Increased performance of thin-film GaAs solar cells by rear contact/mirror patterning

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ABSTRACT

Current thin-film GaAs solar cells typically have a completely metal covered rear side that functions as back contact, rear mirror and stable carrier to the solar cell. However, the bottom contact layer, that typically remains whole in this configuration, absorbs a large amount of photons that would otherwise be reflected back into the cell structure. This work identifies and quantifies the performance limits by non-optimal rear contact reflectance in the standard configuration of thin-film GaAs solar cells and proposes a design solution. In this study we show that the total reflectance at the back of the solar cell is increased from 63.5% to 93.2% by partially etching the absorbing GaAs contact layer and applying a dielectric material in the etched regions. Both shallow junction and deep junction cells were produced, and for deep junction solar cells the patterning resulted in a more than linear increase in the open circuit voltage and a linear increase in the short circuit current, which is in good agreement with previously reported numerical models.

1. Introduction

Most III–V materials used for solar cells, such as GaAs and InGaP, are semiconductors with direct bandgap and high absorption coefficients, therefore only few micrometers thick devices are required to absorb all the incident light. However, in order to obtain good quality single crystal solar cell structures, the materials have to be grown epitaxially on top of a GaAs or Ge wafer, with thicknesses that typically vary from 100 μm to 650 μm . After the growth, the wafer is of no value for the cell performance and it can be removed from the growth structure without any detrimental effect on the efficiency.

Methods to remove the growth template wafer of GaAs solar cells, such as epitaxial lift-off (ELO), produce as final result thin-film devices that can be semitransparent or, more commonly, mounted on a metal foil that works as stable flexible handler and rear contact [1–11]. In the latter configuration, the rear contact also acts as mirror to the incident light, enabling cells to have even smaller thicknesses (of around 2 μm). This thin-film geometry is superior to those of cells processed on the native wafer and have reached higher world record efficiencies [12].

It has been demonstrated that GaAs solar cells can potentially operate close the Shockley-Queisser efficiency limit [13–16] mainly by optimizing the optical advantages of the thin-film geometry. These advantages hold large promises for the applicability of these devices in space applications [17], concentrating solar systems and in combination with third generation photovoltaics.

In thin film-configurations, the production costs can be reduced with wafer reuse. Another major contribution to the cost is the growth of epitaxial structures, and this can be diminished with the production of thinner devices. For ultrathin cells (with few hundred nanometers of thickness), however, not all the incident light can be absorbed and even though these devices can be rapidly grown, it requires the application of nanostructures in the front and rear side. But to date the efficiencies achieved with solar cells of a few hundred nanometers in thickness are still relatively low (below 17%) [18–21]. This indicates that the effective increase in the optical path length of the light captured by the use of gratings and nanostructures has not yet been sufficiently implemented to compensate for drastic reductions in thickness.

The standard design of GaAs solar cells consists of a thin n-doped emitter and thicker p-doped base, here called a shallow junction (SJ) solar cell. With high quality materials, however, the minority carrier diffusion length of the emitter is improved. This allows the production of deep junction (DJ) devices, with thick emitter and thin base, that are able to achieve higher performances, as they operate mainly in the radiative recombination regime at the maximum power point [22]. As the current world-record efficiency of 28.8% for a single junction thin-film GaAs solar cell was achieved in 2012 [12] and has not been surpassed to date, these cells are clearly approaching the limit in material quality. In these cases, radiative recombination becomes the dominant loss mechanism [23–25], and photon recycling has been indicated to be the most important process in order to achieve higher V_{oc} values in

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Fig. 1. Schematic representation of the a) shallow junction (SJ) and b) deep junction (DJ) solar cells processed in this study (not to scale).

previous world record cells [26]. Therefore, confining reemitted photons inside the active layers of the cell becomes fundamental in order to achieve efficiencies closer to the theoretical limit [15,16,27–29].

In the present study we evaluate the behavior of emitted photons in the rear layers of GaAs thin-film solar cells and the role of the rear contact design on the reflectance and photon recycling. Devices with both shallow junction and deep junction configuration were produced with increased reflectance of the rear surfaces, and the effects on the cell key parameters from these changes in the rear contact layout were evaluated.

2. Methods

The solar cells used in this study were grown using low-pressure MOCVD on 2 in. diameter GaAs wafers with (1 0 0) 2° off to [1 1 0] orientation. The grown cell structures are shown in Fig. 1. In this study the wafer was removed by chemical etching with an aqueous citric acid and hydrogen peroxide solution (5:1 in volume), instead of using the more elaborative epitaxial lift-off method. In order to control the fragile thin-film single crystal structures, the front surface of the solar cells were mount to a etch-resistant temporary plastic carrier. To stop the process after etching of the wafer, a thin AllnGaP etch-stop layer was grown before the solar cell structure. Subsequently, the etch-stop layer was removed in a 37% HCl solution.

Using photolithography a pattern of disk-shaped contact areas was created in the GaAs rear contact layer. In between these contact areas with diameters ranging from 260 μm to 580 μm the p-GaAs contact layer was etched away using an $\text{NH}_4\text{:H}_2\text{O}_2\text{:H}_2\text{O}$ (1:2:10) solution, resulting in cells with four different rear surface contact area coverages (C_r) of 100%, 50%, 30% and 10%. Next, a 60 nm thick ZnS insulating layer was deposited by thermal evaporation before removal of the photoresist, so that finally the ZnS covers only the etched regions. Then a 200 nm Au layer was deposited by e-beam evaporation throughout the entire rear surface to promote ohmic contact at the contact areas and simultaneously act as a high quality reflector at the rest of the area, followed by a thick electroplated Cu layer, to facilitate handling of the sample.

The temporary plastic carrier was removed from the front surface

and a 200 nm thick Au front grid contact was deposit by e-beam evaporation. For convenience during processing and cell analysis, a grid pattern with a relatively large front surface coverage (C_f) of 16.6% was applied for most cells. One or two cells in each configuration were created with a square frame-like contact pattern and without grid fingers, which is ideal for optical analysis. The n-contact layer was removed in between the metal contact areas using an $\text{NH}_4\text{:H}_2\text{O}_2\text{:H}_2\text{O}$ (2:1:10) solution and the 25 mm^2 cells were defined by a mesa-etch with a $\text{HBr}:\text{Br}_2\text{:H}_2\text{O}$ (100:1:200) solution for phosphide layers and an $\text{NH}_4\text{:H}_2\text{O}_2\text{:H}_2\text{O}$ (1:2:10) solution for GaAs layers. Finally, an anti-reflection coating consisting of 44 nm of ZnS and 94 nm of MgF_2 was deposited by thermal evaporation. The resulting thin-film with multiple solar cells as well as a schematic depiction of the pattern applied to the contact layer are shown in Fig. 2. In contrast to the processing of wafer based cells, the processing of thin-film geometry is less well developed, but for each C_r value (i.e. from each quarter of the thin-film) 3 to 5 operating cells were obtained.

I–V characterization of the solar cells is performed using an ABET Technologies Sun 2000 Class AAA solar simulator, which provides a uniform illumination resembling the AM1.5G spectrum, over a $100 \times 100 \text{ mm}^2$ area, with a maximum angular off set of 2° . The setup is equipped with a Keithley 2601B source meter and data acquisition is performed using ReRa Tracer3 software. The solar cells are kept at 25°C during measurement using a heating/cooling water thermostat and Pt100 temperature sensing.

The setup is calibrated using an NREL calibrated reference cell before each measurement series. Using this approach the cell parameters can be retrieved from the I–V curves with a systematic error of $\pm 3 \text{ mV}$ in V_{oc} , $\pm 0.8 \text{ mA/cm}^2$ in J_{sc} and ± 0.005 in FF compared to an absolute reference. In mutual comparison the trends in the performance of cells as a consequence of deliberate changes in the cell geometry can be identified with an accuracy of $\pm 2 \text{ mV}$ in V_{oc} , $\pm 0.06 \text{ mA/cm}^2$ in J_{sc} and ± 0.003 in FF.

External Quantum Efficiency (EQE) measurements are performed with a ReRa SpeQuest Quantum Efficiency system. The system uses both a Xenon and Halogen light source to address all wavelengths present in the solar spectrum. A monochromator is used to generate quasi-monochromatic light and a chopper for intensity modulation.

Fig. 2. a) Completely processed 2 in. diameter thin-film GaAs cell structure containing multiple $5 \times 5 \text{ mm}^2$ solar cells with a different rear contact coverage in each quadrant. Note that each quadrant also has one or two devices with a frame-like front contact without grid fingers to facilitate optical measurements. b) Schematic representation of the rear contact pattern design (not to scale).

This generates a test light of variable wavelength while a continuous bias light and bias voltage is used to put the cell under test in operating conditions.

The reflectance calculations were made using the transfer matrix method considering only normal incidence of photons. The required n and k values for GaAs and Au were retrieved from the SOPRA database [30]. For ZnS, MgF_2 and InGaP these data were retrieved from ellipsometry analyses of test samples (typically 100 nm of the material on a GaAs substrate) that were prepared under the same conditions as the corresponding layers in our solar cell structures. Reflectance measurements were performed with a FilMetrics spectrophotometer perpendicular to the analyzed surfaces, using a gold mirror for calibration.

3. Results and discussion

3.1. Photon absorption in the GaAs back contact layer

In order to create good ohmic contacts, the first and last grown layers in thin-film GaAs solar cells are usually highly doped GaAs contact layers. At the front side of the cell this layer is removed in between the grid fingers, but for the rear contact, it typically remains whole. Even though the contact layer is relatively thin (100 nm), a relevant fraction of available photons is absorbed in it. This is because photons emitted in the active layers of the cell as a result of band-to-band recombination are repeatedly reflected inside the cell structure and pass through the contact layer several times. Fig. 3 depicts the calculated reflectance at the rear side interfaces of the thin film cell ($R_{\text{GaAs/Au}}$) considering normal incidence of photons. At room temperature, band-to-band emissions from GaAs peak at around 870 nm (the band-gap wavelength), and the full-width at half maximum (FWHM) of the emission line is 28 nm. The emissions from GaAs cease at the band-gap, so the reflectance should be maximized in the region between 870 nm and 840 nm [31]. At these wavelengths, the calculations show a large absorption ($> 30\%$) of generated photons, with an average reflectance of 63.5% in this region. In a structure without a GaAs contact layer, the reflectance (R_{Au}) could be increased to approximately 94%.

Absorbed photons in the non-active layers of the cell are lost as they do not contribute to the cell performance. Therefore, it will be beneficial to partially remove the rear contact layer, analogous to the procedure applied at the front side. If a transparent insulator material is deposited in the areas where the contact layer is removed and Au remains deposited continuously at the rear surface, at the etched regions the Au layer will function only as a mirror. The benefit of the intermediate layer is that a certain fraction of the light is already reflected at

the semiconductor-dielectric interface before reaching the gold interface that because of its higher extinction coefficient does represent some absorption losses. The thickness of the dielectric layer can be tuned in such a way that in the relevant wavelength range corresponding to the GaAs bandgap in the fraction reflected at the gold interface is minimal due to destructive interference.

The materials considered for the transparent insulator layers were ZnS and MgF_2 , both with a refractive index in between that of InGaP ($n = 3.2$) and gold ($n = 0.2$) at the 870 nm wavelength region. Fig. 4b) and c) shows the wavelength dependent reflectance following from simulations performed using the layer structure in Fig. 4a, with the dielectric materials instead of the contact layer. The simulations show an increase in reflectance if either ZnS or MgF_2 is applied before the Au layer as compared to Au directly applied to the back surface field. According to these simulations, the use of MgF_2 will result in the highest reflectance. In practice, however, the adhesion from the thermally evaporated MgF_2 to the semiconductor material was found to be problematic, as it easily delaminates. Therefore, ZnS was the chosen material for further studies.

In the first place the role of the rear mirror in thin-film solar cells is to reflect the incident light that is not absorbed in the first pass. This enhances overall absorption and increases the short-circuit current density (J_{sc}) or alternatively allows to reduce the active cell layer thickness without loss in J_{sc} . GaAs is a good absorbing material, and in the produced $2 \mu\text{m}$ thick solar cells most of short wavelength light is absorbed in the first pass through the active layers, and the rear mirror reflectance is particularly important for wavelengths above 700 nm. In the second place the rear mirror is important to confine photons that are emitted upon radiative carrier recombination with wavelengths between 840 nm and 870 nm, so they can be reabsorbed in the active layers of the structure. This increase in the photon recycling will result in an increase in the open circuit voltage (V_{oc}) [14,16].

Experimental measurements were made to confirm the increase in reflectance obtained by the deposition of ZnS. For this purpose, $650 \mu\text{m}$ thick GaAs wafers polished on both sides were used. An ARC optimized for longer wavelengths was deposited on the front surface and different thicknesses of ZnS ranging from 0 to 100 nm were evaporated on the rear surface, which were subsequently covered with 200 nm of Au. These structures and the obtained reflectance curves are depicted in Fig. 5. Because of the long optical path that the light has to travel in the GaAs wafer, it is not possible to measure the reflectance mechanisms in the bandgap region. However, a comparative analysis of the different interfaces can be done by observing the reflectances at slightly longer wavelengths. Even though parasitic sub-bandgap absorption in the

Fig. 3. Relevant structure used to simulate the reflectance of photons emitted from the active GaAs layer (n_0) at the rear surface of a) a standard thin-film GaAs solar cell structure ($C_r = 100\%$), b) the same structure without the contact layer ($C_r = 0$), and c) the calculated wavelength dependent reflectance at these structures, considering normal incidence of photons.

650 μm thick GaAs substrate cause the absolute reflectance values to be different from the simulations, at around 1050 nm both simulated and measured curves show similar patterns, confirming the increase in reflectance caused by the deposition of ZnS.

The deposition of 100 nm of ZnS would result in highest reflectance at wavelengths above the bandgap, but the calculated average reflectance between 840 nm and 870 nm (R_{ZnS}) is equal to 96.5% for both 60 nm and 100 nm of ZnS. For shorter wavelengths (important to avoid J_{sc} losses), however, the reflectance of 60 nm of ZnS is higher than for 100 nm. Therefore, 60 nm was the thickness of ZnS applied to the produced devices.

3.2. Rear contact patterning to improve the reflectance

From the previous evaluation it is clear that, compared to the p-GaAs/Au structure, the ZnS/Au structure has an improved reflectance of photons in the relevant wavelength range (840–870 nm) to the active layers cells (approximately 63.5% compared to 96.5%). Therefore the reflectance at the rear side of thin-film GaAs solar cells can be improved by reducing C_r and with the application of an intermediate ZnS layer. To evaluate this experimentally, cells with four different rear contact geometries ($C_r = 100\%$, 50%, 30% and 10%) were produced simultaneously from a single wafer (see Fig. 2). In this way, any performance variation observed between the solar cells can unambiguously be assigned to the rear side patterning, rather than eventual small run to run

differences in wafer processing. The electroluminescence images of the thin-film cells, shown in Fig. 6, clearly reflect the rear side contact geometry of the devices, as this technique injects current in the solar cells through the contacts, leading to radiative recombination in the active area. Because the current density is highest in the direct vicinity of the contact areas, these are brighter due to higher recombination rates. The obtained contact pattern is well defined and there is no apparent defect formation resulting from the patterning.

As first order approximation, the average rear side reflectance in the wavelength range from 840 to 870 nm of the cells with different C_r values can be determined by $R_{cell} = C_r R_{GaAs/Au} + (1 - C_r) R_{ZnS/Au}$. This approximation yields values of 63.5%, 80.0%, 86.6% and 93.2% for $C_r = 100\%$, 50%, 30% and 10% respectively. The total reflectance of the processed DJ cell structures without grid fingers, measured using a spectrophotometer perpendicular to the cell front surface, are shown in Fig. 7. Similar as calculated above for the photons in the 840–870 nm range, photons with wavelengths above this range show enhanced reflectance as C_r is reduced and the area covered with ZnS, beneficial to the reflectance, is increased. However, the absolute value of the reflectance for these photons is higher because they do not suffer from absorption in the GaAs rear contact layer. Unfortunately, the solar cell without grid fingers with $C_r = 50\%$ was damaged, and therefore there is no reflectance spectrum for this configuration.

The illuminated IV parameters under 1 sun conditions of the best performing cells in both the SJ and DJ set of devices with different C_r

Fig. 4. a) Structure used to simulate the reflectance of photons emitted from the active GaAs layer (n_0) at the rear surface of thin-film GaAs solar cells with dielectric materials replacing the normally applied GaAs contact layer, and the calculated wavelength dependent reflectance at these structures considering b) ZnS and c) MgF₂ as the dielectric materials.

Fig. 5. a) Schematic depiction of the structure used for reflectance measurements and b) measured reflectance curves for samples with different thicknesses of ZnS.

Fig. 6. Electroluminescence images of the cells with C_r equals to a)100%, b) 50%, c)30% and d)10%. The camera parameters and the applied current were kept constant for all measurements.

values are shown in Table 1. For the SJ devices, $C_r = 50_r = 50\%$ was not applied and there are no results for this geometry. In the SJ devices, the J_{sc} remained constant, but both the V_{oc} and the FF values decrease as C_r decreases, and no advantages were observed with the patterning. In the DJ configuration, both J_{sc} and V_{oc} increase for lower C_r values. The FF of the cells essentially stays constant only to drop suddenly by more than 3% for $C_r = 10_r = 10\%$, most probably by an increase in series resistances related to the strongly reduced rear contact area. In these samples, the thick and continuous rear contact reduces metal contact resistances to a negligible value, indicating that the main loss mechanism is the lateral resistance that carriers encounter from the position they are generated to the contact points. An optimized design with more evenly dispersed smaller contact points for the rear contact is expected to reduce the fill factor losses considerably, thus allowing for the production of devices with even lower C_r .

Fig. 7. a) Reflectance curves obtained from the front surface of fully processed DJ solar cells without front grid fingers. b) Detail of the reflectance around the bandgap wavelength.

The differences in performances between SJ and DJ devices are explained with the analysis of the dark curve. The diode recombination current density is described by [26]:

$$J_{rec} = J_{01} \left(e^{\frac{qV}{kT}} - 1 \right) + J_{02} \left(e^{\frac{qV}{2kT}} - 1 \right) \quad (1)$$

with $n = 1$ the radiative and $n = 2$ the non-radiative recombination ideality factors. The non-radiative recombination is known to predominantly take place in the depletion zone and the perimeter of the cell, while the radiative recombination is dominant in the neutral regions of the cell. For DJ cells at operating voltage, the contribution of non-radiative recombination is lower than for SJ cells [22]. The ratio between the recombination currents is voltage dependent, with non-radiative recombination dominating J_{rec} at low voltages and radiative recombination at higher voltages. In the analysis of the dark current at lower voltages of the produced samples with $C_r = 100\%$ we find that the non-radiative saturation current density (J_{02}) is one order of magnitude higher for the SJ devices ($3 \times 10^{-11} \text{ A/cm}^2$) compared to the DJ geometry ($4.8 \times 10^{-12} \text{ A/cm}^2$). At the operating voltage, J_{rec} of the SJ cell is therefore mostly non-radiative and consequently the application of an improved geometry for photon recycling in these cells has no noticeable effect on the cell performance.

A $J_{sc} - V_{oc}$ curve composed of data points of I-V curves measured under different light intensities represents the ideal diode dark characteristics, as the series resistance is excluded under these conditions [32]. At higher voltages (the $n = 1$ region) it is possible to determine the radiative saturation current density (J_{01}), and these results are shown in Fig. 8. For the produced DJ cells, with the decrease of C_r (and the increase in reflectance) the J_{01} values also decrease. This irrefutably shows an improvement in re-absorption of emitted photons generated by band-to-band recombination [22,25].

The external quantum efficiencies (EQE) of these devices, shown in Fig. 9, confirm that the mild increase in the J_{sc} in the DJ devices is due to the increased reflection of the incident light. Most of the incident light with shorter wavelengths is absorbed before reaching the rear layers and no differences in the spectrum are expected in this region, but for wavelengths above 700 nm, the EQE of the cells clearly increases.

Miller et al. [16] have modeled the effect that the rear mirror reflectance has on the cell parameters of a $3 \mu\text{m}$ thick GaAs solar cell in the absence of non-radiative recombination. The model predicts a more than linear increase in V_{oc} and a linear increase in J_{sc} as the reflectance approaches 90%. In order to facilitate the comparison between the results from the model and those from solar cells produced in this study, the earlier approximated R_{cell} value for each configuration is related to its V_{oc} and J_{sc} in Fig. 10. For easy comparison, the curves from the

Table 1

Illuminated IV parameters of both SJ and DJ solar cells with different rear surface contact fractions.

	SJ devices			DJ devices		
	J_{sc}	V_{oc}	FF	J_{sc}	V_{oc}	FF
C_r [%]	$\pm 0.06 \left[\frac{\text{mA}}{\text{cm}^2} \right]$	$\pm 2 \text{ [mV]}$	$\pm 0.3 \text{ [%]}$	$\pm 0.06 \left[\frac{\text{mA}}{\text{cm}^2} \right]$	$\pm 2 \text{ [mV]}$	$\pm 0.3 \text{ [%]}$
100	24.17	1006	85.0	24.34	1050	85.8
50	–	–	–	24.23	1053	85.6
30	24.14	1000	84.4	24.43	1055	85.5
10	24.15	0992	83.6	24.46	1058	82.1

Fig. 8. a) $J_{sc} - V_{oc}$ curves measured at different light intensities of the DJ devices with different C_r and b) the calculated J_{01} of each device. **Fig. 9.** a) External quantum efficiency of the DJ samples with different C_r . b) Detail of the curve at the longer wavelength region.

literature are (on a different scale) also depicted in the same graphs. The obvious differences in the absolute V_{oc} and J_{sc} values derived from the model reported in literature and the values obtained in the here reported experiments are due to the fact that the model represents a simplification of the actual situation. The model, for example, neglects the reflection of light at the ARC and grid contacts of the cell and assumes the total absence of non-radiative recombination. Nevertheless, the obtained trends for the produced cells are similar to those following from numerical simulations, providing experimental proof of concept of this relationship.

4. Conclusions and outlook

Reemitted photons by band-to-band recombination in GaAs solar

cells have a wavelength that varies from 840 to 870 nm. In this region, the rear contact layer in thin-film GaAs solar cells absorbs a relevant fraction of photons, that therefore do not contribute to the solar cell performance. Both theoretical and experimental results show that compared to the use of a Au mirror directly on the BSF of the cell a further improved reflectance can be obtained by the application of an intermediate ZnS or MgF_2 layer. Based on these results an approach comprising partial removal of the p-GaAs contact layer and replacement by deposition of ZnS was developed and tested in both shallow junction and deep junction solar cell geometries. The calculated total reflectance at the active layers of these cells increases from 63.5% in the original structure ($C_r = 100\%$), up to 93.2% with the new design ($C_r = 10\%$).

In thin-film SJ devices the non-radiative saturation current density (J_{02}) is higher and no positive effects on the cell performance are

Fig. 10. Measured a) V_{oc} and b) J_{sc} of the DJ solar cells related to the respective average reflectance of the processed cell (R_{device}). The obtained trend in both parameters is similar to that predicted in the literature.

apparent by this approach. However, with the obtained increase in reflectance, the illuminated IV parameters of DJ solar cells improve, and the radiative saturation current density (J_{01}) decreases. This demonstrates an increase in photon recycling and reaffirms the importance of the rear mirror reflectance in the design of thin-film GaAs solar cells.

The results of this study experimentally confirm previously reported theoretical calculations that relate the reflectance of the rear mirror and the solar cell parameters. Furthermore, the technique developed in this work provides a clear reduction of J_{01} without any required changes to the growth structures of the solar cell. Therefore, with further reduction of the non-radiative losses and an optimized rear and front contact geometries, patterning the rear contact represents a viable approach to achieve efficiencies closer to the theoretical limit.

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