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### RAPID COMMUNICATION

# Performance optimization of flexible a-Si:H solar cells with nanotextured plasmonic substrate by tuning the thickness of oxide spacer layer



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#### Abstract

Plasmonic thin film solar cells deposited on periodically textured photonic crystal substrates have been extensively studied since the substantially enhanced light absorption. The reduction of parasitic absorption losses in the metal and spacer layers becomes one of the key issues to achieve high efficiency solar cells. Herein, plasmonic amorphous silicon (a-Si:H) flexible thin film solar cells with different thickness of oxide spacer layers are systematically investigated. An increase of the spacer layer thickness leads to an evolution in surface morphology of AZO and final devices. More intriguingly, the increase of spacer layer thickness reduces the absorption in Ag layer while induces more absorption in spacer layer. The highest light absorption in silicon layer is observed as applying 100 nm spacer layer, which is further verified by electrical measurements. Our observations

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demonstrate a versatile and convenient route towards rational design of light harvesting nanostructure for high performance plasmonic solar cells based on a broad range of materials. © 2014 Elsevier Ltd. All rights reserved.

#### Introduction

Hydrogenated amorphous silicon (a-Si:H) thin-film solar cells are promising candidates for both large scale and portable photovoltaics, owing to their merits of low cost, material abundance, non-toxicity and compatibility with various types of substrates. The energy conversion efficiency of a-Si:H thin film solar cells can achieve up to 27% as its optical bandgap is around 1.7 eV according to the theoretical Shockley-Queisser detailed balance limit [1]. So far, the efficiency of state-of-the-art a-Si:H thin film solar cells is limited by 10.09% [2]. The large discrepancy between the theoretical indication and real device performance is attributed to the high density of tail and defect states in the a-Si:H thin film, which is responsible for the recombination losses and in turn low performance of the solar cells [3]. Reducing i-layer thickness will suppress the lightinduced carriers' recombination loss, while the light absorption is also reduced. In order to comprise the carriers collection and optical absorption, a variety of random nanotextures [4] and periodic nanostructures including nanogratings [5,6], nanodomes [7-10], nanodents [11,12], nanovoids [13,14], nanoholes [15-17] nanopillars [18,19] nanospikes [20], nanorods [21] and inverted nanocones [22], have been developed to improve the light absorption capability while keeping the a-Si:H layer thickness as thin as possible. Metallic thin films are often integrated in these periodic structures, which can couple incident light into both the photonic and surface plasmon resonance modes in the absorption layer [11,23]. In the case of plasmonic a-Si:H thin film solar cells, the optical parasitic absorption loss in the metal layer is a key issue to be overcome. Recently, we have demonstrated a-Si:H thin film solar cells on nanodent plasmonic substrate [11,12]. The light absorption loss in the Ag layer can be easily observed for wavelength beyond 600 nm and became more significant in the device with

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thinner absorbing layer [11]. An Al doped ZnO (AZO) spacer is usually placed between Ag and a-Si:H layer. Besides the limit of materials diffusion [24], the AZO spacer, with a lower refractive index, also acts the function to inhibit the losses in reflected light due to the excitation of surface plasmon resonance. The role of the spacer is to shift the plasmon resonance frequencies toward higher energies. that is already absorbed in the front part of the cell [25,26]. The recent report by Lal et al. indicated that the light confinement in silicon layer can be enhanced by modifying the thickness of AZO spacer layer in nanovoid plasmonic substrate [14]. However, the optical and electrical performance of plasmonic a-Si:H solar cells as a function of the thickness of AZO layer has not been well studied. Herein, a-Si:H solar cells with different thickness of AZO spacer layer on nanodent plasmonic substrate are systematically investigated. Finite difference time domain (FDTD) simulation results demonstrate that the light absorption loss on the Ag surface is reduced as the AZO thickness increases, while the thicker AZO absorbs more light. In addition, solar cell with 100 nm AZO can confine the incident light in silicon layers much more effectively, leading to its highest short circuit current  $(J_{SC})$ . These results not only provide an effective method to understand the coupling effect of light propagation and electrical transport, but also demonstrated a possible strategy to design and optimize the nanostructured thin film solar cells based on other photovoltaic materials.

#### Experimental

Fig. 1a schematically shows the layered structure of an a-Si:H thin-film cell deposited on nanopatterned Al substrate. Detailed fabrication processes of the patterned Al substrate can be found in our previous work [11,12]. A 100 nm thick Ag layer was deposited on the patterned Al substrate by direct current







magnetron sputtering at room temperature in argon plasma atmosphere. Then, the AZO spacer layers, whose thickness varied from 30 to 200 nm, were prepared on the Ag coated substrate by radio frequency magnetron sputtering of a 2 wt% Al<sub>2</sub>O<sub>3</sub> doped ZnO ceramic target (purity 99.99%) under argon plasma at substrate temperature of 150 °C. Subsequently, a stack of n-i-p silicon layers were successively fabricated in plasma enhanced chemical vapor deposition (PECVD) multichamber system. All these chambers have identical capacitively coupled parallel-plate electrode configurations. Doping was achieved by gas mixture of hydrogen-diluted phosphine  $(PH_3)$ for deposition of n layer and hydrogen-diluted diborane  $(B_2H_6)$ for p layer at an excitation frequency of 13.56 MHz. The a-Si:H absorber layers were prepared from a mixture of  $SiH_4$  and  $H_2$ gases at a excitation frequency of 40 MHz. The silane concentration ratio ( $[SiH_4]/([SiH_4]+[H_2])$ ) of i layer was 9.1%. An 80 nm ITO top contact, which also serves as an antireflection coating, was deposited by radio frequency sputtering. Finally, Ag top electrode was evaporated over ITO using a mask at room temperature.

Both the surface and cross-sectional images of the samples were characterized by scanning electron microscope (SEM, Philips, XL30). An X-ray diffraction apparatus (XRD) (Bruker, D8 Advance) with Cu  $K\alpha$  radiation was applied to identify crystalline structure of the AZO and Ag films. Reflectance of the solar cells was investigated using an ultraviolet-visible spectrophotometer (SHIMADZU, UV2700). The initial performance of cells (active area of 0.12 cm<sup>2</sup>) was evaluated by measuring current-voltage (*J-V*) charac-

teristics with the solar simulator (Newport, 94063A-1000) under the standard test conditions (25 °C, Air Mass 1.5G, 100 mW/cm<sup>2</sup>) and external quantum efficiency (EQE) spectra (PV Measurement Inc. QEX10).

The simulations of the optical performance were accomplished by employing FDTD method (Lumerical FDTD solutions). Ag was modeled using Lorentz-Drude fitting following the method of Rakic et al. [27]. The optical constants of AZO and a-Si:H thin film were taken from the measured data, while the refractive index of ITO was set as 2.08+0.004i



Fig. 3 The X-ray diffraction patterns of the patterned Al substrates with 30 nm, 100 nm and 200 nm AZO spacer layers.



Fig. 2 Surface and cross-section views of (a) 100 nm Ag coated Al patterned substrate, and the 100 nm Ag-coated substrates deposited with (b) 30 nm AZO, (c) 100 nm AZO and (d) 200 nm AZO. The scale bars in the insets represent 500 nm.

according to the results from Ferry et al. [28]. Further simulation details such as the select of modeling unit cell can be found in our previous work [11,12].

#### **Results and discussion**

Fig. 2 represents the SEM images of the patterned Al substrates coated with 100 nm Ag, 100 nm Ag/30 nm AZO, 100 nm Ag/100 nm AZO and 100 nm Ag/200 nm AZO,

respectively. A dent-like Ag film is formed due to the underlying patterned Al structure as shown in Fig. 2a. Fig. 2b depicts a 30 nm AZO film deposited on the Ag films, in which the surrounding edges of the dents become thick while the dent-like structure still remain. As the thickness of the AZO films further increases to 100 nm, some nanotextures along the nanodents boundaries emerge out of the film surface. And the sizes of these nano-textures increase as the thickness of AZO films increased to 200 nm (see Fig. 2c and d).



Fig. 4 SEM images of the surface morphology of a-Si solar cells with (a) 30 nm, (b) 100 nm and (c) 200 nm AZO spacer layers. The insets show the cross-section images of corresponding cells. The scale bars in insert images represent  $1 \mu m$ .



**Fig. 5** (a) The simulated and measured absorption of the solar cells with different thickness of the AZO spacer layer. (b) Measured EQE spectra of solar cells with different thickness of the AZO spacer layers.

Fig. 3 shows the XRD patterns of AZO as a function of film thickness. The peaks at  $34.2^{\circ}$ ,  $36.1^{\circ}$  and  $38.0^{\circ}$  correspond to the (0 0 2), (1 0 1) crystal planes of wurtzite ZnO and (1 1 1) crystal plane of Ag, respectively. It can be observed that an increase of the AZO film thickness leads to sharper peak profiles of ZnO (0 0 2) and increased intensity ratio of the (0 0 2) to (1 0 1) peaks. This suggests that the *c*-axial preferred orientation of the film is improved by increasing the AZO film thickness [29]. The appearance of the nanotextures on the AZO films shown in Fig. 2c and d may result from the evolution of *c*-axial preferred orientation.

Fig. 4 shows the surface and cross-sectional SEM images of the solar cells with 30, 100, and 200 nm AZO spacer layer, respectively. For convenience, these nanodent solar cells are denoted as ND30, ND100 and ND200, correspondingly. A morphology evolution from nanodent of the substrate to nanodome of the final device can be observed in comparison with Fig. 2. From the top view of ND30 solar cell, smooth grains with distinct boundary can be seen. Interestingly, an evolution of small grains in the nanodomes can be observed as the AZO layers increased up to 100 and 200 nm (Fig. 4b and c). The appearance of small grains may originate from the nano-textures of thick AZO layers (see Fig. 2c and d). The insets of Fig. 4 indicate that an increase of AZO layer thickness is responsible for a flatter surface of the devices, which have also been found in microcrystalline silicon thin film solar cells [30,31].

Fig. 5a shows the simulated and measured light absorption spectra of ND30, ND100 and ND200. The absorption is obtained by  $1 - R_{total}$ , where  $R_{total}$  is the total reflectence of

the solar cells. The simulated absorption curves of the three devices are overlapped in the wavelength range of 300 to 600 nm. In the same wavelength range, for the measured curves, there are two obvious features. One is the redshift of the valley of the absorption curve. This may attribute to the slight fluctuations of the real pattern size and film thickness from that of the simulated one. Another feature is that the absorption of the ND30 is higher than that of other two solar cells in the wavelength range of 350-600 nm. This can be attributed to the roughest top surface of the ND30 (see inset images of Fig. 4), in which light can be coupled into solar cells most effectively. In the long wavelength range of 600-800 nm, the peaks in the simulated curves are ascribed to the strong resonances occurring in the perfectly regular structures [9,28,32]. While for the measured curves, such peaks are absent due to the imperfect pattern structure of the substrates. Meanwhile, both simulation and experiment results show a decrease trend with the rise of AZO thickness.

Fig. 5b represents the EQE plots of ND30, ND100 and ND200, respectively. For wavelengths shorter than 500 nm, the lowest EQE of ND100 can be predominantly ascribed to its low light absorption, in considering its surface is flatter than that of ND30. While in the same wavelength range, ND200 exhibits slightly higher EQE than that of ND30, although its optical absorption is obviously lower than that of ND30. This could be due to its high quality *i/p* interface resulting from its flattest surface of ND200, which is beneficial for the carrier collection at the short wavelengths [33]. For wavelengths larger than 500 nm, ND200 has the

![](_page_4_Figure_7.jpeg)

Fig. 6 Simulated absorption spectra of each individual functional layers of the cells with (a) 30 nm, (b) 100 nm and (c) 200 nm AZO spacer layers.

lowest EQE, which can be explained by its lowest absorption in long wavelength range (see Fig. 5a). Although ND30 device possesses the highest absorption in this wavelength region, it's EQE value is lower than that of ND100. This mirrors that a substantial number of absorbed photons in the wavelength range of 500-800 nm do not contribute to the photocurrent of the device. It is know that only the part absorbed by silicon layer can be possibly converted to current. However, it is difficult to distinguish light absorption of different layers in the solar cells by experiment measurement.

With the assistance of FDTD simulation, light absorption spectra of each individual functional layers are determined as a function of AZO layer thickness (Fig. 6). Briefly, the absorption of per unit volume can be calculated from the divergence of the Poynting vector [34]:

$$P_{\text{abs}}(x, y, z) = \frac{1}{2} \text{Im}(\varepsilon_{\text{r}}(x, y, z))\varepsilon_0 |\mathbf{E}(x, y, z)|^2 \omega$$

where  $Im(\varepsilon_r(x, y, z))\varepsilon_0$  refers to the imaginary part of the permittivity, E(x, y, z) is the electric field intensity vector and  $\omega$  is the angular frequency of incident light. The absorption is calculated by integrating  $P_{abs}(x, y, z)$  along the spatial domain. In order to separate each individual functional layers,  $P_{abs}(x, y, z)$  is multiplied by a filter before integration. These filters are created according to the distribution of refractive index. At the point where the index equals to that of a specific layer, the filter is set to be 1, otherwise it is set to be 0. In this way, the filters for each layer outline the

spatial region of individual layers. As a result, the integrated values are only the absorption of these specific layers, rather than the overall absorption of the device.

The simulated results illustrate that light absorption of the ITO layers cover the whole wavelength range of 300-800 nm. As the incident light goes across ITO layers, silicon layers absorb most of the light in the wavelength range of 300-550 nm, due to the high absorption coefficient of a-Si:H for the short-wavelength light. For the wavelengths beyond 600 nm, the serious light absorption of the AZO and silver can be observed. As the AZO thickness increases from 30 to 200 nm, the absorption of the AZO film increases due to the increased optical path length, that could be originated from the thicker film and coupled surface plasmon resonance modes. In contrast, the absorption of Ag films decreases along with the increase of the AZO thickness.

Moreover, to investigate how the long wavelength light is coupled into these devices, the cross-sectional electric field intensity  $(|E|^2)$  in the solar cell are simulated as shown in Fig. 7, in which the wavelengths of the incident light are chosen as 600 and 750 nm, respectively. The color index represents the magnitude of  $|E|^2$  normalized with that of the light propagating in free space. Black guidelines illustrate the interfaces of the each layer in the solar cells. High  $|E|^2$  can be always found in the silicon layers near the tips and centers of the textures. Intense  $|E|^2$  can also be found in the AZO layers and on the surface of the silver layers when the ND30 is irradiated by 600 and 750 nm incident light (Fig. 7a and d). As the AZO thicknesses increase to 100 and 200 nm,

![](_page_5_Figure_8.jpeg)

**Fig. 7** Cross-sectional electric field intensity ( $|E|^2$ ) distributions in ((a1) and (a2)) ND0, ((b1) and (b2)) ND30, ((c1) and (c2)) ND100, ((d1) and (d2)) ND200 at a incident light wavelength of ((a1)-(d1)) 600 nm and ((a2)-(d2)) 750 nm.

the strong  $|E|^2$  on Ag surface is gradually transferred into AZO. This suggests that the energy loss on Ag surface will be suppressed as the thickness of the AZO becomes larger. However, the parasitic light absorption in the AZO becomes significant as the AZO thickness increased up to 200 nm, which can hardly contribute to the photocurrent. It is worth pointing out that the presence of an AZO layer between Ag and a-Si:H can efficiently suppress the energy loss occurred on Ag surface. As a demonstration, the spatial distributions of electric field intensity in the solar cell without AZO spacer (denoted as NDO) are also calculated as illustrated in Fig. 7a1 and a2. As expected, the optical absorption in the Ag back contact is distinctly higher compared to that in the devices with AZO layers. The light penetrates the Ag layers by around 30 nm under both 600 and 750 nm illuminations. In this regard, the reflection from the back contact drops and the absorption in the active layer is reduced.

As discussed above, except for silicon layer, other functional layers can hardly convert the absorbed light into electricity. Herein, the influence of the parasitic absorption on the current generation is further investigated. Assuming that the absorbed light in top ITO layers, bottom AZO layers, and bottom silver layers can also generate current, and the absorption of one incident photon induces one electron-hole pairs, the current (*I*) generated in the whole solar cells and individual functional layers can be calculated by *I*=2  $q \times \int Abs(\lambda) S(\lambda) d\lambda$ , where  $Abs(\lambda)$  is the absorption of the device or individual layers based on the simulation results, *S* ( $\lambda$ ) is the photon flux calculated based on the standard AM 1.5G solar spectrum data (ASTM G-173-03) and *q* is the

elemental charge. As shown in Fig. 8a, the calculated current density in ND100, generated by the incident light below 600 nm, is the lowest one among the three devices, while in the wavelength beyond 600 nm, ND100 represents higher photocurrent than ND30 and ND200. This result is consistent with OE data. Similar trend is also found in silicon lavers calculation (Fig. 8b), which delivers the highest photocurrent value of 15.48 mA/cm<sup>2</sup> over the whole wavelength range on ND100 device. The current density contributed from Ag layer reduces from 7.29 to 3.51 mA/cm<sup>2</sup> when the AZO thickness increases from 30 to 200 nm, as shown in Fig. 8c. And the current density coming from the parasitic absorption of AZO layer increases from 2.25 to 4.78 mA/cm<sup>2</sup> with the increase of the AZO thickness, as depicted in Fig. 8d. It is worth noting that optimal AZO thickness and corresponding optical properties in each individual layer are also strongly dependent on the thickness of a-Si layer [11]. More in-depth understanding on the coupling effect are worthy of extensive investigation in the future work.

Fig. 9 plots the experimental results of open circuit voltage ( $V_{OC}$ ), fill factor (*FF*), short circuit current ( $J_{SC}$ ) and efficiency of solar cells as functions of AZO thickness. The average values of  $V_{OC}$  increase with the increase of the AZO films, which should be related to the flattening of the cells surface (Insets of Fig. 4). The flatter cell surface decreases the defects in the i/p interface that enhance the carriers collection [33]. And the thickening of the AZO films may also cut down the number of current leakage pass from the highly conductive substrate. The  $J_{SC}$  of the solar cells increases as the AZO thickness changes from 30 to 100 nm, but declines

![](_page_6_Figure_6.jpeg)

Fig. 8 The calculated light current density of (a) the whole cell, (b) silicon layers, (c) Ag layers and (d) AZO space layers.

![](_page_7_Figure_1.jpeg)

Fig. 9 The electrical performance ( $V_{OC}$ , FF,  $J_{SC}$  and efficiency) of cells plotted as a function of AZO thickness.

![](_page_7_Figure_3.jpeg)

**Fig. 10** (a) Relative efficiency variation under different incident light angles. (b) Relative efficiency variation of ND100 at different bending angles. The inset shows (b1) an optical image of measurement setup and (b2) a schematic of defining bending angle.

on ND200 devices. Quantitatively, the  $J_{SC}$  of ND100 shows an enhancement of 7.7% and 6.9% as compared with ND30 and ND200, respectively. This agrees well with the calculated results as shown in Fig. 8b, in which the highest light induced current in the silicon layer was achieved on ND100 device. *FF*s decrease along with the increase of AZO thickness (Fig. 9c), which can be attributed to the larger series resistance of the solar cells with thicker AZO layer. Overall,

optimized solar cells, with moderate  $V_{OC}$  and FF but the best efficiency of 6.36% are obtained as 100 nm AZO is employed.

It is known that the angle of solar irradiation changes over time in a day, while the above measurements are conducted under normal incident light. In this regard, angular dependent device performance of ND100 is characterized as shown in Fig. 10a. Thanks to the textured top surface, the conversion efficiency remains 60.7% as the incident angle increased from  $0^{\circ}$  to  $60^{\circ}$ . Further improvement on omnidirectional performance is expected to be achieved by antireflection coating on the device top surface [35].

Moreover, the aluminum foils based substrates possess excellent flexibility and its fabrication is highly scalable [20,36], which cannot be achieved in the rigid nanotextured substrate based on glass [9,37]. In order to evaluate the robustness of device performance under bending, the ND100 device is further measured using a customized setup as shown in the left inset of Fig. 10b. The plot in Fig. 10b represents the efficiencies of the ND100 device regardless the change of projection area of a simulated light source under bending condition. The conversion efficiencies only experienced a 8.8% drop even under  $120^{\circ}$ . These results demonstrate the outstanding flexibility of light weight aluminum foil based solar cells.

#### Conclusions

The thickness dependence of AZO spacer layers films on the performance of a-Si:H plasmonic thin film solar cells has been systematically studied. It is found that increasing the thickness of this spacer layer not only causes an evolution in surface morphology of AZO and eventual devices, but also plays an important role on the optical and electrical performance of solar cells. Simulated light absorption in the cells reveals that the absorption loss in Ag layer is reduced whereas parasitic absorption in AZO film increased along with the increase of the AZO thickness. And the highest light absorption in silicon layer is achieved in ND100 device. These results are further verified by experimental measurements where the best performance takes place on ND100 with moderate  $V_{OC}$ , FF and highest  $J_{SC}$ . By employing the plasmonic substrates, this study based on experimental results and optical simulation demonstrates a rational strategy to design and optimize the nanostructured thin film solar cells as well as other optoelectronic devices.

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![](_page_9_Picture_7.jpeg)

Huapeng Xiao is currently a joint M.S. candidate of Nanjing University of Science and Technology and Shanghai Advanced Research Institute, Chinese Academy of Sciences. His current scientific interest focuses on thin film a-Si:H solar cells.

![](_page_9_Picture_9.jpeg)

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![](_page_9_Picture_11.jpeg)

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![](_page_9_Picture_13.jpeg)

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![](_page_9_Picture_15.jpeg)

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![](_page_9_Picture_18.jpeg)

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![](_page_9_Picture_21.jpeg)

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![](_page_9_Picture_24.jpeg)

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![](_page_9_Picture_27.jpeg)

![](_page_9_Picture_28.jpeg)

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