Rear Contact Passivation for High Bandgap Cu(In, Ga)Se₂ Solar Cells With a Flat Ga profile

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Abstract—In this study, Cu(In, Ga)Se₂ solar cells with a high bandgap (1.31 eV) and a flat Ga profile ($[Ga]/([Ga] + [In]) \approx 0.60$) were examined. For absorber layer thicknesses varying from 0.60 to 1.45 μm , the Mo rear contact of one set of samples was passivated with an ultrathin (27 nm) Al₂O₃ layer with point contact openings, and compared with reference samples where the rear contact remained unpassivated. For the passivated samples, mainly large gains in the short-circuit current led to an up to 21% (relative) higher power conversion efficiency compared with unpassivated cells. The differences in temperature-dependent current voltage behavior between the passivated and the unpassivated samples and the thin and the thick samples can be explained by an oppositely poled secondary photodiode at the rear contact.

Index Terms—Back contact, CIGS, CIGSe, Cu(In, Ga)Se₂, energy barrier, passivation, rear contact, thin films, two-diode model.

I. INTRODUCTION

RECENTLY, the power conversion efficiency record for Cu(In, Ga)Se₂ (CIGS) solar cells has increased significantly, mostly due to improvements in the absorber quality and the front contact. In addition, thinning down the absorber layer has been explored by different groups with the aim to save materials and to decrease production costs [1]–[8]. To curtail the increasing influence of recombination at the rear contact for thinner absorbers, rear contact passivation has been successfully applied [9], [10]. Thereby, an extra oxide layer (the passivation layer) between the absorber layer and the metal rear contact is introduced. Hole conduction through the electrically insulating passivation layer is achieved by point contact openings. The passivation layer works in two ways, namely through a field effect (favorable band bending of both the conduction and the valence band) and through chemical passivation due to a lower interface trap density [11], [12].

Manuscript received December 22, 2017; revised February 2, 2018 and February 27, 2018; accepted March 2, 2018. Date of publication April 4, 2018; date of current version April 19, 2018. This work was supported in part by the Swedish Energy Agency and in part by the Strategic Research program STandUP. The work of P. Salomé was supported by Fundação para Ciência e Tecnologia through the project IF/00133/2015. (Corresponding author: Dorothea Ledinek.)

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Digital Object Identifier 10.1109/JPHOTOV.2018.2813259

As another method to curb the rear contact recombination, Ga grading with a high Ga content at the rear contact has been successfully applied in state-of-the-art solar cell devices [2], [4]. If Ga replaces In in CuInSe₂, the bandgap increases mostly by elevating the conduction band edge. Thus, by applying a large quantity of Ga at the rear of the solar cell while keeping a low quantity at the front, a conduction band gradient is introduced and acts as a quasi-electrical field for electrons, lowering rear interface recombination. In Ga graded solar cells a Ga content $[Ga]/([Ga] + [In])$ of the order of 0.6 is often used at the rear contact [13]. Thus, a better understanding of both the bulk properties of high Ga CIGS and the passivation of high Ga CIGS has become more urgent.

In previous works, our group studied the passivation of the rear contact interface for absorber layers without Ga grading and with a low Ga content, about $[Ga]/([Ga] + [In]) \approx 0.3$ [9], [10], [14]. As even thin layers of Al₂O₃ hinder Na diffusion from the soda lime glass, an NaF precursor layer was applied to the Al₂O₃ passivation layer before evaporation of the CIGS absorber layer. In this study, however, an ungraded absorber with much higher Ga content of $[Ga]/([Ga] + [In]) \approx 0.60$ was used and NaF was *in situ* evaporated on top of the CIGS layer (postdeposition treatment).

This study adds also to a previous work [15], in which a two-dimensional (2-D) matrix of solar cells with absorber layers with a Ga content $[Ga]/([Ga] + [In])$ of 0.15, 0.30, 0.45, and 0.60 and a nominal thickness of 0.45, 0.85, and 1.45 μm were studied, but which neither had NaF added nor any rear contact passivation. In this previous work, samples with a Ga content of $[Ga]/([Ga] + [In]) \approx 0.60$ showed nearly identical external quantum efficiencies (EQE) curves for CIGS thicknesses of 0.85 and 1.45 μm , even for long wavelengths. In agreement with [16], a short collection length (minority carrier diffusion length plus space charge region) was proposed. Therefore, we were curious if a passivation effect would be observable in the solar cells with a 1.45 μm thick absorber layer. This paper clarifies this and extends an IEEE PVSC 2017 proceedings article [17] by the same authors by including the results of temperature-dependent current–voltage measurements and their interpretation in terms of a secondary photodiode at the rear contact. At low temperatures, the dark saturation current of the secondary solar cell is reduced, so that the temperature-independent secondary photocurrent can be discerned and compared for both the passivated and the unpassivated case and the different absorber layer thicknesses.

II. EXPERIMENTAL

For this study, a 2-D matrix of six CIGS solar cell samples with a nominal Ga content $[Ga]/([Ga] + [In]) \approx 0.60$ was produced. Three samples were passivated and three were nonpassivated reference samples. The intended thickness of the CIGS absorber was 0.45, 0.85, and 1.45 μm for the two cases, respectively.

Soda-lime glass with a size of $5 \times 5 \text{ cm}^2$ was used as a substrate. In each coevaporation run CIGS was deposited on one sample with a passivated and an unpassivated Mo rear contact each, to reach the most similar process conditions possible. Following the group's baseline procedure [18], a CdS buffer layer was deposited from a chemical bath on top of the CIGS, a ZnO/ZnO:Al window layer was sputtered and a front contact grid was evaporated.

For the samples with a passivated rear contact, a 27 nm thick Al_2O_3 passivation layer was deposited onto the Mo rear contact with atomic layer deposition at 300 $^\circ\text{C}$ using water and trimethylaluminum as precursors and nitrogen as purge gas. A quadratic grid of openings, each with a length of 0.7 μm and a width of 0.4 μm and a pitch of 2 μm between the openings was carefully etched into the Al_2O_3 layer by reactive ion etching after defining the pattern using electron beam lithography as in [19].

For all samples, the CIGS absorber layer was coevaporated in a vacuum chamber at a base pressure of $3 \cdot 10^{-6}$ mbar. Constant evaporation rates were used for all elements using a quadrupole mass spectrometer rate control for the metal sources and temperature control for the Se source during the complete evaporation process. Due to the constant evaporation rates, resulting films have no, or very small, elemental gradients. During the first 125 s after the shutter opened, the substrate temperature was 410 $^\circ\text{C}$ and during the following 125 s the temperature was linearly raised to 530 $^\circ\text{C}$ for all samples. For the evaporation of a 500 nm thick absorber layer the substrate temperature was kept constant until the end of the evaporation after 125 more seconds. The evaporation at high temperature was prolonged to achieve 0.85 and 1.45 μm thick absorbers by multiplying the total evaporation time (i.e., all three stages combined) by a factor of 2 or 3, respectively. After the CIGS deposition, all substrates were allowed to cool down to 490 $^\circ\text{C}$ and then kept at that temperature for 400 s during the postdeposition treatment, where 0.45 mg NaF was evaporated. In the last step, 32 solar cells were scribed on each sample.

For every CIGS evaporation process, the composition and thickness of four dedicated test samples with a glass-Mo-CIGS stack were determined by X-ray fluorescence, calibrated against a CIGS sample with known composition, a $[Ga]/([Ga] + [In])$ ratio of 0.40 and a thickness of 1.7 μm as measured by a profilometer. The average CIGS layer thicknesses and standard deviations were $0.60 \pm 0.01 \mu\text{m}$, $0.85 \pm 0.04 \mu\text{m}$, and $1.45 \pm 0.09 \mu\text{m}$, respectively. $[Cu]/([In] + [Ga])$ varied from 0.83 to 0.89 and $[Ga]/([Ga] + [In])$ from 0.55 to 0.61. As the dummy samples were placed furthest out on each side in the deposition zone, these values represent maximum variations. The thickness of the Al_2O_3 passivation layer and the complex

refractive indices of Mo, the Al_2O_3 , and the CIGS were determined by spectroscopic ellipsometry on different sample stacks with a Woollam VASE instrument. The obtained refractive index and absorption coefficient were used to calculate the light intensity reaching the rear contact at different wavelengths using the transfer matrix method. The reflectance at the unpassivated and passivated rear contact was also calculated. The reflectance of the whole sample stacks was measured using a Perkin Elmer L900 spectrophotometer with an integrating sphere. The diameter and depth of the passivation layer contact openings were determined by atomic force microscopy in tapping mode to control that the etching process was complete.

The current voltage (*JV*) curves for all cells were measured in the dark and under a halogen lamp equipped with a cold mirror with a four point probe. The light intensity was calibrated with a Si solar cell to obtain a photon flux corresponding to 1000 W/m^2 at AM 1.45. The temperature during the measurements was set to 25 $^\circ\text{C}$ by a temperature stage cooled using a Peltier element. EQE were measured on two to three representative cells per sample under ambient light. For the calculation of the short-circuit current J_{SC} and the efficiency of all cells on a sample, J_{SC} from the *JV* measurements was corrected for the spectral mismatch by the arithmetic average of the quotient of the J_{SC} from the *JV* measurements and the EQE measurements on that sample. The bandgap E_g was found by linear regression to the energy axis in a plot of the square of the EQE against energy and by the empirical formula $E_g = 1.010 + 0.626x - 0.167x(1-x) \text{ eV}$ [20]. According to the linear regression, the bandgap is $1.31 \pm 0.01 \text{ eV}$. The empirical formula gives a bandgap between 1.27 and 1.33 eV.

Current–voltage curves at 1%, 10%, and 100% photon flux from a white light-emitting diode were measured in a cryostat at substrate temperatures between 100 and 330 K with temperature steps of 10 K between the measurement blocks (later referred to as *JVT* measurements). The current at 100% light intensity was calibrated so that the magnitude of the short-circuit current is approximately the same as in the previous *JV* measurements.

III. RESULTS AND DISCUSSIONS

A. *JV* and EQE Measurements

The arithmetic average and the standard deviation of the four standard solar cell parameters are given in Table I for the eight best cells for all samples, except for the thinnest, passivated sample. For this sample, the CIGS layer peeled from the Al_2O_3 layer and only 18 cells could be used for the analysis. Therefore, the average and the standard deviation were calculated only for the seven best cells instead of eight. As the peeling continued over time and the current deteriorated, no EQE measurements are shown for this sample. The *JV* curves for the best solar cell of every sample can be seen in Fig. 1.

Mainly due to an increased short-circuit current, the thin sample gained the most in efficiency by passivation, namely 21% (relative), while the medium thick sample gained 13% (relative) and the thick one 14% (relative) compared with the respective reference samples. No roll-over in the current–voltage curves was observed at room temperature which is an indication that

TABLE I
SUMMARY OF SOLAR CELL PARAMETERS (ARITHMETIC AVERAGE $\pm$ STANDARD DEVIATION)

Absorber thickness in μm	Passivation	V_{oc} in V	J_{sc} in mA/cm^2	Fill factor in %	Efficiency in %
0.60	yes	0.732 ± 0.003	18.8 ± 0.45	68 ± 3	9.3 ± 0.45
0.60	no	0.709 ± 0.004	15.6 ± 0.2	69 ± 1	7.7 ± 0.2
0.85	yes	0.721 ± 0.010	20.0 ± 0.2	69 ± 0	9.9 ± 0.1
0.85	no	0.727 ± 0.003	17.6 ± 0.2	69 ± 1	8.8 ± 0.1
1.45	yes	0.747 ± 0.004	22.9 ± 0.7	70 ± 1	12.0 ± 0.4
1.45	no	0.718 ± 0.010	20.7 ± 0.3	71 ± 1	10.5 ± 0.1

Fig. 1. Current-voltage curves of the best cell on each sample for the samples with a (a) 0.60 μm , (b) 0.85 μm , and (c) 1.45 μm thick absorber layer.

the samples have a good supply of Na [9], [21]–[25]. The fill factors for all samples are about 70% and the differences between the passivated and unpassivated samples are not significant, considering the standard deviation, showing that the point contact structure is designed well enough to allow for electrical contacting.

The short-circuit current J_{sc} and the EQE increase with the thickness of the absorber layer. Even for the samples with a 1.45 μm absorber layer, the J_{sc} is about 3 mA/cm^2 higher compared with the samples with a 0.85 μm absorber layer. These increases in EQE and J_{sc} for thicker samples can be explained by a long collection length (i.e., a long minority carrier diffusion length and/or a large space charge region), which makes the collection of additionally generated carriers in the back of the thick samples possible. The increase in EQE when the thickness increases from 0.85 to 1.45 μm contrasts with the previously produced unpassivated samples [15], that were not postdeposition treated. For those samples, the J_{sc} and EQE decreased even slightly for the sample with a 1.45 μm thick absorber layer compared with the one with a 0.85 μm absorber layer. We thus conclude that the samples for the previous paper and the samples for this paper are not comparable. Changing the method of NaF application from a postdeposition treatment to a precursor layer before CIGS evaporation is probably of significant importance to the optoelectronic behavior of the high Ga devices.

The EQE between 530 and 800 nm wavelength (see Fig. 2) is significantly higher for the passivated samples compared with the unpassivated ones for all investigated thicknesses. This difference does probably not only stem from optical effects, since

1) as seen from Fig. 2, the difference in reflectance at the whole sample stack of the passivated and unpassivated samples can only explain a part of the difference in EQE for the 0.85 μm sample and almost nothing for the 1.45 μm sample;

2) according to the absorption coefficient from the ellipsometry measurements, less than 5% of the incoming light with wavelengths below 800 nm is expected to reach the rear contact for the samples with 0.85 and 1.45 μm thick absorber layers;

3) More detailed transfer matrix calculations based on the spectroscopic ellipsometry analysis show that the optical absorption in the CIGS layer varies on the order of 0.1 mA/cm^2 with and without the 27 nm thick passivation layer, whereas the observed photocurrent difference is in the order of 1 mA/cm^2 .

The V_{oc} is higher for the passivated samples than for the unpassivated samples with a 0.6 μm (+23 mV) and a 1.45 μm (+29 mV) thick absorber, but is unaffected within the error margins for the samples with a 0.85 μm thick absorber.

Fig. 2. EQE for two devices per sample and $1-R$ (1-reflectance) of the complete solar cell stack for the samples with a (a) $0.85 \mu\text{m}$ and (b) $1.45 \mu\text{m}$ thick absorber layer.

If we assume that the front interface recombination is similar for all devices, then, differences in the depletion region width of the main diode and bulk recombination due to differences in Na doping and effective rear contact recombination will have to account for all observed differences in J_{SC} and V_{OC} . Even though all samples were equally Na postdeposition treated, the effective doping in the passivated samples might be lower than in the unpassivated ones, as the passivation layer blocks Na diffusion. This possible difference in effective doping would give a wider depletion region and thereby a higher collection length and thus photocurrent for the passivated samples—in agreement with our results. However, it would also decrease the V_{OC} , in contrast to our observation. The passivation effects might therefore compensate for this decrease in V_{OC} .

Without modeling, the task of distinguishing between all these effects is difficult, but there is a strong indication that rear contact recombination plays a role for the V_{OC} differences between the passivated and unpassivated devices even for the thickest sample. Thereby, we need to assume that rear contact recombination is not only a local effect at the rear contact, but an effect of band bending, that extends further into the film.

B. JVT Measurements

In Fig. 3, current voltage measurements of the samples measured at 100 K are shown. The measurements were performed in darkness and for three different illumination intensities. We

Fig. 3. Current-voltage curves measured at a temperature $T = 100 \text{ K}$ for 0%, 1%, 10%, and 100% photon flux for samples with a (a) $0.60 \mu\text{m}$, (b) $0.85 \mu\text{m}$, and (c) $1.45 \mu\text{m}$ absorber layer. The left chart shows the curves for the passivated sample, the right chart shows the curves for the unpassivated reference sample. To be able to depict the currents for the different photon fluxes on one axis, the measured currents have been multiplied with a factor of 10 for the measurements at 10% photon flux and with a factor 100 for the measurements at 1% photon flux.

observe for all samples in this study that for low temperatures the forward current is limited at forward biases larger than V_{OC} , i.e., the JV curves have a roll-over. The magnitude of the roll-over current is—as expected for a photocurrent—larger for higher photon fluxes. It is also larger for the unpassivated samples as compared with the passivated samples. For the unpassivated samples it increases with reduced absorber layer thickness, but not so for the passivated samples. At even higher voltages and for low photon fluxes, the slopes of the current curves increase again for most samples. The dark current is also severely limited for voltages over V_{OC} at low temperatures. When plotting the V_{OC} as a function of temperature for the different illumination intensities, V_{OC} saturates at low temperatures for all illumination intensities, but at different values. The V_{OC} for small light fluxes is always lower than the V_{OC} for larger light fluxes at the same temperature—in agreement with the superposition principle [26] but disagreement with the phototransistor model as presented in [27]–[29], which assumes equal ideality factors for the main diode and the diode at the rear contact and neglects shunt resistances in parallel to the diodes. Furthermore, the roll-over currents are so small that such a phototransistor would have a current gain $\ll 1$, in contrast to the current gains around 1 observed by Ott *et al.* that could not be explained by a secondary photocurrent and made the introduction of the phototransistor model necessary in the first place [28].

To explain these results we propose an energy barrier at the rear contact, without making any definite statement neither about the exact location of this barrier nor about the exact current transport mechanisms. This energy barrier and the associated currents can be described by a secondary photodiode and three current components over this energy barrier can be distinguished.

- 1) An exponentially temperature-dependent dark saturation current consisting of holes injected over the rear barrier from Mo into CIGS. At room temperature this current is several orders of magnitude larger than the photocurrent from the main junction, so that the current is not limited and no roll-over occurs. By measuring at low temperatures (100 K), this current component can be reduced to about the same level as the secondary photocurrent at 1% light intensity, so that the secondary photocurrent can be discerned.
- 2) For high enough voltages over the rear contact barrier the dark saturation current increases due to tunneling or a breakthrough. In our model the junction at the rear contact is reverse biased in the first quadrant. Thus, the applied voltage is divided into a voltage over the forward biased main junction and a voltage over the reverse biased rear junction, where the latter limits the current flow through the whole device. For high enough voltages over the rear contact barrier, the dark saturation current increases due to tunneling or a breakthrough. This is visible in the dark current curves measured at 100 K as a non-ohmic behavior and in the curve measured at 1% light intensity as an increase in the current slope at high voltages.
- 3) A temperature-independent secondary photocurrent, consisting of electrons that diffuse into the depletion zone of the secondary junction and are extracted into the rear contact. At low temperatures, this current dominates the roll-over current for the unpassivated samples. As a photocurrent, it depends on the amount of photons absorbed within the space charge width and the collection length of the secondary junction, and thereby on the incident primary light flux on the solar cell, the absorption profile, and the absorber layer thickness. With decreasing absorber thickness more electrons are generated close to the rear contact and thus have a higher probability of reaching the rear contact and contributing to the secondary photocurrent. For the passivated samples, the fixed charges in the passivation layer reduce the field of the secondary junction. Thereby, the secondary photocurrent and thus the roll-over current decrease and become independent of the absorber layer thickness, and the primary photocurrent increases. In contrast to the unpassivated case, the hole injection current is a significant part of the roll-over current even at low temperatures. Thus, by comparing passivated and unpassivated samples and different absorber thicknesses, the secondary photocurrent can be discerned. As the secondary photocurrent is temperature independent, we can conclude that for unpassivated samples J_{SC} and V_{OC} are reduced due to the rear contact even for these CIGS solar cells, with a high bandgap and a flat Ga profile.

TABLE II
EXTRACTED PARAMETERS FROM THE TEMPERATURE-DEPENDENT
CURRENT-VOLTAGE MEASUREMENTS

Thickness in μm	Passivation	φ_H in eV	φ_L in eV	$\Delta\varphi$ in eV
0.45	yes	1.37	0.97	0.40
0.45	no	1.36	0.98	0.38
0.85	yes	1.25	0.96	0.29
0.85	no	1.46	1.00	0.46
1.45	yes	1.27	0.97	0.29
1.45	no	1.31	1.05	0.26

The secondary diode's saturation current $J_{0\text{sec}}$ is determined by a prefactor $J_{00\text{sec}}$ and a Boltzmann factor $e^{-\Delta\varphi/kT}$. While some [27], [28], [31] have interpreted $\Delta\varphi$ as the effective Schottky barrier height at the rear contact, we are not that specific and more generally interpret $\Delta\varphi$ as the height of a general barrier at the rear contact. For sufficiently small barrier heights, the secondary diode does not limit the current and voltage at room temperature but only at low temperatures. It is then possible to determine $\Delta\varphi$ as the difference $\Delta\varphi = \varphi_H - \varphi_L$ between the extrapolated V_{OC} at 0 K for a linear fit of the high (φ_H) and low (φ_L) temperature regions of the V_{OC} over T curve.

For our JVT measurement results at 100% light flux, no general trend for $\Delta\varphi$ and thus for the saturation current of the secondary diode was observed (compare Table II). This lack of a trend means that the band diagram of the contact for the hole current is not observably influenced by the electrical field stemming from the oxide charge in the passivation layer. This can be explained in two ways: Either the passivation layer does not induce an additional electrical field and the passivation layer only passivates chemically or this electrical field does not spread far enough laterally into the contacts. We assume here that the electrical field spreads both longitudinally into the absorber layer, but also laterally into the CIGS at the point contacts and thus to the CIGS–MoSe₂ interface. As the rear contact barrier height remains unchanged in the experiment, there is probably a path for the holes in the middle of the point contacts through the CIGS–MoSe₂ interface that lies outside of the region affected by the field of the fixed oxide charges. As the smallest dimension of the contact opening is 0.4 μm , the width of the field charge region has to be smaller than half of that value, namely 0.2 μm . Only charge carriers within that distance or less from the rear contact are affected by the additional electrical field.

IV. CONCLUSION

In this paper, a comparison of rear surface passivated samples using Al₂O₃ with hole contact openings and unpassivated reference samples of CIGS solar cells with absorbers with a Ga content of $[Ga]/([Ga]+[In]) \approx 0.6$ and a flat Ga profile has been made. Absorbers with three different thicknesses of 0.60, 0.85, and 1.45 μm were compared. We observe a considerable improvement in the short-circuit current with the passivation layer and a slight improvement in voltage in the case of 0.6 and 1.45 μm thick absorbers. The thinnest sample gained most by passivation, with a relative increase in efficiency of 21%

(relative). The relative gains were 13% (relative) and 14% for the medium and thickest samples, respectively. We also note that a 0.6 μm absorber with low In content and with simple flat profiles achieved a power conversion efficiency value close to 10%, which is an important result for the long-term strategy of chalcopyrite technology to lower the usage of rare In.

The enhanced photocurrent with the passivation layer can only to a small extent be accounted for by optical effects. Rather, our data suggest it to be the result of improved current collection, which probably occurs due to both—an increased depletion region width and reduced rear contact recombination for the passivated samples. In order to explain both the low EQE response for the unpassivated samples in the IR region and the higher voltage for the passivated samples with 1.45 μm thick CIGS, rear contact recombination is most probably not only a local effect at the rear contact, but is the result of band bending that extends into the bulk of the CIGS film.

The temperature, light, and absorber thickness dependence of the *JV* behavior of both the passivated and unpassivated solar cells can be described with a model that postulates a secondary, oppositely poled photodiode at the rear contact. In this model, the passivation layer lowers the secondary photocurrent and increases the primary photocurrent by inhibiting the secondary diode's field at the rear contact. This effect has been observed by measuring *JV* curves at low temperatures (100 K), which reduce the exponentially temperature-dependent dark saturation current of the secondary solar cell to about 1% of the secondary light current at 100% photon flux. The dependence of the currents at the secondary junction on the light flux, the absorber layer thickness, and the existence of a passivation layer, indicate that the secondary photojunction is indeed located at the rear contact.

ACKNOWLEDGMENT

The authors are grateful for input from the colleagues in the Thin Film Solar Cell group at the Ångström Laboratory, especially P. Szaniawski, who introduced D. Ledinek to the JVT measurement set-up.

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