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Optical approaches to improve the photocurrent generation in Cu(In,Ga)Se₂ solar cells with absorber thicknesses down to 0.5 μm

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Improving the optical management is a key issue for ultrathin based solar cells performance. It can be accomplished either by trapping the light in the active layer or by decreasing the parasitic absorptions in the cell. We calculate the absorption of the different layers of Cu(In,Ga)Se₂ (CIGSe) based solar cell and propose to increase the absorption in the CIGSe layer by optimizing three parameters. First, by increasing the transmitted light to the cell using a textured surface of ZnO:Al front contact which functions as a broadband antireflection layer. Second, by replacing the CdS/i-ZnO buffer layers with ZnS/ZnMgO buffer layers which have higher energy bandgaps. Third, by replacing the Mo back contact with a higher reflective metal, such as silver or gold. Calculations show that modifying these layers improves the total absorption by 32% in a 0.5 μm thick CIGSe absorber. These predicted improvements of the short circuit current are confirmed experimentally.

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I. INTRODUCTION

In recent years, chalcopyrite solar cells based on Cu(In,Ga)Se₂ (CIGSe) absorbers have reached the highest efficiency levels of all thin film photovoltaic technologies, and thus turns up to be an ideal combination of low cost and high efficiency. Laboratory scale cells have reached certified efficiencies of 20.3%,¹ which is very close to the efficiency of cells based on multi-crystalline Si wafers. 1 m² modules have reached certified efficiencies of above 14%.² However, this increasing development of CIGSe solar cells may have an impact on the indium world resources when reaching the Tera-Watt scale³ and thus reduce the competitiveness of CIGSe. Reducing the CIGSe thickness has been identified as a key issue to reduce the indium consumption per Wp, allowing both cost reduction and preservation of the material's availability. In order to preserve high efficiencies using much thinner layers, it is necessary to absorb light more efficiently in the absorbers.

The thickness of the CIGSe layer in a standard photovoltaic cell is approximately 2 μm , which is larger than the penetration depth in visible light.^{4,5} Thus, the light is sufficiently absorbed. Based on previous studies,⁶ the light absorption and thereby the short circuit current (J_{sc}) decrease when the absorber thickness is reduced. In addition, when the CIGSe thickness is less than 0.5 μm , the impact of back contact recombination increases, leading to insufficient carrier collection.⁷ In order to improve the efficiency of the cell, it is firstly essential to understand what portion of the absorbed light will generate electrical current. This can be found by: (i) measuring the reflectivity of the cell to derive its total absorption, and (ii) measuring the external quantum efficiency (EQE), associated with the absorption and collection

in the active layer (the CIGSe). However, it is still uncertain whether the photocurrent is lost due to imperfect collection or parasitic absorption. In this paper, we calculate the optical losses in the different layers of the solar cell with an absorber thickness of 0.5 μm . A solar spectrum AM1.5 impinges on the cell in a normal direction. The spectral absorption by the cell was found to be above 80% for wavelengths smaller than 1000 nm. However, by calculating the optical absorption in details, it is seen that light in the visible range is absorbed in the buffer layer (CdS) before it reaches the active layer. Furthermore, in the infrared spectrum, most of the radiation is lost in the back-contact. It should be emphasized that only the light absorbed in the CIGSe contributes to the photocurrent generation. Hence, to enable more absorption in the active layer, the absorption in the other layers should first be minimized. We confirm our calculations by replacing CdS with ZnS and Mo with a more reflective metal such as gold, and finally by measuring the resulting J_{sc} . In addition, we show that when the front contact (ZnO:Al) is textured, it behaves as an antireflection layer for a broad spectrum and thus further increases the absorption of the cell and consequently the EQE and the short circuit current.

Replacing the buffer layer and the back contact enables to maintain the high absorption of the cell almost solely in the active layer. The most remarkable advantage of this cell structure is the simple fabrication process: deposition of thin films having flat interfaces with no further photolithography or embedding of particles on the interfaces, as currently proposed in optical management techniques. In addition, recombination at the back interface is minimized.

II. EXPERIMENT

As a reference solar cell, we used sample (a) in Table I, schematically shown in Fig. 1. The CIGSe layer was

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TABLE I. Summary of CIGSe based solar cell configurations with corresponding short circuit current with absorber thicknesses of 2 μm and 0.5 μm [0.4 μm in sample (d)].

Sample	Configuration	J_{sc} (calc. – 2 μm) (mA/cm ²)	J_{sc} (calc. – 0.5 μm) (mA/cm ²)	J_{sc} (exp. – 0.5 μm) (mA/cm ²)
(a)	Mo/CIGSe/CdS/i-ZnO/ZnO:Al	30.40	23.93	23.34
(b)	Mo/CIGSe/CdS/i-ZnO/ZnO:Al/ARC	33.19	25.60	24.90
(c)	Mo/CIGSe/ZnS/ZnMgO/ZnO:Al	33.20	25.93	26.08
(d)	Au/CIGSe/CdS/i-ZnO/ZnO:Al/Glass	32.67	26.68	25.20
(e)	Au/CIGSe/ZnS/ZnMgO/ZnO:Al/ARC	34.27	31.63	—

deposited at Würth Solar by co-evaporation⁸ on Mo coated glass substrate. In order to investigate how the solar cell performance depends on the CIGSe layer, its thickness was reduced using chemical etching in a HBr/Br₂ solution as described by Bouttemy *et al.*⁹ The CdS/i-ZnO layers are replaced by ZnS/ZnMgO layers [sample (c)]. The ZnS buffer layer was deposited by chemical bath deposition (CBD) technique using zinc sulfide, thiourea, and ammonia, and the ZnMgO was prepared by sputtering as presented elsewhere.^{10,11} Furthermore, the Mo layer is replaced by Au for different CIGSe thicknesses, as described in Ref. 12 [sample (d) in Table I]. Cells with antireflection coating (ARC) were also made by a deposition of 100 nm MgF₂ layer on top of the solar cells [sample (b)]. Texturing the ZnO:Al front contact was performed by wet chemical etching in diluted hydrochloride acid (0.5% in volume). In addition, we measured the quantum efficiency of the solar cells and the short circuit current using a class A double light source solar simulator (Wacom-WXS-140S-Super).

III. OPTICAL MODELING

For optical modeling, we consider the solar cell as a stratified media with flat interfaces, where the electromagnetic waves can be calculated at each position for any wavelength λ , incident angle θ_i , and polarization p . Here, the transfer matrix method¹³ is used to calculate the spectral absorption in layer m with thickness L by

$$A_\lambda(\theta_i, m, p) = \frac{[\mathbf{S}(m, 0) - \mathbf{S}(m, L)] \cdot \hat{z}}{\mathbf{S}_i(\lambda, \theta_i, p) \cdot \hat{z}}, \quad (1)$$

where $\mathbf{S}(m, l)$ is the Poynting vector of the electromagnetic field in layer m at position l , \mathbf{S}_i is the Poynting vector of the incident field, and \hat{z} is a unit vector normal to the interface (here, in direction z). We assume the solar light is unpolar-

ized; therefore, the spectral absorption is given by an average of both polarizations

$$A_\lambda(\theta_i, m) = \frac{1}{2} [A_\lambda(\theta_i, m, \text{TE}) + A_\lambda(\theta_i, m, \text{TM})]. \quad (2)$$

The back contact is considered as a semi-infinite layer since its thickness is much larger than the penetration depth of light. We found that the absorption in the layers does not depend on the angle of incidence up to 70° (not shown here). Therefore, for simplicity, we show calculations when illuminating light in normal direction. We begin by calculating the spectral absorption in all the layers of a CIGSe solar cell of 2~2.5 μm thick—hereafter, regarded as thick CIGSe. The results are shown in Fig. 2(a). As can be seen, the absorption by the cell (all the layers in the figure) is about 90% in the spectral range where CIGSe absorbs: between 400 and 1100 nm. However, detailed calculations show that a substantial part of the absorbed light is not in the active layer; therefore, it is considered as a loss in the device. We can see that there are mainly two sources for these losses: the CdS in the visible and the Mo in the infrared (IR) spectra. Below 500 nm wavelengths, the buffer layer significantly absorbs although its thickness is only 50 nm. This absorption should not be avoided because the solar spectrum has its maximum power in this spectral range which leads to significant reduction in the photocurrent. Since light passes the buffer layer before it reaches the CIGSe, similar effect is expected in the thin solar cell. Indeed, calculating the absorption in a cell with a CIGSe thickness of 0.5 μm (referred as thin CIGSe) shows similar absorption in the CdS layer [Fig. 3(a)]. This loss can be prevented by using materials with a higher energy bandgap such as ZnS which has an energy bandgap of 3.6 eV. Calculations for solar cell ZnS buffer layer are shown in Fig. 2(b), for the thick CIGSe layer, and in Fig. 3(b) for the thin one. We assess the advantage of changing the buffer layer by computing the total absorption in the CIGSe layer (in the relevant spectrum, from $\lambda_1 = 350$ nm to $\lambda_2 = 1200$ nm),

$$A(\theta_i, m) = \frac{\int_{\lambda_1}^{\lambda_2} A_\lambda(\theta_i, m) I_{\text{solar}}(\lambda) d\lambda}{\int_{\lambda_1}^{\lambda_2} I_{\text{solar}}(\lambda) d\lambda}, \quad (3)$$

where I_{solar} is the solar spectrum (AM1.5). The total absorption in the thin CIGSe layer is increased from 57% to 63%.

In the IR spectra, the radiation that is not absorbed by the front contact nor by the CIGSe layer (at wavelength

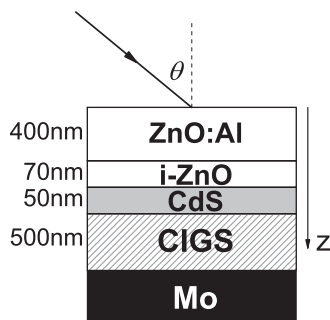


FIG. 1. Schematic set-up of CIGSe solar cell.

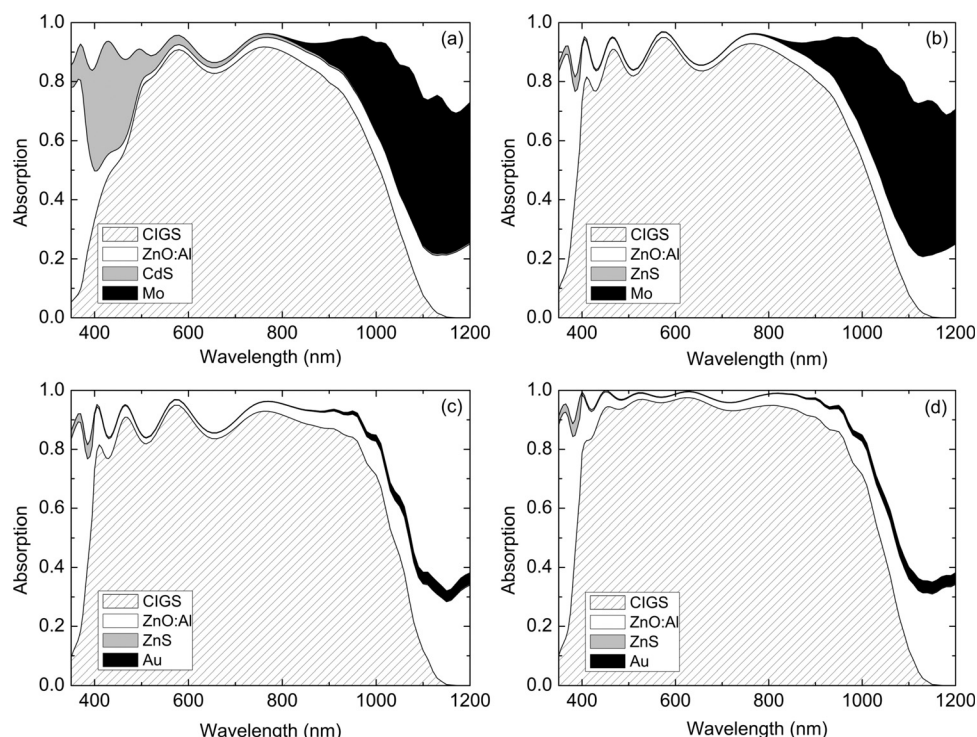


FIG. 2. Calculated spectral absorption of different layers of thick CIGSe solar cell ($2\sim 2.5\ \mu\text{m}$ thickness). (a) Same configuration as sample (a) in Table I with thick CIGSe. Total absorption in CIGSe layer is $A = 69\%$, derived from Eq. (3). (b) As sample (c) in Table I; $A = 76\%$. (c) As sample (e) in Table I without ARC; $A = 78\%$. (d) Sample (e) in Table I; $A = 83\%$.

larger than 700 nm) decays into the back contact, Figs. 2(a) and 2(b). Herein, the Mo has two functions: from the electrical point of view, it functions as the back contact electrode, but from the optical point of view, it should reflect the light back to the CIGSe layer in order to increase its absorption. However, since the reflection at the Mo/CIGSe interface is less than 20%, it was proposed to replace it by another metal such as Ag, Au, Cu, or Al which are known as good mirrors and conductors^{12,14} or to add a dielectric layer between the absorber and the back contact.¹⁵ The necessity to replace the

Mo layer is even more essential in ultrathin cells—a CIGSe thickness below $0.5\ \mu\text{m}$ —since less light is absorbed by the CIGSe in a single pass and therefore more light will eventually be lost in the Mo. This is observed in Figs. 3(a) and 3(b) for CIGSe thickness of $0.5\ \mu\text{m}$. All the light that is not absorbed by the CIGSe is absorbed by the Mo such that the total absorption by the cell remains the same. Based on our calculations, one can observe a gain in light absorption when further replacing the Mo back reflector by gold [see Fig. 2(c) for thick CIGSe and Fig. 3(c) for thin with total absorption

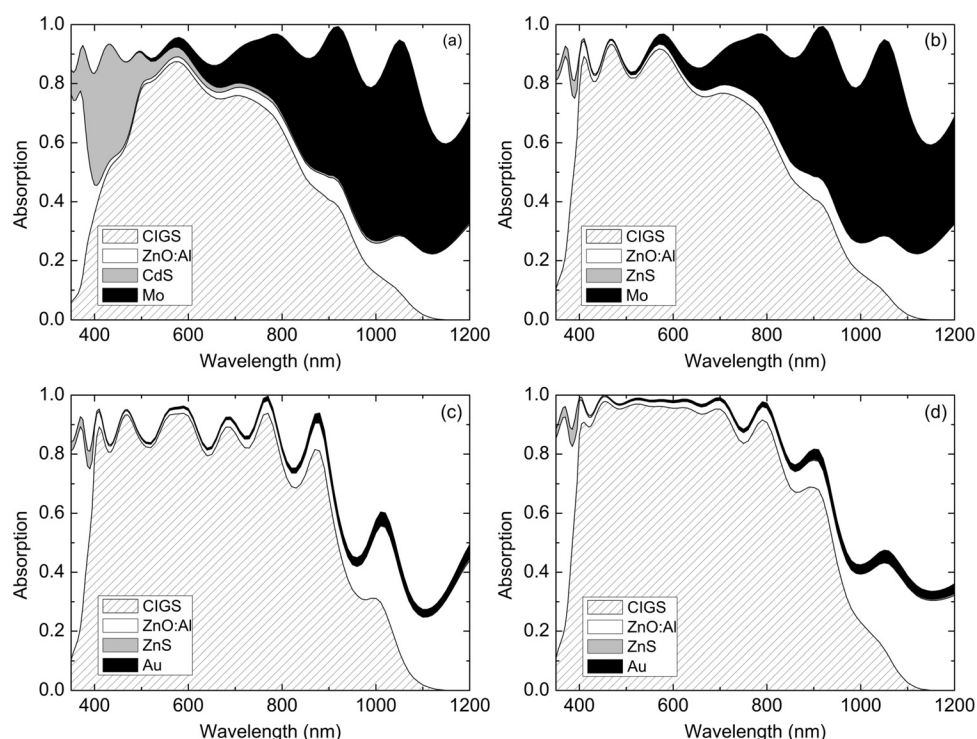


FIG. 3. As Fig. 2 with CIGSe thickness of $0.5\ \mu\text{m}$. (a) Sample (a) in Table I; $A = 57\%$. (b) Sample (c) in Table I; $A = 63\%$. (c) Sample (e) in Table I without ARC; $A = 71\%$. (d) Sample (e) in Table I; $A = 76\%$.

$A = 71\%$]. Although the reflection by gold is lower than by silver, it is still more interesting to use gold due to its chemical stability such that no protective layer is required. A third issue is the portion of light that is absorbed by the front contact, the ZnO:Al and i-ZnO layers. This absorption can be reduced by replacing the ZnO:Al with a more transparent material; however, finding an electrical conductor that is also transparent for light in the visible range is a complex issue. Despite its absorption, the front contact can serve as a substrate for antireflection layer, e.g., MgF_2 of 100 nm thick. Consequently, more light is transmitted into the cell and eventually increases the absorption in the CIGSe by an additional 5% [Fig. 2(d) and Fig. 3(d) for thick and thin CIGSe layer, respectively]. To summarize, the total absorption is increased from 57% in the reference configuration [sample (a)—thin CIGSe] up to 76% in configuration of sample (e). For comparison, in the thick cell, the same modifications lead to an increase from 69% [Fig. 2(a)] to 83% [Fig. 2(d)]. Since the effects of the front and buffer contacts are similar for thick and thin solar cells, it is noticeable that the difference in the total absorption comes from changing the back contact.

The short circuit current can be calculated with the following relation to the spectral absorption in the CIGSe layer:

$$J_{sc} = q \int A_\lambda(0; m_{\text{CIGSe}}) I_{\text{solar}}(\lambda) \frac{\lambda}{hc} d\lambda, \quad (4)$$

where q is the electron charge, $A_\lambda(0; m_{\text{CIGSe}})$ is the spectral absorption in the layer of the CIGSe when illuminating in normal incident angle, and hc/λ is the energy per wavelength. In this notation, $I_{\text{solar}}(\lambda)$ is given in units of $\text{W}/(\text{m}^2 \text{ nm})$. The integration is over all the solar spectrum or over the spectra where CIGSe absorbs. Herein, we used the limits as shown in the figures, from 350 nm to 1200 nm. In this equation, a collection efficiency of unity is assumed, i.e., all the absorbed photon contribute to electrical current. Their values for each configuration are summarized in Table I, for a CIGSe thickness of $0.5 \mu\text{m}$.

It is well known that a reduction of reflection through index matching between the air and the cell can be achieved either by encapsulation by glass or deposition of MgF_2 layer. Furthermore, a similar effect can be achieved by texturing the surface of the front contact.¹⁶ The textured surface can be modeled as a medium with an effective index of refraction if the diffuse light is negligible. Its value is between the refractive index of the substrate (ZnO:Al) and of air according to the ratio between them, which is the geometrical filling factor f . Note that in general, it may depend on the height of roughness, i.e., $f = f(z)$. Here, we use the Bruggeman method to calculate the effective index by solving the following equation¹⁷

$$f(z) \frac{\varepsilon_1 - \varepsilon_{\text{eff}}(z)}{\varepsilon_1 + 2\varepsilon_{\text{eff}}(z)} + [1 - f(z)] \frac{\varepsilon_2 - \varepsilon_{\text{eff}}(z)}{\varepsilon_2 + 2\varepsilon_{\text{eff}}(z)} = 0, \quad (5)$$

where $\varepsilon_j = (n_j + ik_j)^2$ is the dielectric constant of medium 1 (air) and medium 2 (ZnO:Al). $f(z)$ is obtained by measuring the surface profile of the solar cell. A similar procedure was

reported by Stephens and Cody where they modeled the textured surface by graded density of particles.¹⁸ Then, the textured surface is modeled as a stack of multilayer structure, each layer with homogeneous refractive index $n_{\text{eff}} = n_{\text{eff}}(z)$. Finally, this effective medium is implemented on top of the ZnO:Al layer and the absorption in the different layer of the cell is calculated using the transfer matrix method. We calculate the spectral absorption of the cell (derived by the reflection, $1 - R$) to check that the reflection is actually reduced. As can be seen in Fig. 4(a), the textured interface functions as an antireflection layer in a broad spectral range.

IV. EXPERIMENTAL RESULTS AND DISCUSSIONS

In this part, cells with the four different configurations suggested above are studied [samples (a)–(d) in Table I]. For these cells, we compared the theoretical and experimental improvement of the short circuit current when the CIGSe thickness is reduced down to $0.3 \mu\text{m}$. The theoretical J_{sc} were calculated using Eq. (4) for each thickness. We remind that only the optical absorption in the CIGSe layer is accounted for. The experimental J_{sc} were measured using the solar simulator, as mentioned in Sec. II. In order to assess the improvements obtained by each modification (antireflection, buffer layer, and back contact), we normalized the short circuit current values with their reference cell, i.e., sample (a) with a $2.5 \mu\text{m}$ thick CIGSe layer.

A. Effect of the front contact

Our aim in this subsection is to show that fine textured surface behaves as antireflection layer. In order to compare optical modeling and measurements, we have measured the reflectivity by cells with CIGSe thickness of $0.5 \mu\text{m}$ with flat and textured front contacts. We show in Fig. 4(b) the spectral

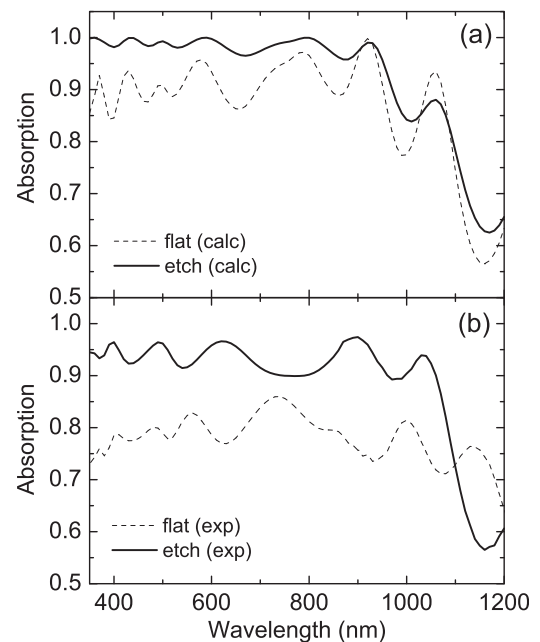


FIG. 4. (a) Calculated and (b) measured absorption by a solar cell with $0.5 \mu\text{m}$ CIGSe thickness. Solid and dash lines denote absorption by textured and flat ZnO:Al front contact, respectively.

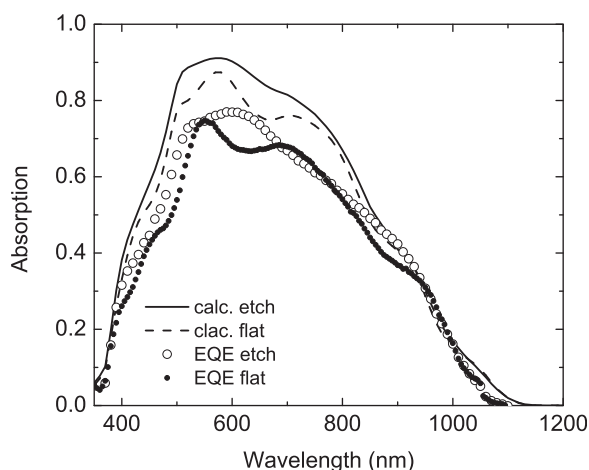


FIG. 5. EQE (dots) and calculated absorption (lines) in $0.5\ \mu\text{m}$ thick CIGSe layer. Comparison between flat and textured (etch) ZnO:Al interface.

absorption by the cells (derived by $1 - R$). As shown by the experiment and the calculation, a cell with a textured surface absorbs more light in all the spectral range, up to a $1100\ \text{nm}$ wavelength. For the same solar cells, we compare in Fig. 5, the calculated absorption in the active layer and the EQE. A good agreement is obtained in the curve shapes and spectral peaks. In order to evaluate experimentally the effect of ARC (or protective glass, for this purpose) and textured surface on the absorption in the active layer, we measure the EQE for solar cells with flat interface, ARC, and textured ZnO:Al. In addition, for each of these configurations, we include measurements for $2.5\ \mu\text{m}$ and $0.5\ \mu\text{m}$ CIGSe absorber. The results shown in Fig. 6(a) correspond to a flat interface with or without ARC. In Fig. 6(b), textured ZnO:Al is compared with flat interface (without ARC). We observe that the effect of

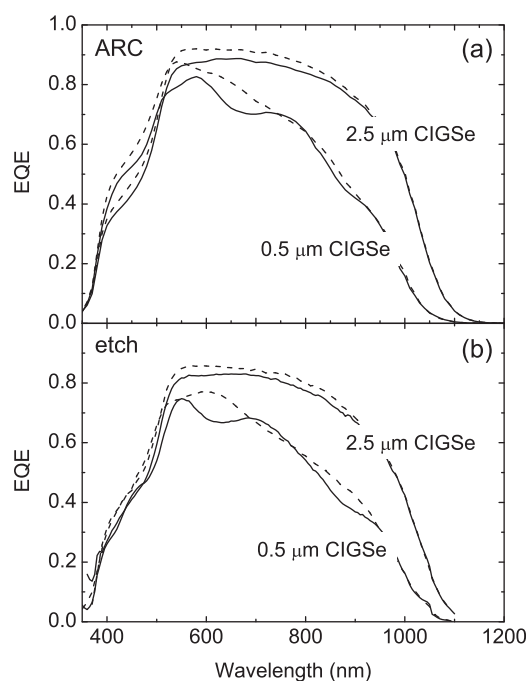


FIG. 6. Comparison of experimental EQE of $0.5\ \mu\text{m}$ and $2.5\ \mu\text{m}$ CIGSe based solar cells with (a) antireflection layer and (b) ZnO:Al textured layer. Dash lines present the gain with ARC or textured ZnO:Al.

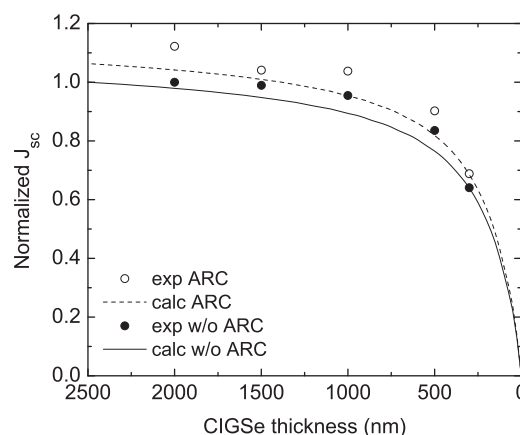


FIG. 7. Short circuit current versus CIGSe thickness normalized by the reference cell [sample (a) with CIGSe thickness of $2.5\ \mu\text{m}$]. Experiments deduced from J-V measurements under AM1.5 illumination for solar cells before (filled circles) and after (open circles) ARC. Calculations derived from Eq. (4) are given by lines.

textured ZnO:Al structure is indeed similar to that obtained by the ARC, i.e., less light is reflected due to a graded refractive index. Furthermore, the effect on a thin or a thick active layer is almost the same, as pointed out in Sec. III. In addition, the short circuit current of solar cells with ARC [sample (b)] and flat interface are compared in Fig. 7 for absorber thickness between $0.3\ \mu\text{m}$ and $2.5\ \mu\text{m}$. The J_{sc} is improved by 5 to 10% for all thicknesses larger than $0.5\ \mu\text{m}$. These improvements show good agreement with our theoretical predictions (error of 5%).

B. Influence of the buffer layer

We have replaced CdS/i-ZnO buffer layer by ZnS/ZnMgO in our solar cells, sample (c) in Table I. CBD-CdS and CBD-ZnS buffer layers are distinguished by their bandgap energy.¹⁹ The bandgap of i-ZnO and ZnMgO layers has also been measured showing a higher bandgap of $\text{Zn}_{0.74}\text{Mg}_{0.26}\text{O}$ (3.8 eV) compared to that of i-ZnO (3.2 eV). For our calculations, we considered that the optical properties of ZnMgO are similar to those of i-ZnO in the spectra beneath the bandgap energy.²⁰ Due to the higher transmission of the ZnS/ZnMgO, the Cd-free cells show significantly enhanced quantum efficiency in the spectral range between 350 nm and 550 nm wavelengths.^{10,11} At the absorption edge of the CIGSe, the EQE of the Cd-free cells is identical to the one with CdS buffer down to a 500 nm wavelength, which indicates that the collection length is not affected by replacing the buffer. This explains the higher J_{sc} values obtained for cells with ZnS buffer layers. A comparison of the experimental and calculated J_{sc} for solar cell thicknesses between $2.5\ \mu\text{m}$ and $0.3\ \mu\text{m}$, with CdS and ZnS buffer layers, shows an improvement of about 7% for all thicknesses down to $0.5\ \mu\text{m}$ thick CIGSe (cf. Fig. 8).

C. Influence of the back contact

A cell with a gold back contact and a ZnS buffer layer could not be accomplished for the moment due to electrical contact difficulties. To overcome this problem, we study

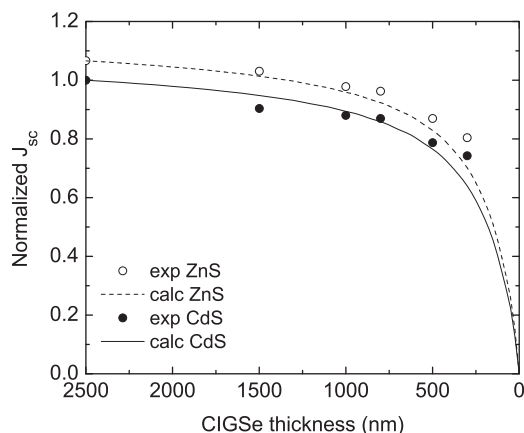


FIG. 8. As Fig. 7 when replacing the buffer layer from CdS to ZnS.

only the optical effect of the back contact [sample (d)]. Using gold as a back contact in ultrathin CIGSe solar cell has been realized by combining chemical etching and lift-off process, as presented elsewhere.¹² Therein, we observed that the Au back contact does not degrade the other parameters of the cell, and thus a good ohmicity on CIGSe is achieved as well as good reflectivity in the IR spectra. We investigate a solar cell with 300 nm thick gold back contact, and different absorber thicknesses between 0.4 μm , and 2 μm . Although there might be differences in electrical contacts between thin and thick CIGSe layers, as was reported in Ref. 12, we do not consider them here and compare the measured J_{sc} only with optical absorptions. In Fig. 9, short circuit current as a function of the absorber thickness is compared for solar cells with Au and Mo back contacts [samples (a) and (d) in Table I]. We observe that the J_{sc} difference between solar cells with Au or Mo increases as the absorber thickness is smaller. The influence of the back contact reflectivity becomes pre-eminent for CIGSe thicknesses lower than 1.2 μm , with 20% difference at 0.4 μm CIGSe thickness. However, the J_{sc} difference for thick absorber ($>1.2 \mu\text{m}$) cannot be explained by the reflectivity of the back contact. We suggest that it is due to the differences in the etching process between the two type of solar cells [front etch for sample (a), front and back etch for sample (d)]. This process leads to two main differences. The first is the necessity of

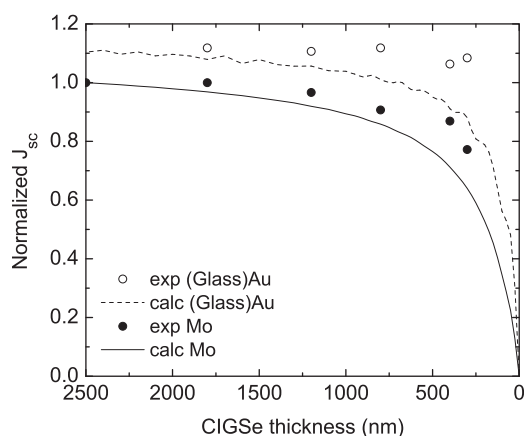


FIG. 9. As Fig. 7 when replacing the back contact from Mo to Au. In addition, the solar cell with gold back contact includes glass superstrate.

glass as a substrate on the front contact in the back etching process. Its influence is similar to the use of an ARC due to the graded refractive index. The second reason is originated by the graded composition of the CIGSe layer with a bandgap energy difference of +50 meV between its front and back sides.⁶ In our case, it means that CIGSe after front etching [sample (a)] has a 50 meV higher bandgap than CIGSe obtained by back etching [sample (d)]. This bandgap difference is materialized by $\Delta J_{sc} \approx 1 \text{ mA/cm}^2$. It is noticeable that the J_{sc} is maintained at a high value with Au back contact, with an improvement of 20% for thin 0.4 μm CIGSe solar cell. An efficiency of about 10% was achieved for cells with CIGSe thickness larger than 0.4 μm .¹²

V. CONCLUSION

An effective integration of both fundamental understanding and innovative processing developments is the key to efficient photon manipulations for ultrathin solar cell applications. Based on theoretical and experimental studies, we show that enhanced absorption in the active layer is achieved by minimizing the parasitic optical losses in the buffer and the back contact layers. As a result, the short circuit current can be theoretically improved by 32%, particularly for thin film down to 0.5 μm thickness. This modeling is reinforced by experimental results when replacing the buffer layer and the back contact, separately. Moreover, we point out that the measured J_{sc} at ultrathin cells (CIGSe thickness lower than 0.5 μm) are higher than the values anticipated from modeling. These enhancements should be further investigated. Consequently, if sample (a) with absorber thickness of 0.5 μm yields $J_{sc} = 23 \text{ mA/cm}^2$, based on our calculations we expect sample (e) to produce 30 mA/cm^2 . This value is comparable to commercial cells—sample (a) with CIGSe thickness of 2.5 μm and rough interface. Since these results are very encouraging, we plan as a next step to include all improvements in a single solar cell.

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