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Optical modeling of chalcopyrite-based tandems considering realistic layer properties

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Previous models of chalcopyrite-based tandem solar cells have not taken into account the limited optical transmission of the top cell observed. We use a quantitative model derived from measured optical properties of the different layers of the top cell to re-evaluate the benefits and limitations of the tandems. Guidelines are provided for minimizing optical losses in the structure. Optimization of the bottom absorber band gap and top absorber thickness is carried out. In combination with straightforward assumptions concerning the electronic cell properties, we calculate tandem maximum efficiencies in the range of 26%–28% depending on the degree of nonideal optical absorption. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3077613]

Tandem solar cells attract attention with respect to an improved exploitation of the solar spectrum as a route toward higher efficiencies. In the case of perfect absorbers, only specified by their band gaps and including radiative recombination as single source of losses, the maximum tandem output was found to be 42.9%.[1] A more realistic calculation based on the extrapolation of experimentally achieved best single cell results states a maximum achievable efficiency of 28.2% for absorber band gaps $E_{g,\mathrm{top}}=1.74$ and $E_{g,\mathrm{bottom}}=1.15$ eV.[2] To reap the benefits of the tandem, the top cell needs to be highly transparent in the energy range below its absorber's band gap. In chalcopyrite thin film solar cells $CuGaSe_2$ (CGS) with $E_g=1.68$ eV seems a good choice as top cell absorber. However, only around 60% transmission was achieved experimentally for *n*-ZnO/*i*-ZnO/CdS/CGS/TCO/SLG (TCO denotes transparent conducting oxide, here fluorine-doped tin oxide, type AsahiU,[3] SLG denotes soda lime glass) cells.[4]

In this contribution, we will take into account these findings and evaluate in more realistic detail the benefits and limitations of a chalcopyrite tandem with a CGS top cell. In contrast to previous work, we start for the first time with a detailed optical model of an actual transparent CGS solar cell on glass (initial stack), which allows us to include (case A) or exclude (case B) nonideal absorption mechanisms (defects, band tails, and free carriers). Further on we suggest what would be the optimal structure (optimized stack) with minimized reflection losses. This structure is then extended to an optical model of the complete tandem cell with bottom cell $Cu(In,Ga)Se_2$ (CIGS) absorber.[5] The photocurrents are calculated from the absorption in CGS and CIGS assuming complete collection of photogenerated carriers. $j(V)$ curves of the bottom and top cell are each evaluated in a standard diode model. Saturation current j_0 and diode factor A are extrapolated from the experiment (latest at 19.9% efficient cell[6]). We assume all cells to have the same A and recalculate j_0 as a function of the band gap according to[7]

$$j_0 = j_{00} \exp[E_g/(Ak_BT)],$$

where k_B is the Boltzmann constant, e is the elementary charge, T is the temperature, and j_{00} is the extracted form.[6] Since the bottom and top cells are stacked monolithically (two-terminal configuration) the $j(V)$ curve of the tandem is calculated by adding the voltages across the bottom and top cells, respectively, thus considering current match. The maximum power point of the combined curve is extracted numerically and finally used to calculate the efficiency.

Based on optical characterization of the different layers, prepared and measured individually and in all relevant combinations, we had already derived an optical model of the initial stack.[8] The DIPLOT[9] numerical modeling software allowed us to quantify the influence of nonideal absorption mechanisms in these layers. In order to assess in similar detail the losses due to partial reflection at the interfaces the wavelength-dependent data (refractive index n, coefficient of extinction k) were extracted from DIPLOT and used here as input for further calculations with the SunShine simulator.[10] According to this, the ranking of the most critical interfaces and the resulting options for improvement in the CGS top cell are as follows: (1) air/*n*-ZnO (entrance at the front, responsible for 64% of the total reflection calculated as shape of photocurrent loss), to be reduced by adding a MgF_2-antireflection coating with intermediate refractive index; (2) TCO/SLG, SLG/air (exit at the back, 19%), to be overcome by switching to monolithic integration; in the model this is expressed by an imaginary CdS substrate (replacing SLG) and suppressed back at the side reflection in order to represent the transition into the bottom cell; (3) CdS/CGS, CGS/AsahiU ($CuGaSe_2$ interfaces, 17%), not considered here. Based on the results of optical losses in the top cell, due to reflectances as well as parasitic absorptances, we propose a top cell stacking optimized with regard to optical transmission [optimized stack, Fig. 1(b)].

Besides antireflective measures mentioned, the layer thicknesses were adjusted as well. Starting values were generated from recent experimental findings concerning window layers with reduced thickness[11] and from estimating optimum layer thicknesses according to the $\lambda/4$ condition

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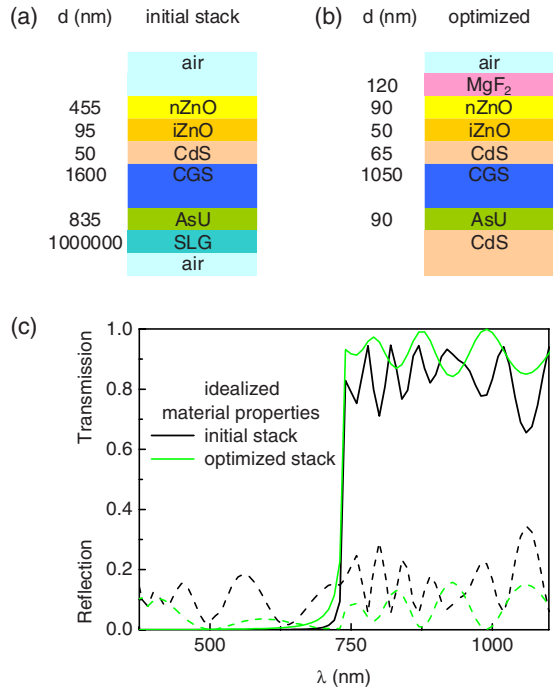

FIG. 1. (Color online) Initial top cell stack (a) and optimization (b) derived from optical modeling. Calculations were carried out assuming no parasitic absorption and reveal (c) the reduced reflection/increased transmission due to improved optical matching.

$d=m(\lambda/4n)$ (m an odd integer), evaluated at $\lambda=800$ nm. The thicknesses were then refined in iterations with the optimization tool of the SunShine simulator, aiming at minimum reflectance or maximum transmittance of the stack, keeping the absorption in the CGS high. The resulting thicknesses are given in Fig. 1(b) in comparison to the initial values Fig. 1(a). The calculated transmission and reflection curves are shown in Fig. 1(c) for case B (no parasitic absorption). The average subgap transmission of the initial stack is in the average of 80% and could be increased to 90% for the optimized stack. In comparison to the experimental value of 60%, this would imply that roughly two thirds of transmission losses are due to nonideal material properties achieved in the experiment but one third is due to the stack layout and can be avoided.

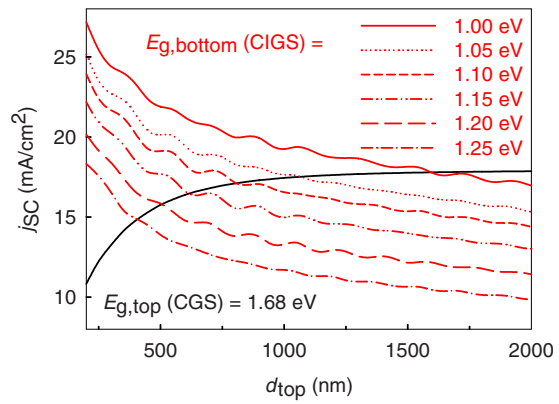

FIG. 2. (Color online) Photocurrent densities of top (fixed band gap $E_{g,\mathrm{top}}=1.68$ eV) and bottom cells with different band gaps ($E_{g,\mathrm{bottom}}$) as a function of top absorber thickness d_{top}. The calculation is based on optical properties achieved in the experiment.

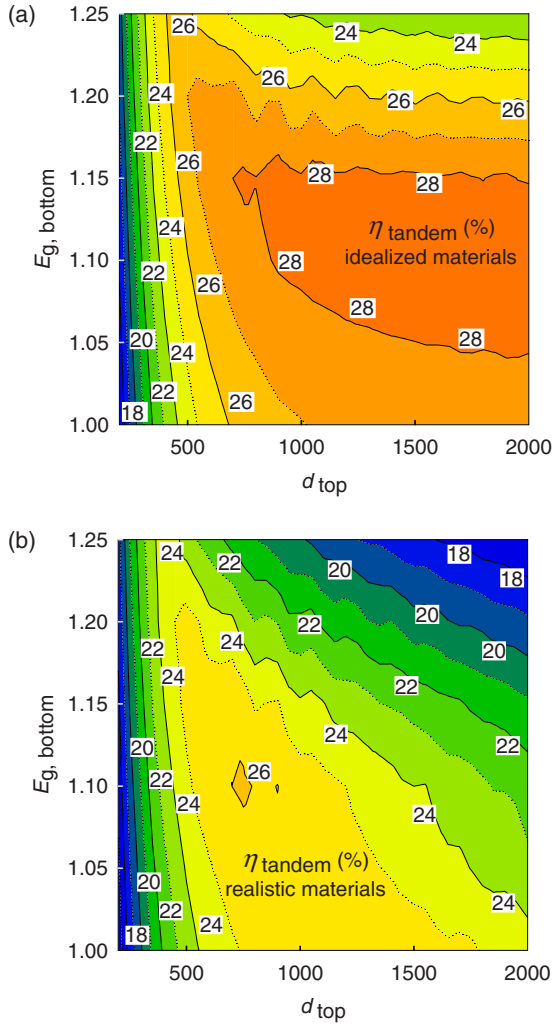

FIG. 3. (Color online) Tandem efficiencies calculated for varied bottom absorber band gap $E_{g,\mathrm{bottom}}$ and top absorber thickness d_{top}. In the absence of parasitic absorption the maximum efficiency is 28% (a). It drops to 26% (b) when calculations are based on experimentally achieved optical properties. The correct choice of design parameters ($E_{g,\mathrm{bottom}}$, d_{top}) is crucial in the latter situation.

This optimized top cell stack of Fig. 1(b) was now used to revise the overall tandem construction. In the model CIGS the bottom cells on molybdenum (Mo) back contact were added to the monolithic structure resulting in a MgF_2/n-ZnO/i-ZnO/CdS/CGS/AsU/CdS/CIGS $\times$(2000 nm)/Mo/SLG tandem. As mentioned before we used n,k-data from Ref. 5 for the bottom absorber. They were translated with DIPLOT to varied band gap and then used as input for SunShine. The top cell properties were fixed at this step except for the CGS layer thickness.

The light absorbed in the top and bottom absorbers was calculated for a top CGS material of fixed $E_{g,\mathrm{top}}=1.68$ eV and a bottom CIGS with varying energy gap. The result, expressed in terms of photocurrent densities (assuming complete carrier collection under AM 1.5 spectrum for both absorbers in the tandem), is presented as a function of top absorber thickness d_{top} in Fig. 2 (case A). The photocurrent generated in the top cell saturates according to the high absorption associated with the band-to-band transitions at d_{top} $\approx$1000 nm. Absorption caused by band tails and defects does not contribute to the current but—with increasing top absorber thickness—significantly reduces the amount of light

coupled into the bottom cell. An intersection of top and bottom absorber current density (perfect current match) is achievable for a wide range of bottom band gaps. In contrast, current match in the absence of parasitic absorption (case B, not shown here) is possible only for $E_{g,\mathrm{bottom}} > 1.10$ eV. However, judging the design parameters only on the basis of current match can be misleading (voltages not included). It is therefore mandatory to calculate the tandem efficiency.

Further analysis requires the calculation of $j(V)$ curves and, hence, additional assumptions concerning the saturation current and diode factor. As outlined above, our assumptions were based on experimental data ($A=1.14$, $j_0=2.1\times10^{-9}$ mA/cm^2 at $E_g=1.18$ eV), which were extrapolated to all band gaps required for the model. Resulting calculated bottom cell efficiencies range from 17.2% at $E_g=1.0$ eV to 20.7% at 1.25 eV. The tandem efficiency in two-terminal configuration is mapped in Fig. 3 as a function of d_{top} and $E_{g,\mathrm{bottom}}$. In case B of ideal materials, an efficiency of $\eta_{\mathrm{tandem}} \geq 25\%$ is easily achievable, and also for values $\geq 28\%$, we find an acceptable parameter window [Fig. 3(a)]. This is no longer the case when the calculation is repeated for optical properties derived from the experiment [case A, Fig. 3(b)]. The maximum efficiency decreases to 26% and the parameters become more critical.

For achieving relatively high efficiencies of the optimized tandems, one should further focus on improving the performance of the wide gap top cell, which is the most critical issue in practice. As we have shown here in the first quantitative model, the optical properties of the transparent top cell are also a major concern. Mismatch of refractive indices causes reflection losses. They can be minimized by our suggested optimized stack, which would still reach an efficiency of 28% assuming the electrical properties of the top cell to be comparable to those of the best chalcopyrite low-gap cell realized in practice. When nonideal absorption mechanisms are taken into account, the maximum efficiency drops to 26% and the bottom absorber band gap as well as top absorber thickness needs to be adjusted precisely. This clearly represents a worst case scenario as parasitic absorption in the CGS absorber will be less significant once the necessary electrical properties have been achieved (reduced defect density). We conclude that the optical properties of the top cell do not prevent the realization of efficient chalcopyrite-based tandems. However, monolithic stacking appears to be mandatory and is a significant technological challenge in itself.[12] Whether the required electrical performance of the top cell can be achieved remains an open question.

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