To verify the AR effect, reflectance of two devices grown simultaneously, with and without the 50 nm ZnO HRT layer, were measured (Fig. 4a). Compared to the model data (Fig. 1), suppression of the reflectance at most visible wavelengths is evident for the HRT device which demonstrates that the ZnO layer indeed introduced AR to the ITO substrate. In Fig. 4b, the change in the quantum efficiency is compared to that of the optical reflectance for the same devices. Between 400 and 700 nm the spectra are in close agreement, indicating that device spectral response and the Jsc benefit strongly from the AR property in this region. Above 700 nm EQE of the HRT device is markedly superior to that of the non-HRT device, beyond the prediction of the reflectance gain. The additional gain in EQE may result from a change in the electronic properties of the semiconductor films due to a change in the growth substrate, i.e. ITO vs. ZnO/ITO. A small change in the MOCVD kinetics (e.g. due to the deposition substrate) can influence the film composition and properties, especially in terms of dopant.

Table 3
8-cell mean J-V data of CdZnS/CdTe devices with different CdZnS thicknesses. HRT refers to the 50 nm ZnO buffer film between ITO front contact and CdZnS window.

<table>
<thead>
<tr>
<th>HRT</th>
<th>CdZnS (nm)</th>
<th>Voc (mV)</th>
<th>Jsc (mA cm⁻²)</th>
<th>FF (%)</th>
<th>η (%)</th>
<th>Rs (Ω)</th>
<th>Rsh (Ω)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No</td>
<td>60</td>
<td>432 ± 37</td>
<td>23.4 ± 0.9</td>
<td>60.2 ± 2.3</td>
<td>6.1 ± 0.8</td>
<td>3.5 ± 0.3</td>
<td>642 ± 130</td>
</tr>
<tr>
<td>Yes</td>
<td>60</td>
<td>641 ± 21</td>
<td>24.9 ± 0.5</td>
<td>64.0 ± 3.1</td>
<td>10.2 ± 0.7</td>
<td>3.5 ± 0.2</td>
<td>1247 ± 297</td>
</tr>
<tr>
<td>No</td>
<td>100</td>
<td>790 ± 13</td>
<td>21.7 ± 0.4</td>
<td>72.6 ± 4.8</td>
<td>12.5 ± 1.0</td>
<td>3.2 ± 0.4</td>
<td>911 ± 148</td>
</tr>
<tr>
<td>Yes</td>
<td>100</td>
<td>768 ± 11</td>
<td>23.9 ± 0.4</td>
<td>77.3 ± 0.9</td>
<td>14.2 ± 0.5</td>
<td>2.8 ± 0.2</td>
<td>3077 ± 596</td>
</tr>
<tr>
<td>No</td>
<td>150</td>
<td>798 ± 11</td>
<td>21.9 ± 0.6</td>
<td>75.9 ± 2.3</td>
<td>13.3 ± 0.7</td>
<td>3.5 ± 0.2</td>
<td>1979 ± 567</td>
</tr>
<tr>
<td>Yes</td>
<td>150</td>
<td>775 ± 15</td>
<td>23.1 ± 0.5</td>
<td>78.1 ± 1.5</td>
<td>14.0 ± 0.4</td>
<td>2.6 ± 0.2</td>
<td>3127 ± 751</td>
</tr>
</tbody>
</table>

Fig. 3. The light J-V curve (a) and EQE spectrum (b) of the best cells found using 100 nm CdZnS, with and without the HRT layer. The extracted J-V parameters are given as inset to (a).

Fig. 4. (a) Reflectance spectrum of CdTe solar cells grown with and without the ZnO HRT layer, and (b) comparison of the change in reflectance and quantum efficiency for the same cells.

Table 4
Summary of the simulated J-V data for CdZnS/CdTe devices, with and without the 50 nm ZnO HRT buffer layer. Results for equivalent absorber carrier density (N_a = 1 × 10¹⁶ cm⁻³) show no significant V oc change between the two device structures, hinting that the V oc is more sensitive to the absorber carrier density than the band alignment.

<table>
<thead>
<tr>
<th>HRT</th>
<th>N_a (cm⁻³)</th>
<th>V oc (mV)</th>
<th>J sc (mA cm⁻²)</th>
<th>FF (%)</th>
<th>η (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No</td>
<td>1.7 × 10¹⁶</td>
<td>857.20</td>
<td>25.00</td>
<td>73.79</td>
<td>15.24</td>
</tr>
<tr>
<td>No</td>
<td>1.0 × 10¹⁶</td>
<td>834.60</td>
<td>24.24</td>
<td>73.15</td>
<td>14.80</td>
</tr>
<tr>
<td>Yes</td>
<td>1.0 × 10¹⁶</td>
<td>835.30</td>
<td>24.10</td>
<td>75.46</td>
<td>15.19</td>
</tr>
</tbody>
</table>
distribution and carrier density. Such a small difference is indeed observed in the As concentration and consequent p-type doping density in the CdTe absorber (see Fig. 1 in Ref. [20]). Slightly higher bulk recombination at long wavelengths for the non-HRT device may be linked with its higher As density, whereby most As atoms are compensated for due to the low dopant activation ratio (~1%). Finally, the loss of EQE at long wavelengths for the non-HRT device may be linked with the CdS/CdTe solar cells is a new experimental observation.

Here, the small loss of $V_{oc}$ encountered even for 150 nm thick window layer, is likely to arise from (1) the energy band alignment at the material interfaces with the ZnO film incorporated into the device structure [8] or (2) the electronic quality (carrier density and/or lifetime) of the absorber [6,23]. A change particularly in the conduction band offset (CBO) between the absorber/window layers for CdTe solar cells can alter the interfacial surface recombination and hence device parameters [24]. Using SCAPS, we modelled our thin film device structures to examine the band alignment at the absorber/window/front contact interfaces. Although, the insertion of HRT layer produced narrow spikes at the interfaces to CdZnS and ITO (see Fig. 2 in Ref. [20]) these do not seem to lead to a change in the $V_{oc}$ for a constant absorber carrier density of $1 \times 10^{16}$ cm$^{-3}$ (Table 4). However, when the apparent CdTe carrier densities from the C-V data (see Fig. 1 in Ref. [20]) are used in device simulations the $V_{oc}$ of the HRT device is observed to reduce by ~20 mV, which agrees well with the experimental data. Thus, it is sensible to suggest that the $V_{oc}$ of HRT devices can also be optimized by adjusting the p-doping density via the As concentration.

Devices with the thinnest (60 nm) CdZnS window layer showed a significant decrease in the $V_{oc}$ and FF (Table 1). This observation agrees with previous reports where the $V_{oc}$ and FF fall rapidly as the CdS window thickness in CdS/CdTe solar cells is reduced to below 100 nm [4,25,26]. This effect is usually attributed to insufficient substrate coverage by the window layer. Extra-thin regions, or pin-holes, in the window layer would result in weak diodes between CdTe and the TCO layer with an extra-thin film incorporated into the device structure (version 50), Prog. Photovolt.: Res. Appl. 25 (2017) 668–676.


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