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Ultrathin Cu(In,Ga)Se₂ solar cells with point-like back contact in experiment and simulation

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ABSTRACT

We apply the concept of point contact solar cells to a model system having 190 nm thick Cu(In,Ga)Se₂ (CIGSe) films by using a SiO₂ back side film with periodic openings to the molybdenum layer being the electrical contact. The openings are plasma etched through a mask prepared by laser interference lithography. We find a maximum increase of the short circuit current density of 25% for a SiO₂ thickness of 60 nm and structure length of 1,1 μm. This gain is due to (1) coherent optical reflection, (2) light scattering enhancing the quantum efficiency at all wavelengths and (3) an anticipated wave guide effect which boosts the quantum efficiency particularly at long wavelength. The best efficient solar cells with 190 nm thick CIGSe has a conversion efficiency of 9% (fill factor = 70%) without anti-reflection coating. These experimental results can largely be confirmed by simulation.

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1. Introduction

Thickness reduction in solar cells can reduce material and capital costs. However, thickness reduction shall not compromise the solar-to-electric conversion efficiency of the cells. In principle, there are 4 different obstacles for thickness reduction: (1) Shunt-forming pinholes, (2) Back-contact recombination, (3) Reduced bulk lifetime due to reduced processing time, and (4) Optical losses. In Cu(In,Ga)Se₂ (CIGSe) technology, pinholes can partly be isolated by the non-doped ZnO layer [1]. Concerning back contact recombination there is still some controversy. Vermang showed that the non-passivated Mo/MoSe₂/CIGSe back contact has an electron recombination velocity of minimum $S_n = 3 \times 10^4$ cm/s and maximum $S_n = 8 \times 10^5$ cm/s thus rendering surface passivation extremely important [2]. We recently found that Sodium passivates the Mo/MoSe₂/CIGSe back contact down to $S_n = 10^2$ cm/s leaving little room for further passivation by the point contact concept [3]. Assuming the latter, what remains are obstacles (3) and (4) on which this work shall concentrate on. Optical losses may be related to the modified film structure due to thinning and the mere absorption due to optical path length. In [4] we showed that the absorption coefficient of a CIGSe film is not dependent on the film thickness, thus voids or density fluctuations play a negligible role. Recently, Vermang et al. showed that a reflective rear surface passivation layer can improve the short circuit current J_{sc} of CIGSe solar cells with 400 nm CIGS layer [5]. The thickness of the passivation layer out of Al₂O₃ or out of combined

MgF₂ and Al₂O₃ was limited to <65 nm due to technical reasons. On the other hand, the process applied in [5] in order to form nano-sized local point contact openings is interesting for large scale production. Van Lare et al. used imprint lithography on 460 nm thick CIGS films [6]. Here, we employ even thinner CIGS layers of 190 nm and form local point contacts of Mo in a matrix of SiO₂ by laser-interference-lithography. We take the flat Mo/MoSe₂ back contact as the benchmark reference. We find that with appropriate SiO₂ thickness, the short circuit current can be increased by 15% due to coherent reflectivity at the back contact. A further 10% current increase can be obtained by a light trapping effect at the structured SiO₂. We find indications for a near infrared waveguide effect in experiment and simulation at appropriate structuring scale. Using theoretical absorption spectra, the gain in short circuit current due to back contact structuring can be explained to a large extend – thus supporting our view of the Sodium passivated Mo/MoSe₂ back contact.

2. Experimental

Solar cells are prepared on 3 mm soda-lime-glass substrates covered with a SiO_xN_y barrier layer and 500 nm sputtered molybdenum of 0,3 Ω/sq. sheet resistance. The substrate size is 2 × 2 cm². For the fabrication of point-like back contacts, the substrates were covered by a SiO₂ layer, which was structured using laser interference lithography (LIL) and reactive ion etching. A detailed description of the LIL setup and procedure can be found in [7]. For hexagonal lattices with periods from 230 nm to 460 nm, a three beam setup with single exposure was used; for larger periods of 1100 nm and quadratic lattice a two beam setup and double

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exposure with rotation of the substrate by 90° after first exposure was applied. After masking by LIL the SiO₂ layer was etched in a Plasmalab100 system (Oxford Instruments) using a C₄F₈/O₂ plasma (C₄F₈: 40 sccm, O₂: 15 sccm, RF: 105 W, Inductive coupled plasma: 2500 W for 65 s). The SiO₂ coverages of the three employed periods are given in Table 1. Fig. 1 shows the example of a hexagonal lattice of back contact openings for the 0,46 μm period.

CIGSe films are deposited in a vacuum chamber using the 3-stage deposition process [4,8] with laser light scattering [9] and substrate temperature monitoring [10]. The metal source fluxes in the 2nd and 3rd stages were reduced for smaller CIGSe target film thickness in order to enable the detection of stoichiometric points and in order to have a similar temperature budget on the growing film as for thicker CIGS. All samples have similar chemical composition of [Cu] / ([Ga] + [In]) = 0,88–0,92 and [Ga] / ([Ga] + [In]) = 0,28–0,29 according to energy dispersive X-ray spectroscopy in the Scanning Electron Microscope (SEM). Film thicknesses were measured by profilometry and SEM cross sections. Time resolved photoluminescence transients were recorded directly after CIGSe preparation (for experimental details see [3]). Solar cells of 0,5 cm² were completed with a CdS buffer layer, sputtered i-ZnO/ZnO:Al, and Ni/Al/Ni grid. No antireflection coating has been applied. Admittance spectroscopy measurements and secondary ion mass spectrometry depth profiles were performed as reported elsewhere [4]. The optical properties of our films that we use for device simulation as determined by transmittance and reflectance measurements are very much in agreement with the data of Orgassa [11]. Optical simulations were performed using the Stanford Stratified Structure Solver S⁴ [12] which solves the linear Maxwell equations by “rigorous coupled-wave analysis”.

3. Results and discussion

The main objective of a back contact dielectric is the increased reflectance due to (i) a large difference in refractive indices between SiO₂ ($n = 1,54$) and CIGSe ($n = 3,0$) and (ii) the constructive interference of coherent light. In the simulated data of Fig. 2 we observe a broad band reflectance of up to 70–80% for SiO₂ thicknesses around 120 nm – much higher than the Mo/MoSe₂ reflectance [11]. We selected 60 nm and 130 nm SiO₂ for 3D simulation and for the experimental verification. Fig. 3 shows the cross section of samples with 130 nm SiO₂ and structure dimensions as listed in Table 1 for series VI. Due to etching phenomena, the walls of the holes in the SiO₂ layer are conical. Nevertheless it was possible to form a regular structure of holes in the SiO₂ – opening the electrical contact to the Mo back contact as can be seen in Fig. 1.

The optical effects of this back contact structures were studied in Fig. 4a–c by 3D simulations using the program S⁴. Here, the optical absorption for the complete material stack in front of the Mo layer is given. Due to low absorption of SiO₂ in the considered wavelength range, this quantity reasonably approximates the CIGSe absorption. The

comparison between SimA (Mo/MoSe₂ back contact) and SimE (Mo/MoSe₂/SiO₂ back contact) shows the mere effect of increased reflectance due to constructive interference of dielectric reflectance. As already depicted in Figs. 2, 130 nm SiO₂ layer reflection slightly outperforms the 60 nm reflection in that respect. Next we simulated the structured SiO₂ layer as schematically shown in Fig. 4d. The inclined hole edges have been taken into account by a step function of up to 20 steps (schematically represented by only one step in Fig. 4d). In the simulated absorption of the structured SiO₂ (SimB–D), the interference maxima become shifted and absorption generally increases for SimD and SimC. This is true for cases I (60 nm SiO₂) and II (130 nm SiO₂). It is particularly instructive to compare SimEII and SimCII in Fig. 4b: There is an absorption gain at around 800 nm which we assign to scattering. Only for the 130 nm SiO₂ layer with 460 nm period (SIMCII), there is an absorption gain at around 1100 nm. This gain we tentatively assign to a waveguide effect. The interpretation of a waveguide effect is in accordance with the results of finite-difference time-domain simulations in Ref. [6]. If we assume complete collection of the excited charge carriers in the finished solar cell, we can calculate the photocurrent J_{ph} from the simulated absorption. Fig. 4c shows relative values of J_{ph} as referenced to the Mo/MoSe₂ cases of SimAI and SimAII. In addition, Fig. 4c gives the calculated photocurrent for the hypothetical cases SimEI (black dashed line) and SimEII (red dashed line). The current of SimEII is increased by 22% due to increased reflectance. Given that the structured SiO₂ has an average coverage of 70%, we find that about 15% (0,7 × 22%) of photocurrent can be gained from improved optical reflection at the Mo/MoSe₂/SiO₂ back contact and another 10% gain results from structuring the back contact inducing additional scattering and waveguide effects. In total the optical gain can be 25%.

The back contact structures of Table 1 have been completed to solar cells. By adjusting the Sodium content in the layer using 1,6 nm of NaF in a post-deposition-treatment we can achieve a decay time of the photoluminescence (TRPL) of 7 ns. As shown in Ref. [3] this Sodium doping is appropriate for back surface passivation. On the other hand it is low enough to avoid bulk recombination. We find that the Na concentration of 190 nm films grown without diffusion barrier is 3 times higher than the one of 1,5 μm films and for that reason films without diffusion barrier suffer from bulk recombination with TRPL decay times of 2 ns. The carrier concentration as determined by admittance spectroscopy of thin layers of adjusted Na concentration is about $1 \times 10^{16} \text{ cm}^{-3}$ (value after light soaking). Table 2 gives the solar cell parameters as determined from AM 1,5 illuminated cells and parasitic resistances as determined from dark JV measurements. We find that besides for sample DV there is trend of increasing series resistance with period length. This we tentatively assign to the longer path of majority carriers to the back contact due to structuring. However, this assignment would need corroboration by simulation. There is no clear trend of the shunt resistances. Also the fill factors, FF, do not show a clear trend with maximum values above 70% which is reasonable for these non-optimised cells. The clearest trend comes with the short circuit current,

Table 1
Structure sizes and film thicknesses from experimental series V and VI and simulation series I and II. Lattice parameter a_{SiO_2} with hexagonal or quadratic symmetry. Hole diameter within the SiO₂ layer ϕ_{SiO_2} and SiO₂ coverage percentage. Samples AV und AVI with simulations SimAI and SimAII represent the reference case with Mo/MoSe₂ back contact. Simulations SimEI and SimEII show the hypothetical cases of complete SiO₂ coverage without electrical contact openings. [Cu] / ([Ga] + [In]) ratio CGI and [Ga] / ([Ga] + [In]) ratio CGI.

Sample	Simulation	d_{CIGS} [μm]	CGI	GGI	d_{SiO_2} [μm]	d_{NaF} [nm]	Symmetry	a_{SiO_2} [μm]	ϕ_{SiO_2} [μm]	SiO ₂ [%]
AV	SimAI	0,19	0,88	0,28	0	1,6				
BV	SimBI	0,19	0,88	0,28	0,06	1,6	Hex.	0,23	0,14	67
CV	SimCI	0,19	0,88	0,28	0,06	1,6	Hex.	0,46	0,23	77
DV	SimDI	0,19	0,88	0,28	0,05	1,6	Quad.	1,1	0,66	72
	SimEI	0,19			0,06					100
AVI	SimAII	0,19	0,92	0,29	0	1,6				
BVI	SimBII	0,19	0,92	0,29	0,13	1,6	Hex.	0,23	0,14	40
CVI	SimCII	0,19	0,92	0,29	0,13	1,6	Hex.	0,46	0,23	67
DVI	SimDII	0,19	0,92	0,29	0,13	1,6	Quad.	1,1	0,66	72
	SimEII	0,19			0,13					100

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