

Light trapping in ultrathin $\text{CuIn}_{1-x}\text{Ga}_x\text{Se}_2$ solar cells by dielectric nanoparticles

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ABSTRACT

In this work, light trapping effects based on dielectric nanoparticles (NPs) are numerically evaluated in ultrathin $\text{CuIn}_{1-x}\text{Ga}_x\text{Se}_2$ (CIGSe) solar cells for different locations of NPs, cell architectures and illumination directions, with relevant implications and optimized NP parameters being specified. The severe absorption in Mo is the main constraining factor for the effective implementation of light trapping NPs in ultrathin CIGSe cells. For a significant light absorption enhancement, it is favoured to integrate dielectric NPs at the rear interface of CIGSe/back contact and employ transparent conductive oxide (TCO) back contacts rather than the conventional Mo. It is demonstrated, that under front illumination, the low-index ($n = 1.5$) hemispherical NPs at the CIGSe/TCO interface cause significant light trapping effects. The NP-patterned ultrathin cells achieve a maximum short circuit current density (J_{sc}) of 36.4 mA/cm^2 at an upper limit of 500 nm CIGSe thickness, which is as high as 94% J_{sc} of their thick flat counterparts with a CIGSe thickness of 2000 nm. In contrast, under back illumination, the patterned ultrathin cells realize comparable absorption to the corresponding thick counterpart at a CIGSe thickness of only 300 nm and the maximum J_{sc} (35.2 mA/cm^2) saturates at a CIGSe thickness of 425 nm. Further, J_{sc} is less attenuated by the parasitic absorption in Al:ZnO (AZO) under back illumination than under front illumination in module configuration, where a much thicker AZO is required. This suggests that patterned ultrathin CIGSe solar cells under back illumination will be a promising cell architecture simultaneously for high efficiencies and less material usage in industrial module production.

1. Introduction

Photovoltaics, which convert sunlight to electricity, are a promising technology to address the environmental problems arising from burning fossil fuels by providing renewable and clean energy. Compared to the conventional bulk crystalline Si solar cells in the market, thin-film solar cells hold the promise to be more competitive, because they allow the reduction of material consumption and resulting manufacturing cost as well as improved flexibility in roll-to-roll manufacturing on bendable substrates. Among various thin-film solar cell technologies, $\text{CuIn}_{1-x}\text{Ga}_x\text{Se}_2$ (CIGSe) solar cells are unique for their high record efficiencies of 22.6% (<https://www.zsw-bw.de>, 2016). Furthermore, CIGSe solar cells offer a remarkably short energy payback time and a great tolerance towards environmental influences like shading and illumination intensity variations (CIGS community, 2016). Despite these achievements and advantages, further cost reduction is desirable for driving mass production and large-scale deployment of CIGSe solar cells. Presently, the raw materials induced cost accounts for 50% of the module

production (CIGS community, 2016) and In scarcity is likely to be a bottleneck when envisaging mass production (Tao and et al., 2011; Fthenakis, 2012). To address the problems, one key approach is to thinning the CIGSe photoactive layers from typical 2–3 μm to below 500 nm. However, this will cause incomplete absorption, giving rise to a significantly reduced short circuit current density (a loss of more than 6 mA/cm^2 in J_{sc}) and poor cell performance (Gloeckler and Sites, 2005; Jehl et al., 2011; Lundberg et al., 2003; Yin et al., 2014). Therefore, light trapping is crucial to maintain high efficiencies of ultrathin CIGSe cells (with a sub-500 nm absorber thickness).

Metallic nanoparticles (NPs) have been extensively explored for absorption enhancement in various types of solar cells, because they are able to exhibit plasmonic resonances allowing large-angular scattering and strong near-fields (Catchpole and Polman, 2008; Spinelli and Polman, 2012; Tan et al., 2012; Atwater and Polman, 2010; Ferry et al., 2008). Yet, plasmonic NPs suffer from intrinsic parasitic absorption, limiting their potential for maximum absorption enhancement in photovoltaic devices (Ferry et al., 2008, 2010; Schmid et al., 2014). More

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critically, plasmonic materials (e.g. Ag, Au) are not thermally compatible with the high-temperature CIGSe fabrication (Yin et al., 2015).

In recent years, wavelength-scale dielectric NPs have also gained tremendous attention in light trapping due to their strong scattering ability (relative to metallic ones) while being absorption free (Spinelli et al., 2012; Brongersma et al., 2014; Mann et al., 2011; Wang et al., 2014). Furthermore, inorganic dielectric materials (e.g. SiO₂, Al₂O₃) are thermally stable, which permits their experimental integration into CIGSe cells. Additionally, certain dielectric materials were proved to contain intrinsic charges which can electrically benefit solar cells (Hoex et al., 2007; Moors et al., 2012; Vermang et al., 2013). These distinctive features favour dielectric NPs as a light trapping tool for ultrathin CIGSe cells.

Compared to the intensive investigation in Si-based solar cells, the application of dielectric NPs to CIGSe solar cells for absorption enhancement is just emerging (Yin et al., 2016a; Lare et al., 2015; Yin et al., 2016b, 2016c; Salome et al., 2017). Though initial work has realized absorption enhancement to varied success, no systematic research has been performed to identify how to effectively implement light trapping NPs in ultrathin CIGSe solar cells. Plus, CIGSe solar cells have a distinctively different cell architecture and opto-electronic properties from Si-based cells. Those suggest that the effective implementation of light trapping NPs in ultrathin CIGSe solar cells may not be easily generalized from similar technologies.

In this contribution, we start from analyzing optical losses in ultrathin CIGSe solar cells. To identify the potential of NPs in ultrathin CIGSe solar cells from the perspective of absorption enhancement, light trapping effects of dielectric hemispheres for different locations of NPs, cell architectures, illumination directions, are comprehensively evaluated with relevant implications and optimized NP parameters being specified.

2. Simulation

The theoretical CIGSe solar cell model is based on our lab-scale architectures with a typical structure of Al:ZnO(AZO)/ZnO/CdS/CIGSe/back contact/substrate from top to bottom. For the window and buffer layers (AZO/ZnO/CdS), the thicknesses are 240/130/50 nm. The back contact with a thickness of 200 nm is Mo or alternatively Sn:In₂O₃ (ITO) and a sheet resistance < 10 Ω/sq. The CIGSe layer here corresponds to a Ga/[Ga + In] ratio of 0.35 and a minimum bandgap of 1.05 eV. The input optical constants (refractive index *n*, extinction coefficient *k*) of each individual layer (except Mo) are extracted by transfer-matrix method (Yin et al., 2013). Mo data was obtained using spectroscopic ellipsometry. The optical constants for the individual layers of a CIGSe solar cell are plotted in supporting Fig. S1.

For optical simulations on flat CIGSe solar cells, an analytical program based on transfer-matrix method (RefDex) is used (<https://www.helmholtz-berlin.de>). To understand the optical influences of NPs on ultrathin CIGSe solar cells, we perform finite element method (FEM) simulations using the software package JCMsuite (<http://www.jcmwave.com/>). In all optical simulations, interface roughness is not taken into account, but we don't expect that the overall trend will be substantially influenced since the interface roughness is typically low in experimental samples. For overall comparison, reflection (*R*), absorption (*Abs*) and transmission (*T*) are converted into current density *J*, assuming the complete conversion under standard AM 1.5 solar irradiation. The current density out of absorption in the CIGSe layer (*Abs_{CIGSe}*) is specially referred to as short circuit current density *J_{sc}*.

3. Identification of optical losses

To identify optical losses in ultra-thin CIGSe solar cells, we start from the simulations of flat devices and compare optical responses (*R*/*T*/*Abs*) for cells – with varied absorber thicknesses (*d_{CIGSe}*) as well as different back contacts. The detailed cell structures are listed in Table 1.

Table 1

Cell structures for identifying optical losses in ultrathin CIGSe solar cells.

No.	Structure (from top to bottom)	<i>d_{CIGSe}</i> (nm)
N1	AZO/ZnO/CdS/CIGSe/Mo/glass substrate	2000 (thick)
N2	AZO/ZnO/CdS/CIGSe/Mo/glass substrate	400 (ultrathin)
N3	AZO/ZnO/CdS/CIGSe/Ag/glass substrate	400
N4	AZO/ZnO/CdS/CIGSe/ITO/glass substrate/Ag mirror	400

Amongst all the layers in CIGSe solar cell devices, only the absorption in the CIGSe absorber layer (*Abs_{CIGSe}*) contributes to the effective photocurrent, *R*, *T* and *Abs* in other layers are lost. We start from thick cells on a conventional Mo back contact at *d_{CIGSe}* = 2000 nm (N1) and its optical responses (*R*/*T*/*Abs*) are plotted in Fig. 1(a). The weights of *R*/*T*/*Abs* are also labelled in terms of $\frac{J(Abs/R/T)}{J(AM1.5)}$ in Fig. 1(a). It can be observed that *Abs_{CIGSe}* dominates the spectrum of interest and the optical losses are sub-divided into 3 sections: *R*, *Abs* in AZO/ZnO/CdS (*Abs_{top}*) and back contact (*Abs_{back}*). *R* loss covers the whole spectrum of interest. The parasitic *Abs_{top}* dominates the absorption in the visible range (< 550 nm) due to inter-band absorption of AZO/ZnO/CdS. As to *Abs_{top}* in the long wavelength range, this is related to free carrier-induced absorption in AZO. Since AZO/ZnO/CdS layers are on top of the CIGSe layer, *Abs_{top}* is inevitable. The parasitic *Abs_{back}* starts from the wavelength where light is not completely absorbed by the CIGSe layer and is negligible for thick cells.

For comparison, $\frac{J(Abs/R/T)}{J(AM1.5)}$ shares for *d_{CIGSe}* = 400 nm (N2) are studied and listed in Fig. 1(b). As observed, the ultrathin cells feature increased *Abs_{back}* and *R* at a cost of a dramatic drop in *Abs_{CIGSe}*, and *Abs_{back}* is becoming the main optical loss. It accounts for a share of 16.3%, whereas the corresponding value in the thick cells (N1) is only 2.5%. This can be interpreted due to the poor reflective ability of Mo: light reaching Mo will be mainly absorbed by Mo rather than being reflected back into CIGSe (Onwudinanti et al., 2016; Krc et al., 2016). As the CIGSe thickness reduces, more light hits Mo and the parasitic absorption in Mo thereby increases. For the increase in *R*, this is related to the increased intensity of light escaping ultrathin solar cells out of incomplete absorption in the absorber, which mainly lies in the infrared range.

The ratios, $\frac{J(Abs/R/T)}{J(AM1.5)}$ for ultrathin cells on Ag back contact with nearly perfect rear reflectivity (N3) are also shown in Fig. 1(b). The parasitic *Abs_{back}* reduces to a negligible level and the share *Abs_{CIGSe}* accounts for significantly enhances from 62.6% to 70.7% compared to the cell on Mo (N2), despite of being still lower than the value of thick cells N1. Unfortunately, the direct implementation of metallic materials (Ag, Al, Au) having much better reflectivity as back contact is not experimentally feasible due to their thermal incompatibility with high-temperature CIGSe fabrication. Lifting off and transferring solar cells from Mo onto Au back contact for improving interface reflectivity has been experimentally realized (Li-kao et al., 2012). However, the lift-off process is applicable to the size of a few cm² and the absorption enhancement is limited by only a double optical path. An alternative back contact, which is experimentally feasible and exhibits low absorption ability, are the transparent conductive oxides (TCOs) (Young et al., 2002; Nakada, 2005; Nakada et al., 2004). Their back interface reflectivity is also poor, which can however be overcome by a reflector at the rear side of the glass substrate. $\frac{J(Abs/R/T)}{J(AM1.5)}$ fractions for ultrathin cells on ITO/glass/Ag back reflector (N4) are listed in Fig. 1(b) as well. The parasitic *Abs_{back}* (both ITO and Ag mirror) takes a small portion of 5.1% and is greatly lower than 16.3% in the case of Mo back contact. As a result, the *Abs_{CIGSe}* fraction improves to 68.3%, which is nearly 6% higher than the value on Mo (N2).

In conclusion, the parasitic *Abs_{back}* is the main optical loss for ultrathin cells on Mo back contact. This indicates that, in order to obtain a comparable *Abs_{CIGSe}* to their thick counterparts, ultrathin CIGSe solar cells should reduce light intensity reaching Mo into a negligible level,

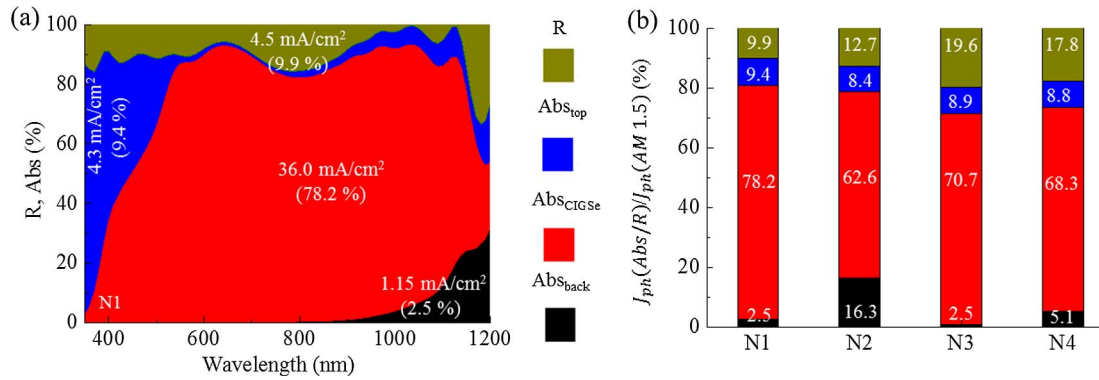


Fig. 1. (a) Optical responses ($R/T/Abs$) of thick CIGSe solar cells at $d_{CIGSe} = 2000$ nm (N1), (b) weights of each individual response $\frac{J_{ph}(Abs/R)/J_{ph}(AM\ 1.5)}$ of cell architectures in Table 1.

otherwise a dramatic Abs_{back} is inevitable. For ITO/glass/Ag mirror back contact, ultrathin cells allows light hitting back contact in multiple passes, which mitigates the requirements for light trapping NPs. It should be noted here that CIGSe solar cells normally exhibit inferior electrical performance on TCO than on Mo due to the mismatch of work function between CIGSe and TCO. However, recent reports have shown that inserting a few nm thick hole transporting layer can address this problem, holding the promise to utilize the advantages of TCO back contacts (Larsen et al., 2014; Simchi et al., 2014).

4. Light trapping NPs on top

For the stability consideration of electrical properties in photovoltaic devices, light trapping NPs are typically placed out of the p-n junction, in many cases on top of the cells. Light trapping effects from the top can be generally decomposed into improved coupling (R reduction) and large angular scattering. However, previous studies of light trapping nanostructures on top of CIGSe solar cells (e.g. closely packed low-index SiO_2 nanospheres (Yin et al., 2016a) and high-index TiO_2 nanopatterns (Lare et al., 2015) and textured AZO (Dahan et al., 2012)), have ended with a modest J_{sc} enhancement, showing an optimum light trapping effect comparable to the anti-reflection effect from a conventional MgF_2 layer. The underlying reasons are related to the specific cell architecture of CIGSe. On top of the photoactive CIGSe layer, there are three top layers (AZO/ZnO/CdS). According to Snell's law, scattering will suffer an angular shrinkage due to the continuously increasing refractive indices from AZO to CIGSe, which thus constrains the optical path enhancement in the CIGSe layer. For example at $\lambda = 900$ nm, as shown in Fig. 2, a scattering angle as high as 70° in AZO

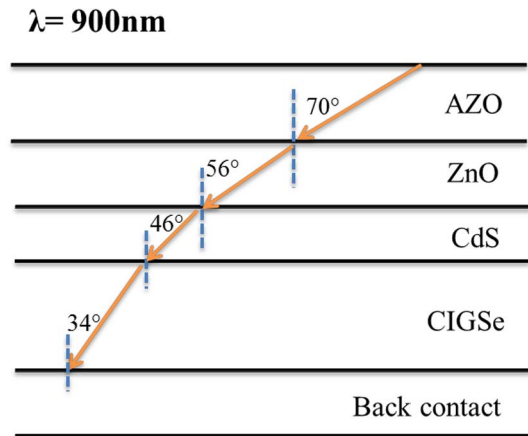


Fig. 2. Sketch of angle shrinkage of light ($\lambda = 900$ nm) scattered from top: Based on Snell's law, the refractive indices gradually increasing from AZO to CIGSe reduce the scattering angle of 70° (in AZO) to 34° (in CIGSe).

shrinks sharply to 34° in CIGSe. This corresponds to an optical path of only $1.2d_{CIGSe}$ in a single propagation and is much shorter than the absorption length at $\lambda = 900$ nm. Further, AZO/ZnO/CdS layers exhibit broadband parasitic absorption (see Fig. 1(a)), light passing through AZO/ZnO/CdS layers prior to the CIGSe absorber will attenuate the benefit of improved coupling as a result of a prior increase in Abs_{top} , which is aggravated by the prolonged optical path arising from angular scattering. This issue is particularly severe for cells on Mo back contact: on the one hand large angular scattering is required for a significant Abs_{CIGSe} enhancement in a single pass, this will on the other hand induce an increase in Abs_{top} and R , thus constrains or even worsens Abs_{CIGSe} .

Another disadvantage for light trapping NPs on top is, since light is propagating from a low-index material (AZO) to a high-index medium (CIGSe), scattering angles in CIGSe can not be beyond the critical angles and thus the possibility to trap light in CIGSe is lost. Combined with the above-mentioned attenuation points of light trapping effects from top, obtaining a significant Abs_{CIGSe} enhancement and achieving a comparable Abs_{CIGSe} to thick CIGSe cells is quite challenging for ultrathin cells with NPs on top.

5. Light trapping NPs at CIGSe/back contact

5.1. Front illumination

Placing light trapping NPs at interfaces touching absorbers allows light trapping effects to be directly utilized by CIGSe absorbers and avoid the attenuation effects from the top layers, a similar phenomenon was also observed in Si solar cells (Sanchez-Sobrado et al., 2017). Compared to the critical p-n junction interface at CdS/CIGSe, it is electrically favourable to place light trapping NPs at CIGSe/back contact. 3D FEM simulations using JCMwave are performed to study optical behavior of hemispherical NPs in ultrathin CIGSe solar cells. Fig. 3(a) shows the sketch of patterned CIGSe solar cells with NPs at CIGSe/back contact interface under front illumination. An anti-reflection MgF_2 layer with a thickness of 110 nm is added on top to reduce the Fabry-Pérot interferences. In the FEM simulation, a hexagonal computational domain with three sets of periodic boundary conditions in the x-y plane and perfectly matched layer (PML) boundary conditions in the z direction are used. A plane wave source is used incident anti-parallel to the z axis, in order to simulate light incident from above the solar cell. To be comparable to the experimental morphology, the normal growth (Sever et al., 2013) of layers over NPs is assumed considering the NP incurred deformation of layers on top and the volume of individual layer is adjusted equally to the corresponding volume in the configuration of the flat reference. The shape of dielectric NPs is confined to represent a hemisphere, since we also studied other commonly used shapes like cylinder, conical frustum and sphere and they exhibited comparable light absorption enhancement. They are in a hexagonal order with a radius of r and an edge-to-edge distance l as shown

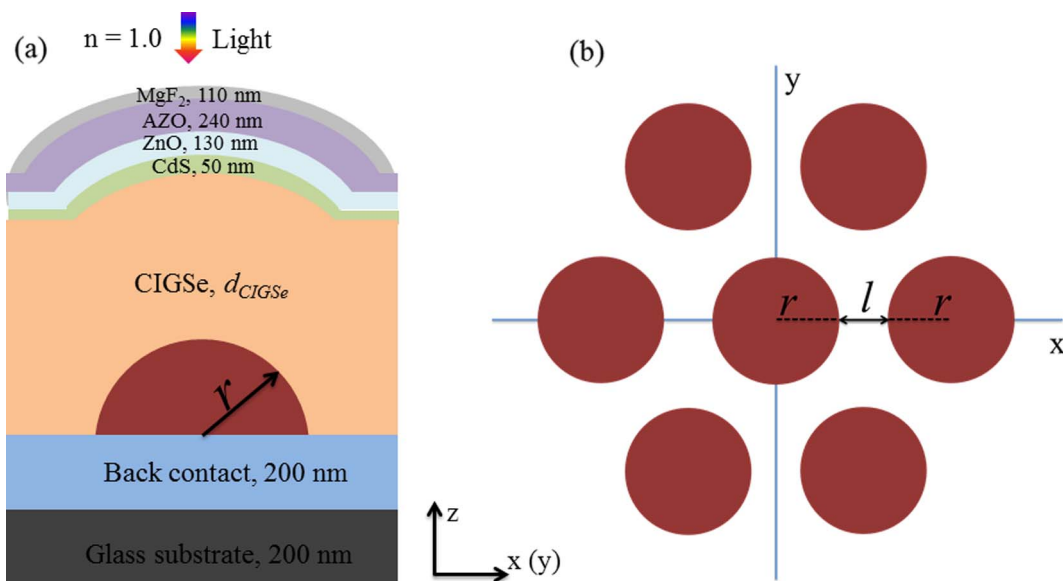


Fig. 3. (a) Sketch of hemisphere-patterned CIGSe solar cells under front illumination: the hemispheres are located at CIGSe/back contact interface, the layers on top of the hemisphere have a conformal growth. (b) Top view of the hexagonally arrayed hemispheres at the interface of CIGSe/back contact with a radius r and an edge-to-edge distance l .

in Fig. 3(b). To calculate the absorption in the layers of solar cells, the total field volume integration inside these individual layers is used. The unit simulation mesh structure is shown in supporting Fig. S2. The index of NPs is set to 1.5, which is comparable to the thermally stable SiO_2 and Al_2O_3 and exhibits large index-contrast to CIGSe absorbers (~ 2.8).

We start from ultrathin cells on the conventional Mo back contact and Fig. 4(a) compares R/Abs of NP-patterned solar cells (solid lines) to their flat counterparts (without NPs, dashed lines) at $d_{\text{CIGSe}} = 400$ nm and $(r, l) = (275, 75)$ nm. The (r, l) geometry parameter is within the optimum geometry range, which will be shown later. For flat cells, $\text{Abs}_{\text{CIGSe}}$ (red dashed line) reaches a maximum value of 95% at 550 nm and then incomplete absorption leads to a drop in $\text{Abs}_{\text{CIGSe}}$ for longer wavelengths. Remarkably, the patterned cell shows a pronounced $\text{Abs}_{\text{CIGSe}}$ enhancement (red dashed to red solid line) in the poor absorbing wavelengths, yielding a J_{sc} enhancement of 3.2 mA/cm^2 . This is mainly as a result of Abs_{Mo} reduction (green dashed to green solid line). Furthermore, the R spectrum overall reduces after patterning and also contributes to the $\text{Abs}_{\text{CIGSe}}$ enhancement. Abs_{top} does not significantly change upon the patterning and is not shown here.

Even the major optical loss Abs_{Mo} is substantially reduced after patterning, the remaining loss (green solid, 5.3 mA/cm^2) is still severe, which limits further $\text{Abs}_{\text{CIGSe}}$ enhancement. Actually, it is quite challenging to completely eliminate Abs_{Mo} even by optimizing geometries of

NPs, the reasons will be mentioned later. As stated above, TCO back contacts exhibit less absorption ability and thus render the potential of a lower Abs_{back} . For comparison, Fig. 4(b) shows the plot of $R/T/\text{Abs}$ between flat and patterned cells on ITO back contact. For flat cells, Abs_{ITO} (green dashed, Fig. 4(b)) is around 10%, whereas Abs_{Mo} (green dashed, Fig. 4(a)) shows up to 30% beyond 900 nm in wavelength. T (blue dashed) is becoming the main optical loss and shows up from the wavelength around 500 nm where incomplete absorption starts and is significantly increasing as the wavelength goes up. With the addition of NPs, similar to the case on Mo, cells on ITO exhibit a great $\text{Abs}_{\text{CIGSe}}$ enhancement (3.6 mA/cm^2) with a concomitant decrease in T from 8.0 to 3.9 mA/cm^2 and in Abs_{ITO} from 1.3 to 1.1 mA/cm^2 . Overall, NP patterning gives a nearly equal $\text{Abs}_{\text{CIGSe}}$ between cells on ITO and on Mo. However, the remaining T (3.9 mA/cm^2) for the patterned cells on ITO can be further utilized to a great extent by a reflector (e.g. Ag film, TiO_2 paste) at the rear side of glass substrate, which indicates that solar cells on ITO hold the potential to have a higher $\text{Abs}_{\text{CIGSe}}$. Due to this, we will limit the following investigations to cells on ITO.

We note that there are four pronounced $\text{Abs}_{\text{CIGSe}}$ enhancement peaks ($\lambda = 820, 910, 1040, 1090$ nm) for the patterned cells as the arrows indicate in Fig. 4(b). A batch of simulations are done by tuning the edge-to-edge distance l from 25 nm to 75 nm and the four resonance peaks are characterized by a linear red-shift in wavelength as l (or pitch, $2r + l$) is enlarged (see supporting Fig. S3). This suggests that

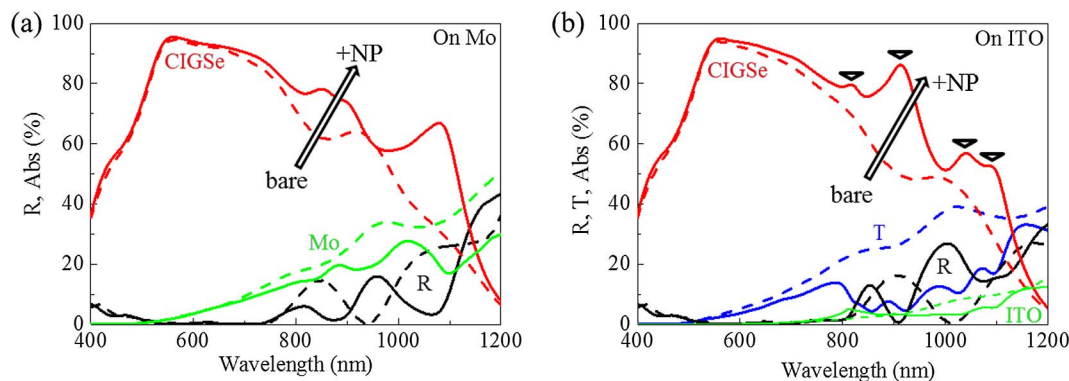


Fig. 4. Optical responses (R/Abs) between flat (dashed lines) and patterned (solid lines) CIGSe solar cells (a) on Mo and (b) on ITO at $d_{\text{CIGSe}} = 400$ nm and $(r, l) = (275, 75)$ nm under front illumination.

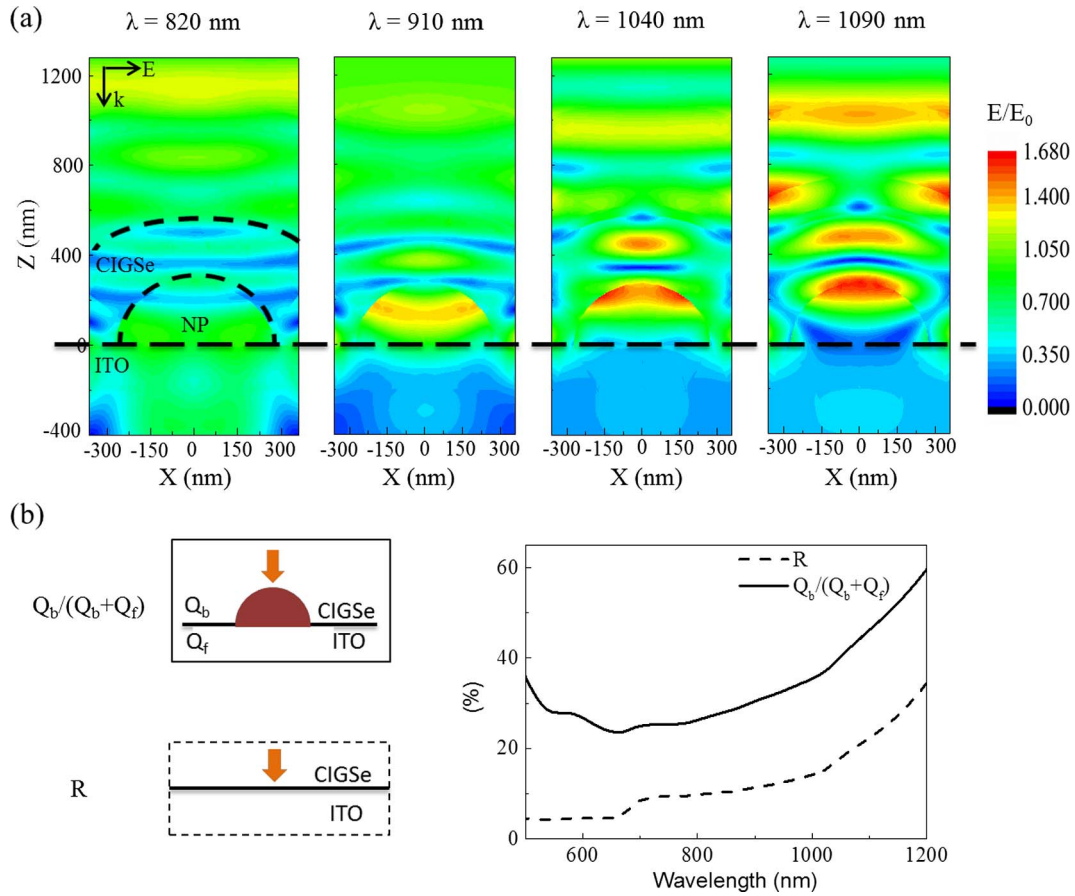


Fig. 5. (a) Near field distributions of patterned CIGSe solar cells on ITO at $\lambda = 820, 910, 1040, 1090$ nm, (b) comparison of interface reflectivity and backward scattering efficiency ($Q_b/(Q_b + Q_f)$) of a single NP at a CIGSe/ITO interface. Absorption ability for both CIGSe and ITO is turned off for the calculation in order to obtain the far field scattering, Q_b and Q_f are the normalized backward and forward cross sections, respectively.

diffraction from the NP array excites waveguide modes (Mokkapati and Catchpole, 2012), which confine light within CIGSe absorbers and enhance Abs_{CIGSe} . The corresponding near field distributions are shown in Fig. 5(a). The near field profiles are characteristic of regions of constructive and destructive interferences. At a free-space wavelength of 820 nm, where the bare CIGSe absorption is still relatively strong, the peak field strength in the CIGSe absorber is only comparable to the incident light E_0 . However, at the other three peaks, significant field confinements (higher than E_0) are present on the top part of hemispheres within the absorber and are located away from the ITO substrate. This visually explains the reduction of Abs_{ITO} and T after patterning as shown in Fig. 4(b). Since the waveguide modes lie in the poor-absorbing range, the influence on the Abs_{CIGSe} enhancement is substantial.

Besides the waveguide modes, the NPs are able to improve the CIGSe/ITO interface reflectivity, which arises from the backward scattering of NPs and contributes to Abs_{CIGSe} enhancement as well. To investigate scattering behavior of a single hemisphere at CIGSe/ITO interface, all outer boundaries are set to PML. The forward and backward scattering behavior is calculated and normalized to the geometric cross section of a single hemisphere, and expressed in forward (Q_f) and backward (Q_b) scattering cross sections. The backward scattering efficiency ($= Q_b/(Q_b + Q_f)$) is compared to the bare CIGSe/ITO interface reflectivity in Fig. 5(b). The extinction coefficient is neglected in both CIGSe and ITO for the calculation. We observe that the bare CIGSe/ITO reflectivity (dashed line) is quite low with a value under 20% in most of the spectral range, whereas a single NP exhibits a backward scattering efficiency (solid line) around 10% higher than the bare CIGSe/ITO reflectivity for all wavelengths. This indicates that the NPs are able to

improve the CIGSe/ITO reflectivity and directly explains the broadband Abs_{CIGSe} enhancement in the wavelength range from 550 to 800 nm, where waveguide modes are absent. It should be stressed that dielectric NPs with sizes comparable to or above the incident wavelength λ scatter light preferentially towards the incident wave propagation direction (forward scattering) (Heifetz et al., 2009; Chen et al., 2004; Itagi and Challener, 2005). In the case of front illumination, this indicates that light will be preferentially scattered into ITO. Nevertheless, compared to the bare interface, the presence of NPs enhances the backward scattering efficiency (towards CIGSe), which ($Q_b/(Q_b + Q_f)$) however still remains less than 50% in most of the spectral range. This is unlike plasmonic metallic NPs, which are expected to dominantly scatter light into the high index medium of CIGSe (a greater refractive index of CIGSe than ITO). This is due to the larger number of multipole resonances present in dielectric NPs compared to metallic ones (Schmid et al., 2014). It also implies, in the case of Mo back contact in Fig. 4(a), Abs_{back} couldn't be eliminated to a marginal level and Abs_{CIGSe} is thus suppressed. To clarify that the Abs_{CIGSe} enhancement in the wavelength range of 550–800 nm is not arising from the modulation of Fabry–Pérot interferences due to the NP incurred d_{CIGSe} change, the simulation is redone assuming the NPs to be CIGSe but neglecting the extinction coefficient. Since NPs have the same refractive index as CIGSe absorbers, little light trapping effect from NPs is expected (Supporting Fig. S4).

In order to evaluate the influence of the geometry (r, l) of hemisphere arrays on the Abs_{CIGSe} enhancement, we map in Fig. 6 integrated J_{sc} as a function of (r, l) for $d_{CIGSe} = 400$ nm. r ranges from 200 to 300 nm and l from 25 to 100 nm in steps of 25 nm. Notably, an apparent Abs_{CIGSe} enhancement is demonstrated in the whole investigated

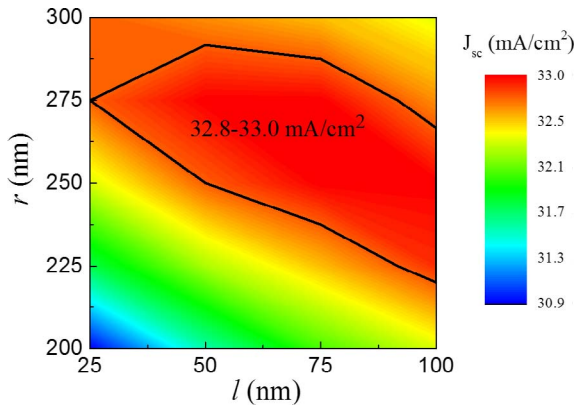


Fig. 6. J_{sc} contour as a function of (r, l) for CIGSe solar cells on ITO at $d_{CIGSe} = 400$ nm under front illumination.

geometry range (29.2 mA/cm² for the flat reference), with a maximum J_{sc} of 33.0 mA/cm² achieved at $(r, l) = (275, 75)$ nm. The broad geometry coverage can be correlated to the joint benefits of the broadband improvement of interface reflectivity and especially the multiplicity of waveguide modes covering the poor-absorbing wavelength range. From the fabrication perspective, the broad geometrical parameters of NPs represent a strong advantage inasmuch as the fabrication constraints are relaxed.

We arbitrarily define the optimum J_{sc} values in the range from 32.8 to 33.0 mA/cm². Another feature in Fig. 6 is that the optimum geometry region follows an overall trend of an increasing l coupled with a decreasing r . Since the pitch is the summation of l and twice r , increasing one while decreasing the other will tend to leave the pitch constant, and thus the wavelength positions of waveguide modes in the optimum range, which confirms the above-mentioned statement of substantial influence of waveguide modes on the J_{sc} enhancement. This also suggests that the optimum (r, l) range for $d_{CIGSe} = 400$ nm is also applicable for other absorber thicknesses.

It should be specially noted here that CIGSe solar cells have an excellent hole diffusion length up to more than 10 μ m (Casper et al., 2016). The sizes of the hemispheres investigated are within the diffusion length of holes. Further, it was experimentally demonstrated that dielectric nanostructures at the back interface are potentially able to passivate the back interface and reduce the back recombination rather than degrade CIGSe qualities or increase the interface recombination for ultrathin CIGSe cells (Vermang et al., 2013; Lare et al., 2015; Yin et al., 2016b, 2016c; Salome et al., 2017; Casper et al., 2016). Regarding the fabrication complexities of NPs, several lithography technologies such as nanosphere lithography (Kandulski, 2007), substrate conformal imprint lithography (Verschuuren, 2010), are able to fabricate large scale NP arrays with a high tolerance to the substrate and other environment factors. Envisaging the vast consumption of absorber materials in mass production, it is promising to employ these technologies into the fabrication of ultrathin CIGSe solar cells without a cost increase.

To estimate the dependence of Abs_{CIGSe} limit on d_{CIGSe} for patterned cells, Fig. 7 depicts Abs_{CIGSe} and T in terms of J as a function of d_{CIGSe} ranging from 300 to 500 nm. The hemisphere geometry parameter (r, l) is fixed at (275, 75) nm, which is within the optimum geometry range. Abs_{CIGSe} for flat cells is also added as a reference for each d_{CIGSe} . The patterned cells exhibit a significant Abs_{CIGSe} gain at each d_{CIGSe} , with a total J_{sc} value from 30.4 to 34.5 mA/cm² and yielding a T decrease from 5.7 to 3.2 mA/cm² as d_{CIGSe} increases. For $d_{CIGSe} > 400$ nm, around 3.0 mA/cm² gain in Abs_{CIGSe} is obtained. Whereas, for $d_{CIGSe} < 400$ nm, the Abs_{CIGSe} increase is beyond 4 mA/cm² since more light interacts with NPs. The summation of Abs_{CIGSe} for patterned cells and the corresponding T linearly decreases with d_{CIGSe} . This means that even with total collection of the transmitted light via a lossless back reflector, the

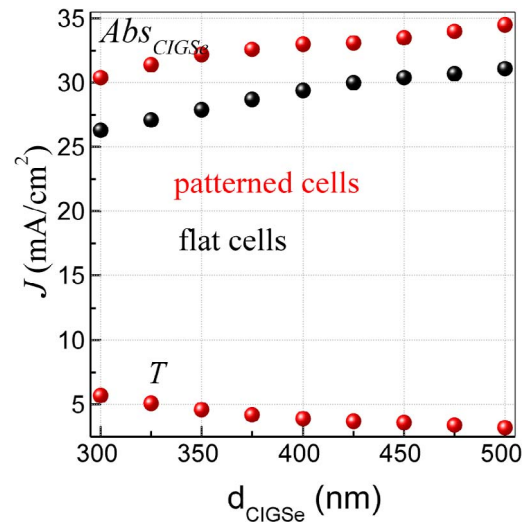


Fig. 7. Abs_{CIGSe} and T in terms of J as a function of d_{CIGSe} at $(r, l) = (275, 75)$ nm under front illumination.

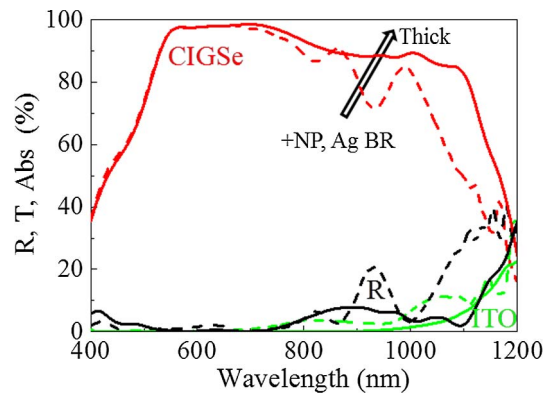


Fig. 8. Optical responses (R/Abs) between patterned ultrathin CIGSe solar cells with the addition of a Ag back reflector at the upper limit $d_{CIGSe} = 500$ nm and $(r, l) = (275, 75)$ nm (dashed lines) and thick flat CIGSe solar cells at $d_{CIGSe} = 2000$ nm (solid lines).

maximum Abs_{CIGSe} would be achieved at the upper limit d_{CIGSe} of 500 nm for ultrathin CIGSe solar cells.

Fig. 8 depicts R/Abs (dashed lines) of patterned cells at $d_{CIGSe} = 500$ nm after adding a Ag back reflector. As the remaining T is reflected back into the cells, the J_{sc} further increases up to 36.4 mA/cm², which is 1.9 mA/cm² higher than in the case without a Ag back reflector. To identify the Abs_{CIGSe} gap between ultrathin cells and thick cells, R/Abs of flat thick solar cells is also attached for comparison in Fig. 8 (solid lines, $d_{CIGSe} = 2000$ nm) and Abs_{CIGSe} corresponds to a J_{sc} of 38.7 mA/cm². This means that the ultrathin patterned cells achieve 94% J_{sc} of the thick flat counterparts. The Abs_{CIGSe} gap (2.3 mA/cm²) lies in the infrared range, R and the parasitic Abs_{ITO} in the patterned cells are mainly responsible due to the still imperfect absorption.

5.2. Back illumination

CIGSe solar cells on a TCO back contact allow back illumination of light from the glass substrate, which is termed as a backwall structure (Larsen et al., 2014). In contrast to front illumination, the anti-reflection MgF_2 is not needed and a back reflector can be coated on top of the device to reflect transmitted light back into devices. Compared to the superstrate configuration with direct absorber deposition on buffer layers, the backwall structure offers the advantage of avoiding the deterioration of p-n junctions incurred by the high substrate temperature growth of CIGSe (Young et al., 2002). Simultaneously, ultrathin CIGSe

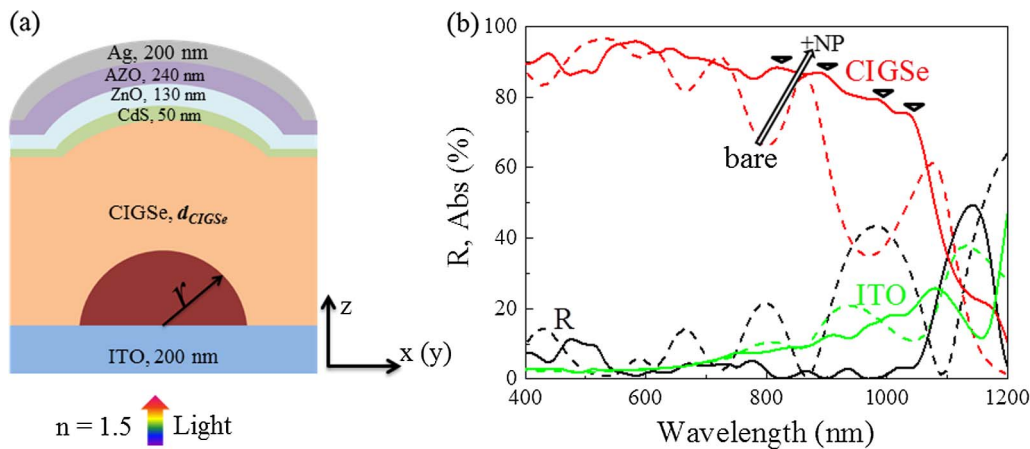


Fig. 9. (a) Sketch of a patterned CIGSe solar cells on ITO under back illumination, (b) corresponding optical responses (R / Abs) of flat and patterned CIGSe solar cells at $d_{CIGSe} = 400$ nm and $(r, l) = (275, 25)$ nm.

cells are free of the carrier collection issue thick cells under back illumination are encountering, arising from the primary absorption of light close to the back of the absorber rather than at the p-n junction (Larsen et al., 2014).

Fig. 9(a) illustrates the sketch of backwall cells with a Ag back reflector at the front side, the corresponding simulation mesh structure is shown in supporting Fig. S5. To avoid the enormous computation efforts for incorporating a thick glass substrate, the approximation is assumed that the incident medium is the glass substrate. Fig. 9(b) compares R and Abs between flat (dashed lines) and patterned cells (solid lines) at $d_{CIGSe} = 400$ nm and $(r, l) = (275, 25)$ nm under back illumination. Abs_{CIGSe} in flat cells (red dashed line) exhibits pronounced Fabry-Pérot interferences and overall taper off towards longer wavelengths due to incomplete absorption. R (black dashed line) is becoming the main optical loss channel. The patterning flattens the Fabry-Pérot interferences of Abs_{CIGSe} (red dashed to red solid lines) and overall gives a J_{sc} gain as high as 4.1 mA/cm^2 (from 32.7 to 36.8 mA/cm^2). As compared in Fig. 9(b), the significant J_{sc} gain is originating from the constraint of R : after patterning, R (black solid line) is overall reduced to a marginal level (below 5%) with much smoother interferences.

As stated in the case of front illumination, dielectric NPs with sizes comparable or larger than the incident wavelengths feature dominant forward scattering ability. Back illumination can utilize this feature and the preferential direction is towards the CIGSe layer rather than towards ITO under front illumination. For a better understanding of how the dominant forward scattering contributes to the Abs_{CIGSe} enhancement, we simulate cells with thick absorbers, where incident light can be completely absorbed without reaching the back contact. Fig. 10(a) compares R/Abs curves between flat and patterned cells at $d_{CIGSe} = 2000$ nm. Surprisingly, a further broadband Abs_{CIGSe} enhancement (3.2 mA/cm^2) is observed. Similar to the ultrathin cells in Fig. 9(b), R is greatly reduced to almost zero in the wavelength range $800\text{--}1000$ nm and is mainly responsible for Abs_{CIGSe} enhancement. Since the absorbers are thick enough to allow complete absorption of incident light, R reduction and incurred Abs_{CIGSe} enhancement are then expected to be arising from improved incoupling efficiency (towards CIGSe absorbers) at ITO/CIGSe interface due to the preferential forward scattering of NPs. The preferential forward scattering behavior of dielectric NPs is typically characterized by a concentrated electric fields close to their shadow-side surface, forming a jet-like electric field distribution (Chen et al., 2004; Itagi and Challener, 2005). Fig. 10(b) shows the near field contour at $\lambda = 800$ nm and we can observe that the electrical field is most concentrated within the NP and gradually reduces in intensity as it propagates towards the CIGSe absorber, forming a jet-like field distribution (also seen at other wavelengths). However, the jet-like field distribution shown in Fig. 10(b) is not observed for

ultrathin CIGSe cells in Fig. 9(b), this is because the light concentration phenomenon in ultrathin cells is combined with the waveguide modes (see below) and back reflecting of Ag back reflector.

Besides the dominant forward coupling efficiency at CIGSe/ITO, waveguide modes are also excited in ultrathin cells. The four Abs_{CIGSe} peaks as highlighted in Fig. 9(b) are also linearly red-shifting as l is increasing. Therefore, the Abs_{CIGSe} enhancement with the addition of NPs under back illumination is from the combined benefits of preferential forward scattering and multiple waveguide modes.

For discovering the optimum geometry range of NPs for Abs_{CIGSe} enhancement under back illumination, J_{sc} as a function of (r, l) for $d_{CIGSe} = 400$ nm is mapped in Fig. 11. Here, all J_{sc} values calculated from the model in Fig. 9(a) are calibrated with a 5% deduction for considering the loss of the single reflection at air/glass substrate. In the whole geometry range, a Abs_{CIGSe} enhancement is demonstrated at least 2.1 mA/cm^2 and the maximum enhancement up to 3.9 mA/cm^2 is achievable at $(r, l) = (275, 25)$ nm compared to flat references. Remarkably, in the most geometry ranges investigated, the patterned ultrathin cells realize a higher Abs_{CIGSe} than thick flat cells at $d_{CIGSe} = 2000$ nm (33.7 mA/cm^2). To identify the lower limit of d_{CIGSe} for obtaining comparable Abs_{CIGSe} to thick flat cells, Fig. 12 depicts Abs_{CIGSe} in terms of J_{sc} as a function of d_{CIGSe} ranging from 300 to 500 nm under a fixed NP geometry parameter $(r, l) = (275, 25)$ nm. Patterning gives rise to a great Abs_{CIGSe} enhancement at each individual d_{CIGSe} . As d_{CIGSe} increases, the patterned Abs_{CIGSe} gradually increases and tends to saturate at 35.0 mA/cm^2 from $d_{CIGSe} = 425$ nm. Remarkably, the patterned J_{sc} at $d_{CIGSe} = 300$ nm (33.6 mA/cm^2) is almost equal to that of the thick flat cells at $d_{CIGSe} = 2000$ nm, which indicates that the patterned cells under back illumination with a d_{CIGSe} of only 300 nm are able to realize comparable absorption to thick flat cells (33.7 mA/cm^2).

Under front illumination, patterned ultrathin CIGSe solar cells reach 94% Abs_{CIGSe} of their thick counterparts at the upper limit d_{CIGSe} of 500 nm. In contrast, under back illumination, ultrathin cells achieve an Abs_{CIGSe} equal to their thick counterparts at a d_{CIGSe} of only 300 nm. However, the absolute maximum J_{sc} under back illumination (35.2 mA/cm^2 , Fig. 12) is lower than the maximum value under front illumination (36.8 mA/cm^2 , Fig. 8). Reviewing Fig. 9(b), Abs_{ITO} is the main factor suppressing a greater maximum Abs_{CIGSe} under back illumination. The parasitic Abs_{ITO} is mainly stemming from the free carriers necessary for a good conductivity as a back contact. This indicates that the advantage of back illumination for achieving the maximum Abs_{CIGSe} at a lower d_{CIGSe} is dissipated due to a lower maximum Abs_{CIGSe} compared to front illumination. High-mobility TCOs (e.g. $\text{H:In}_2\text{O}_3$) feature higher carrier mobility and less carrier concentration, which guarantees good conductivity without increasing absorption ability (Onwudinanti et al.,

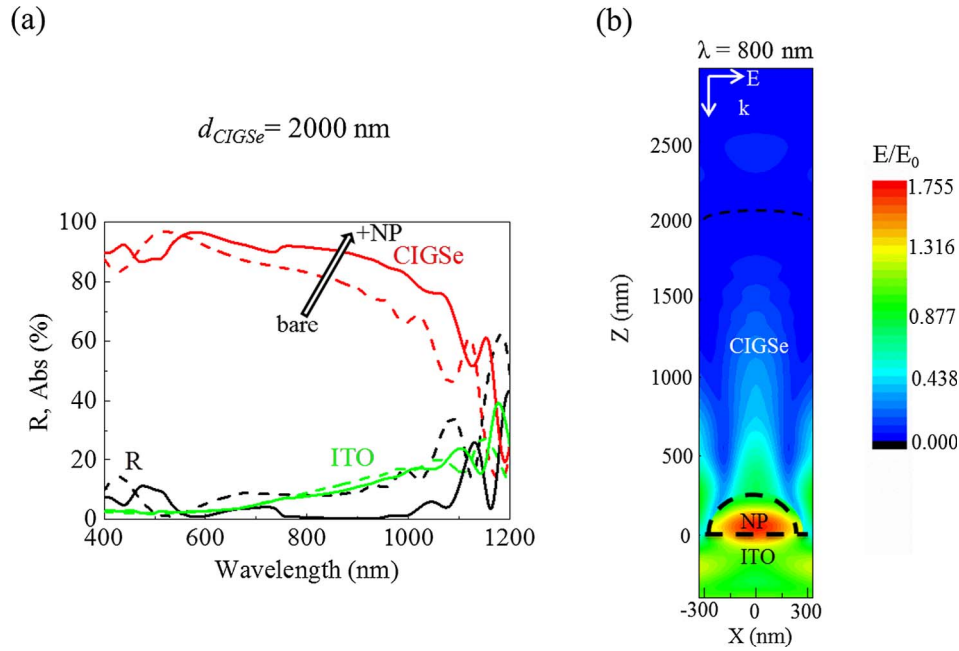


Fig. 10. (a) Optical responses (R/Abs) of flat and patterned CIGSe solar cells at $d_{\text{CIGSe}} = 2000$ nm and $(r, l) = (275, 25)$ nm under back illumination, (b) near field distribution at $\lambda = 800$ nm.

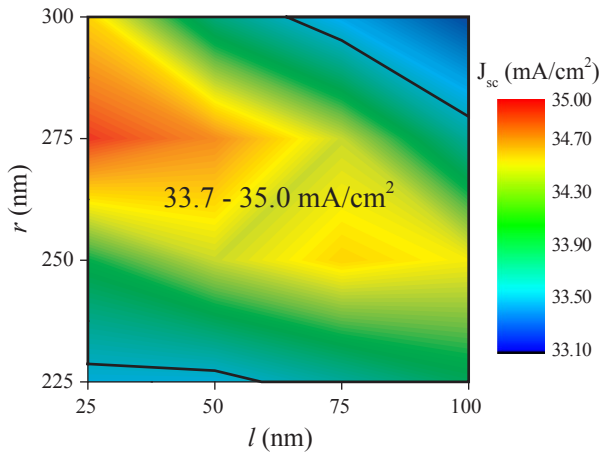


Fig. 11. J_{sc} contour as a function of (r, l) at $d_{\text{CIGSe}} = 400$ nm under back illumination.

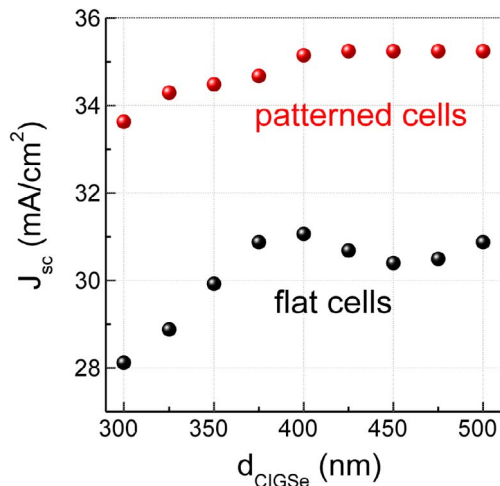


Fig. 12. Abs_{CIGSe} in terms of J_{sc} as a function of d_{CIGSe} for flat and patterned CIGSe solar cells under back illumination.

2016; Yin et al., 2015). Applying them to replace ITO may be a promising option to further reduce the parasitic absorption in the back contact and utilize the advantage of back illumination.

Additionally, in module configuration, (1) MgF_2 will be replaced by encapsulation EVA and glass substrate with an index around 1.5, (2) a much thicker AZO (around 1000 nm) will be applied for lateral transport of electrons under front illuminated cells. This will respectively give rise to an increased R and parasitic Abs_{top} , leading to a significantly reduced J_{sc} compared to the lab-scale cells shown in this work. Whereas under back illumination, due to the very good conductivity of the Ag back reflector, AZO is even not necessary. Even with a dielectric back reflector (e.g. TiO_2 paste), absorption in AZO/ ZnO / CdS with a thicker AZO layer will be still negligible since most light is first absorbed by the patterned CIGSe absorbers. Therefore, the maximum J_{sc} is expected to be much greater under back illumination than under front illumination in module configuration. To simply quantify this, Fig. 13 shows the simulated Abs of patterned ultrathin solar cells in module configuration with a 1000 nm thick AZO under two different illumination directions. For calculation simplicity, light is assumed incident from glass substrate in both illumination cases. As expected, the parasitic absorption in AZO under front illumination is quite severe, which causes a much reduced Abs_{CIGSe} with respect to the corresponding lab-scale cells. In contrast,

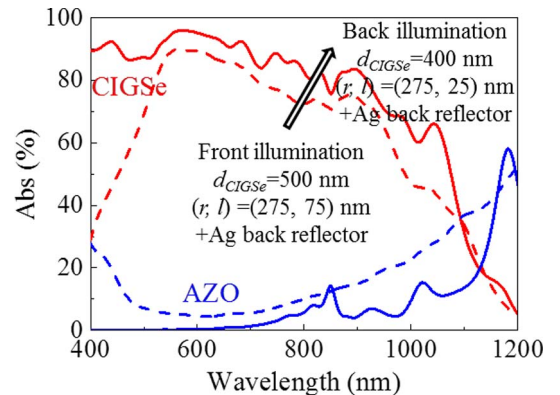


Fig. 13. Abs curves of patterned ultrathin CIGSe solar cells with a 1000 nm thick AZO and a Ag back reflector under both front and back illuminations.

under back illumination, Abs_{CIGSe} can reach 33.5 mA/cm^2 , which is 3.6 mA/cm^2 higher than the value under front illumination. This indicates that the back illuminated hemisphere patterned ultrathin CIGSe solar cells will be a promising structure for high efficiencies in module production.

6. Conclusion

In this work, we extensively examined light trapping effects of dielectric NPs in ultrathin CIGSe solar cells from three aspects: interface locations of NPs, back contacts and illumination configurations. The conclusions are as follows:

- (1) The parasitic absorption in Mo is the main optical loss for flat ultrathin CIGSe solar cells and is also the main constraining factor for achieving a significant Abs_{CIGSe} enhancement in NP-patterned ultrathin cells. The TCO/glass/back reflector back contact structure (ITO/glass/Ag in this work), exhibiting much less parasitic absorption and excellent back-reflecting ability, is recommended to replace the conventional Mo for a higher Abs_{CIGSe} .
- (2) NPs are favoured to be directly placed at the interface of CIGSe/back contact. This allows light trapping effects of NPs being utilized to the greatest extent by the CIGSe absorber rather than being attenuated by other non-active layers.
- (3) Under front illumination, the hemisphere ($n = 1.5$) patterned ultrathin CIGSe solar cells on ITO show a significant Abs_{CIGSe} enhancement, which is due to the joint effects of waveguides modes and back scattering of NPs. The broadband improvement of back scattering and especially the multiplicity of waveguide modes determine the broadness of the geometry range of hemisphere arrays for a maximum Abs_{CIGSe} . In conjunction with a Ag back reflector, the maximum J_{sc} reaches 36.4 mA/cm^2 at the upper $d_{CIGSe} = 500 \text{ nm}$, 94% of that of their thick flat counterparts.
- (4) The ITO substrate also allows back illumination and utilization of a back reflector in the front. The hemispherical NPs contribute to a broadband Abs_{CIGSe} gain via their preferential forward scattering and waveguide modes, which reduces R to minimum and traps light within CIGSe absorbers. Consequently, the patterned CIGSe solar cells are able to reach comparable Abs_{CIGSe} to their thick cells at a d_{CIGSe} of only 300 nm , and the maximum J_{sc} saturates at a value of 35.2 mA/cm^2 at $d_{CIGSe} = 425 \text{ nm}$.
- (5) For lab-scale cell configurations, in contrast to front illumination, the advantage of back illumination of achieving its maximum J_{sc} at a low d_{CIGSe} is shadowed because of a relatively lower maximum J_{sc} due to the strong parasitic absorption in ITO back contact. However, in module configuration with a much thicker AZO, the patterned cells are less affected by the heavy absorption in AZO under back illumination than under front illumination and thus exhibit a much higher Abs_{CIGSe} . This suggests that patterned ultrathin CIGSe solar cells on TCO substrate under back illumination will be a promising structure for high efficiencies in industrial module production.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.solener.2018.01.096>.

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