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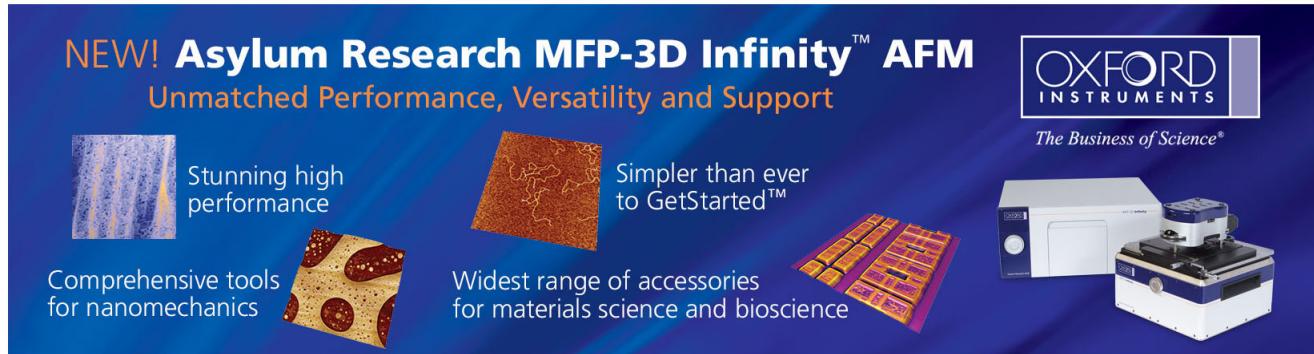
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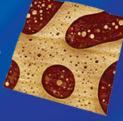
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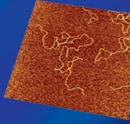
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# Multi-functional stacked light-trapping structure for stabilizing and boosting solar-electricity efficiency of hydrogenated amorphous silicon solar cells

Wen-Hsien Huang,<sup>1</sup> Jia-Min Shieh,<sup>2,3,a)</sup> Fu-Ming Pan,<sup>1,b)</sup> Chang-Hong Shen,<sup>2</sup> Jung Y. Huang,<sup>3</sup> Tsung-Ta Wu,<sup>2</sup> Ming-Hsuan Kao,<sup>3</sup> Tzu-Hsuan Hsiao,<sup>3</sup> Peichen Yu,<sup>3</sup> Hao-Chung Kuo,<sup>3</sup> and Ching-Ting Lee<sup>4</sup>

<sup>1</sup>Department of Materials Science and Engineering, National Chiao-Tung University, Hsinchu 30010, Taiwan

<sup>2</sup>National Nano Device Laboratories, No. 26, Prosperity Road 1, Hsinchu 30078, Taiwan

<sup>3</sup>Departments of Photonics and Institute of Electro-Optical Engineering, National Chiao-Tung University, Hsinchu 30010, Taiwan

<sup>4</sup>Institute of Microelectronics, Department of Electrical Engineering, National Cheng Kung University, Tainan 701, Taiwan

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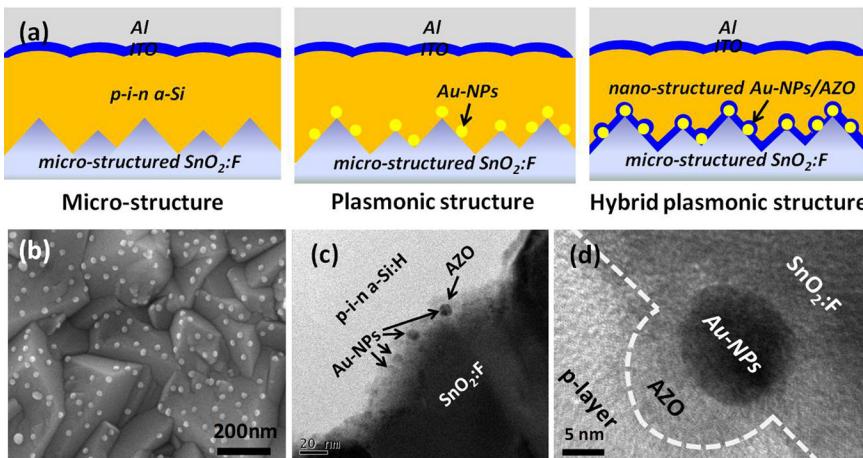
A sandwiched light-trapping electrode structure, which consists of a capping aluminum-doped ZnO (AZO) layer, dispersed plasmonic Au-nanoparticles (Au-NPs), and a micro-structured transparent conductive substrate, is employed to stabilize and boost the conversion-efficiency of hydrogenated amorphous silicon (a-Si:H) solar cells. The conformal AZO ultrathin layer (5 nm) smoothed the Au-NP-dispersed electrode surface, thereby reducing defects across the AZO/a-Si:H interface and resulting in a high resistance to photo-degradation in the ultraviolet-blue photoresponse band. With the plasmonic light-trapping structure, the cell has a high conversion-efficiency of 10.1% and the photo-degradation is as small as 7%. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4818621>]

Hydrogenated amorphous silicon (a-Si:H) thin film solar cells have attracted a wide attention because of their low-cost and low-thermal budget characteristics. To maximize the photoconversion efficiency of a solar cell, one needs to improve photo-absorption, photocurrent generation, and carrier-collection for the cell; these can be realized by optimization of the cell structure, including band-gap-engineered active layers of low defect, highly transparent conductive metal-oxide (TCO) layers with a micro-structured texture or multi-scale-structure,<sup>1,2</sup> and the introduction of plasmonic nanoparticles (NPs).<sup>3</sup> However, textured transparent electrodes barely provide multi-functionalities and efficient carrier collection for broadband light-trapping structures. Moreover, they are not suitable substrates for stacking multi-layered photovoltaic devices of high quality. A front-side ZnO electrode with a three-dimensional hexagonal micro-hole array, which was patterned by electron beam lithography, has been demonstrated to enhance the conversion efficiency of micromorph Si-based thin-film solar cells. The optical modeling shows that a-Si:H/ $\mu$ -c-Si:H micromorph cells with the micro-hole array electrode can achieve a conversion efficiency of 15%. However, experimental result yields a conversion efficiency of only 10.3%.<sup>4</sup> Boccard *et al.*, recently reported a new broadband light-trapping architecture that produced a conversion efficiency of 14.1% for micromorph solar cells.<sup>1,2</sup> The front-side electrode of the trapping structure had a homogeneous multi-scale ZnO layer prepared by the combination of nano-imprint technique and low pressure chemical vapor deposition (CVD). In addition, when metal nano-structures<sup>5,6</sup> or metal NPs,<sup>3</sup> such Ag and Au, are combined with a thick ZnO layer for the use as the back-reflector, back light-

scattering is strongly enhanced. Such nanostructures can avoid the generation of extra interface and bulk defects in a-Si:H multilayers. However, substrate-type (n-i-p) single junction solar cells fabricated with these light-trapping schemes exhibit a low conversion efficiency (<8.5%). In our previous study, highly UV-transparent dielectric particles can scatter the sunlight into the a-Si:H layer mainly via the waveguide mode,<sup>7,8</sup> enhancing light absorption in the ultraviolet-visible wavelengths regime and, thus, raising the conversion efficiency to 8.5%.<sup>9</sup> For superstrate-type (p-i-n) solar cells, the incorporation of metal nano-structures or metal NPs is rarely used in the front-side light-trapping scheme for the fabrication of stable Si thin-film solar cells with high conversion efficiencies. This is because the plasmonic enhancement in the quantum efficiency (QE) occurs only in a narrow bandwidth and the formation of interface structural defects between a-Si:H absorptive layer and TCO is inevitable.<sup>10-13</sup> In this letter, we propose a sandwiched light-trapping structure, which consists of a capping aluminum-doped ZnO (AZO) thin film, dispersed plasmonic Au nanoparticles (Au-NPs), and a micro-structured transparent conductive electrode (Asahi SnO<sub>2</sub>:F, FTO). Such the sandwiched structure provides the immunity to metal diffusion and a low-defect AZO/a-Si:H interface, where the AZO acted as a protective layer eliminating H<sup>+</sup> ion bombardment on the electrode substrate.<sup>14</sup> The low plasma power density for the a-Si:H deposition can also minimize the ion bombardment on the substrate.<sup>15</sup> Moreover, the nano-scale smooth surface of the TCO electrode by smoothing the protrusion and sharp valley of FTO/Au-NPs is beneficial to the growth of the dense a-Si:H film of low-defect density.<sup>16</sup> The Au-NPs/AZO layer enhances the absorption of the green-red solar energy via the plasmonic effect, and the micro-structured FTO functions like a short-wavelength scatter, which increases the ultraviolet-blue solar-energy utilization.<sup>1</sup> Because of these merits, the sandwich structure enhances

<sup>a)</sup>E-mail: jmshieh@ndl.narl.org.tw (or jmshieh@faculty.nctu.edu.tw). Tel.: 886-3-5726100-7617. Fax: 886-3-5722715

<sup>b)</sup>E-mail: fmpan@faculty.nctu.edu.tw. Tel.: 886-3-5712121-31322. Fax: 886-3-5724727



broadband light-harvesting and reduce the defect density across the AZO/a-Si:H interface for the a-Si:H thin film solar cell that demands a high conversion efficient and high resistance to photo-degradation. The solar cell integrated the sandwich scheme with the low-defect a-Si:H stacked layer<sup>16</sup> exhibits a short circuit current density ( $J_{sc}$ ) of  $16.6 \text{ mA/cm}^2$ , filling factor (FF) of 72.4%, conversion efficiency ( $\eta$ ) of 10.1%, and high resistance to photo-degradation (<7%).

The fabrication of the a-Si:H solar cell begins with the preparation of the multi-functional front-side electrode. First Au-NPs were self-assembled on the micro-structured Asahi-type substrate ( $\text{SnO}_2:\text{F}$ , FTO), followed by the deposition of an ultra-thin AZO capping layer. The a-Si:H p-i-n multilayer (12-nm p-layer/400-nm i-layer/20-nm n-layer) was then deposited on the light-trapping electrode by high-density inductively coupled plasma CVD (ICP-CVD),<sup>15</sup> which can produce a-Si:H thin-films of low defect at the substrate temperature of  $140^\circ\text{C}$ . A 80-nm-thick indium tin oxide (ITO) and a 500-nm-thick Al metal were then deposited on the ICP a-Si:H p-i-n multilayer as the back-electrode. The mono-dispersed Au-NPs of 15 nm in size in water solution were synthesized by the citrate chemical reduction method at low temperatures.<sup>17</sup> To form the self-assembled Au-NPs monolayer on the micro-textured FTO substrate, we sequentially immersed the bare FTO-covered glass substrate in the 3-aminopropyltrimethoxysilane (APTMS)/ethanol solution and the solution containing dispersed Au-NPs. The APTMS forms strong chemical bonds with FTO and can selectively attach to Au-NPs via the terminal ligand (-C-NH<sub>3</sub>). The deposition of the conformal AZO capping layer (5 nm) on the self-assembled Au-NPs monolayer was carried out using scanning-mode DC-sputtering deposition, which was operated at a low power density (0.75 W/cm<sup>2</sup>), a low pressure (2.4 mTorr), and a low scanning speed (6 mm/s) to reduce the deposition rate (2 A/s). The FTO substrate without Au-NPs was used as the control sample. The photovoltaic performance was characterized by an AM1.5G Global sun simulator (Oriel Sol3A) with the white light of  $1000 \text{ W/m}^2$  irradiance. The light-soaking measurement was performed under the white-light irradiance of  $6000 \text{ W/m}^2$  (6-Sun). The device reached accordingly a steady-state temperature at  $60^\circ\text{C}$  due to the irradiation, thereby accelerating photo-degradation of the device.<sup>15</sup> The defect density of the intrinsic a-Si:H layer on different electrodes was determined by the drive-level capacitance profiling (DLCP).<sup>18,19</sup>

FIG. 1. (a) The schematic diagram of p-i-n-type a-Si:H thin film solar cell for micro-structured electrode of FTO (MS), plasmonic-structured electrode of FTO/Au-NPs (PS) and FTO/Au-NPs/AZO hybrid electrode (HPS); (b) 15 nm Au-NPs self-assembled on FTO electrode; (c) the cross-sectional TEM image of p-i-n-type a-Si:H thin film solar cells on HPS electrode; (d) the HRTEM image across FTO, Au-NPs, AZO, and a-Si:H p-layer.

Figure 1(a) schematically shows three structures of the p-i-n a-Si:H thin film solar cell; one is with the micro-structured FTO electrode (MS), one with the FTO/Au-NPs plasmonic electrode (PS), and one with the FTO/Au-NPs/AZO hybrid electrode (HPS). The SEM image of Fig. 1(b) shows that Au-NPs of 15 nm in size are well dispersed on the FTO substrate with the NPs density of  $\sim 2 \times 10^{10} \text{ cm}^{-2}$ . Excessive Au-NPs can reduce light absorption of the active layers and may result in a non-conformal AZO capping, which may induce microstructure defects, such as cracks or voids, across the AZO/a-Si:H interface. Figures 1(c) and 1(d) show the cross-sectional transmission electron microscopy (TEM) images of the p-i-n a-Si:H thin film solar cell with the FTO/Au-NPs/AZO structure. The conformal AZO layer tends to smoothen the Au-NPs dispersed FTO electrode, and microstructural defects are absent across the AZO layer, the p-layer and the intrinsic a-Si:H layer.

The current-voltage (I-V) characteristics of the a-Si:H thin film solar cells integrated with different front-side light-trapping electrodes are shown in Fig. 2. Table I lists the photovoltaic characteristics of the three light-trapping-structured solar cells. According to our previous study, the ICP intrinsic a-Si:H layer has a low-defect density of  $3 \times 10^{15} \text{ cm}^{-3}$ .<sup>15,19</sup> The MS-PV shows a high  $J_{sc}$  of  $16.3 \text{ mA/cm}^2$  with the conversion efficiency as high as 9.6%. In addition, the device has a low dark saturation current of  $1.2 \times 10^{-9} \text{ A/cm}^2$  and a high FF of 70.4%. When plasmonic Au-NPs are deposited on the micro-structured FTO electrode (PS-PV), the solar cell shows a degraded photovoltaic performance with  $J_{sc} = 15.5 \text{ mA/cm}^2$ , FF = 67.8%, and  $\eta = 8.85\%$ . On the other hand, the insertion of the AZO layer between the p-layer and

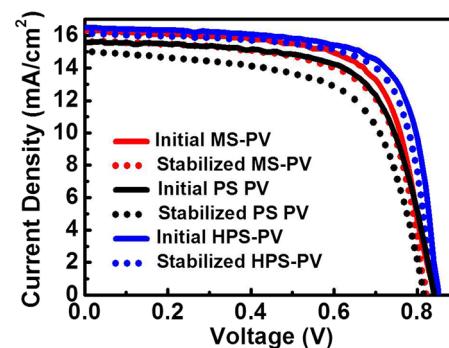


FIG. 2. The I-V characteristics of MS-, PS-, and HPS-PV devices before (initial state) and after (stabilized state) light-soaking.

TABLE I. The cell-performances comparison for the MS-, PS-, and HPS-PVs before (initial state) and after (stabilized state) light-soaking test. The cell area is  $1\text{ cm}^2$ .

Cell type	$V_{oc}$ (volt)	FF (%)	$J_{sc}$ (mA/cm $^2$ )	$\eta$ (%)
MS-PV (initial state)	0.84	70.4	16.3	9.6
MS-PV (stabilized state)	0.83	62.6	15.9	8.4
PS-PV (initial state)	0.84	67.8	15.5	8.85
PS-PV (stabilized state)	0.82	62.0	15	7.65
HPS-PV (initial state)	0.84	72.4	16.6	10.1
HPS-PV (stabilized state)	0.84	68.3	16.3	9.4

the Au-NPs dispersed FTO electrode leads to an increase in the FF (72.4%) and  $J_{sc}$  (16.6 mA/cm $^2$ ), thereby greatly enhancing the conversion efficiency of the HPS-PV cell to 10.1%. The improved FF of the HPS-PV cell may be attributed to the more stable AZO/a-Si:H interface in comparison with the PS-PV cell. Because of the stable interface, the HPS-PV cell is less vulnerable to photo-degradation and, therefore, exhibits a higher conversion efficiency after light-soaking (stabilized state) as to be discussed later.

Figure 3(a) shows the initial QE spectra of the three PV cells before light-soaking. The QE of the PS-PV cell is lower in the range of 350–500 nm than that of the MS-PV cell. However, with the conformal AZO capping layer on Au-NPs, the QE of the HPS-PV cell is comparable to that of the MS-PV cell in the ultraviolet-blue band. The lower QE of the PS-PV cell in the ultraviolet-blue band is likely due to the optical loss and microstructural defects induced by Au-NPs. However, the presence of the AZO capping layer reduces the microstructural defects at the AZO/a-Si:H interface and compensates the photocurrent loss via the stable interface. Moreover, by properly adjusting the size and density of Au-NPs, and the dielectric constant of the surrounding media, one can match the localized surface plasmon resonance (LSPR) wavelength of metal-NPs in a hybrid

plasmonic light-trapping structure with the specific photoresponse band of an a-Si:H PV device.<sup>20,21</sup> In this study, we carefully optimized the fabrication conditions of the HPS electrode, including the size and density of Au-NPs and the thickness of the AZO layer, to enhance the plasmonic effect in the green-red band. The photocurrent loss in the short wavelength range can be effectively compensated by the gain of the enhanced electromagnetic field due to the Au-NPs/AZO induced plasmonic effect. Therefore, more hole carriers are photogenerated in the active layer by the enhanced field and are extracted from the p-layer to the AZO layer, leading to a higher photocurrent in the green-red band.<sup>11</sup> As a result, the HPS-PV cell has a better quantum efficiency in the range of 550–650 nm than the MS-PV cell.

The reverse-bias QE method is usually employed to study the photovoltaic behavior of photogenerated carriers trapped by interface and bulk defects because the applied reverse-bias can increase the electric field across the p-i-n multilayer and, thus, enhance the extraction of photogenerated carriers. Figures 3(b)–3(d) show the QE spectra under the zero-bias (0 V) and the reverse-bias (−1 V) for the three PV devices. In a reverse-bias QE spectrum, a higher QE indicates that more photogenerated holes and electrons are trapped by defects near the p-i-layer and the i-n-layer interfaces, respectively.<sup>22,23</sup> The reverse-bias QE of the HPS-PV cell is lower in the ultraviolet-blue wavelengths regime than that of the MS-PV and PS-PV cells, suggesting a lower defect density across the interfaces. This is likely because the conformal AZO layer encourages the formation of the interface of low defect density with the intrinsic a-Si:H layer. As a result, the photocurrent loss can be compensated by a better carrier collection due to the improved microstructural property of the interface.

Figure 4(a) is the energy dispersive spectroscopy (EDS) mappings that show the elemental distribution in a selected area of the HPS-PV cell. From the mappings, Au-NPs are strictly confined in the AZO capping layer without

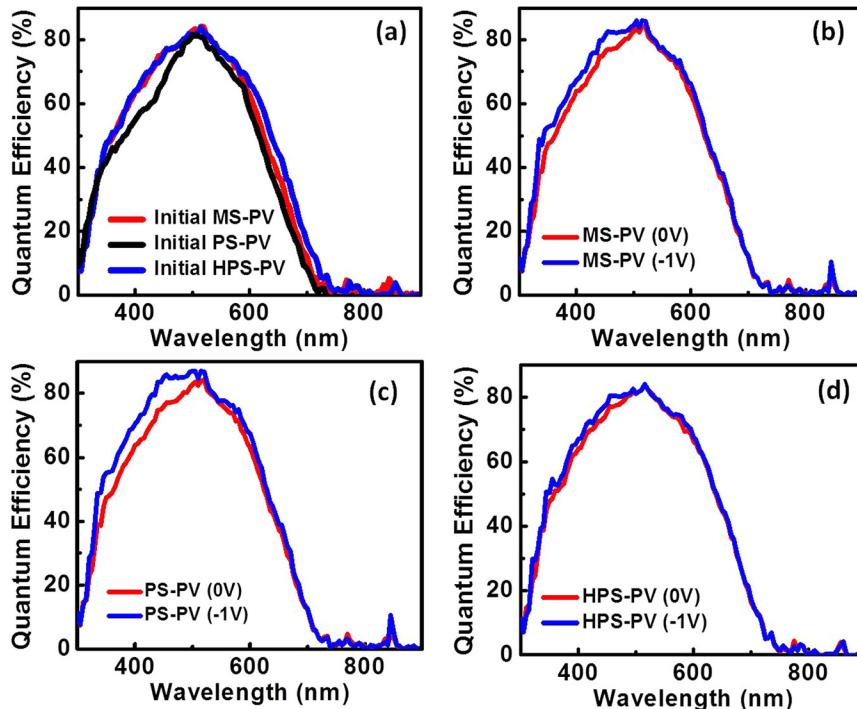


FIG. 3. (a) The initial QE of MS-, PS-, and HPS-PV devices and the QE operated under 0 V and −1 V bias for (b) MS-PV device, (c) PS-PV device, and (d) HPS-PV device.

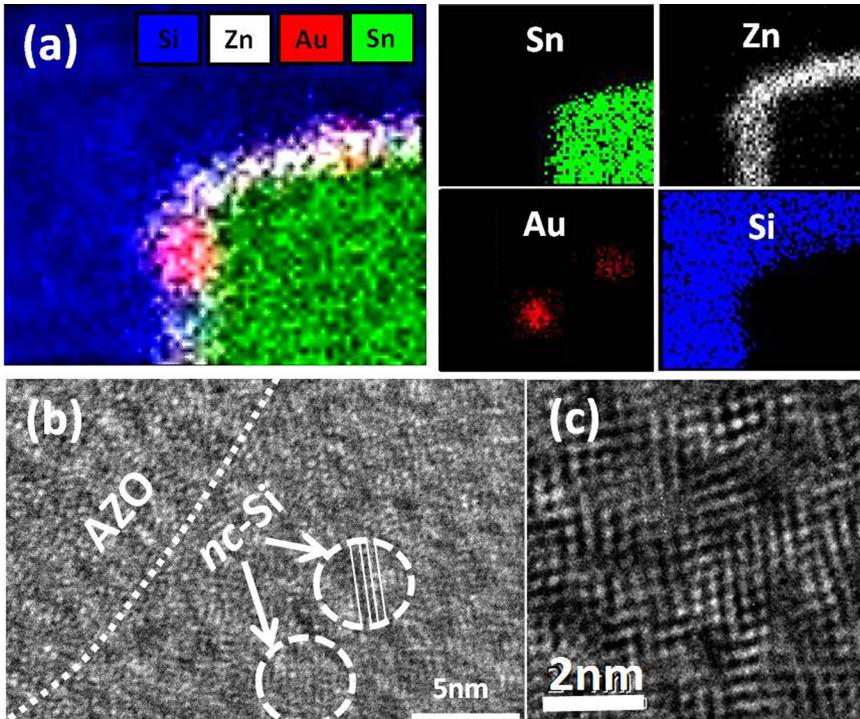


FIG. 4. (a) The elemental distribution in a selected area of the HPS-PV cell for Sn, Zn, Au, and Si by TEM-EDS, (b) the dispersed nc-Si in the a-Si:H p-layer, and (c) the inverse fast-Fourier transform image of nc-Si.

perceivable Au diffusion into the a-Si:H multilayer. The high resolution TEM (HRTEM) image in Fig. 4(b) shows the presence of crystalline Si nanograins near the interface between the p-layer and the AZO layer as indicated by the dashed circles and the inverse fast-Fourier transform image in Fig. 4(c). Raman spectroscopy also revealed the formation of nanocrystalline-Si (nc-Si) in the p-layer of the HPS-PV cell (not shown). The nc-Si grains were not found in the p-layer of the MS- and PS-PV cells, suggesting that the conformal AZO layer induces the partial crystallinity. Because the a-Si:H/nc-Si mixed-phase layer is analogous to the polymorphous Si (pm-Si) in the aspect of microstructure, it can exhibit a better photocarrier transport efficiency compared with the a-Si:H layer.<sup>24,25</sup>

Figure 5(a) shows the  $QE_{loss}$  of the three PV cells as a function of the light wavelength. The  $QE_{loss}$  is herein defined by the ratio of QE at  $-1$  V to QE at  $0$  V. All the three cells demonstrate a higher  $QE_{loss}$  in the ultraviolet-blue wavelengths range and a lower  $QE_{loss}$  in the green-red wavelengths range, suggesting that trapped charge carriers near the p/i-layer interface influence the photovoltaic behavior much more than those near the i/n-layer interface.<sup>22,23</sup> The high  $QE_{loss}$  in the ultraviolet-blue wavelengths range primarily arises from the recombination of photogenerated electrons with holes trapped by defects near the p/i-layer interface.

Therefore, the lowest  $QE_{loss}$  of the HPS-PV cell in the ultraviolet-blue wavelengths range can be ascribed to a less hole accumulation across the p/i-layer interface. In addition to a smaller defect density at the interface, an efficient collection of photogenerated carriers can also improve the  $QE_{loss}$ . It has been reported that the high work function of the AZO layer can moderate the abrupt band bending across the AZO/a-Si:H interface leading to a barrier height lowering and, thus, to a better collection efficiency of hole carriers.<sup>26,27</sup> Therefore, the introduction of the AZO layer may result in the barrier height lowering between the electrode and the p-layer and, thus, yields the better  $QE_{loss}$  of the HPS-PV cell.

To study the influence of the three transparent electrodes on the film quality of the intrinsic a-Si:H layer, we measured the bulk-defect density of the intrinsic a-Si:H layer after light illumination at  $120$  °C for  $10^4$  s by DLCP.<sup>15,18,19</sup> Figure 5(b) shows the integrated defect density ( $N_D$ ) of the PV cells as a function of the depth from the top of the p-i-n active layer after light-soaking. The HPS-PV cell exhibits the lowest bulk-defect density of  $\sim 6 \times 10^{15} \text{ cm}^{-3}$ , and both the MS- and the PS-PV cells have a  $N_D$  higher than  $1.4 \times 10^{16} \text{ cm}^{-3}$ . The DLCP analysis suggests that the FTO/Au-NPs/AZO structure facilitates the growth of an intrinsic a-Si:H layer of low-defect density, which improves the stability of the HPS-PV cell after light-soaking.

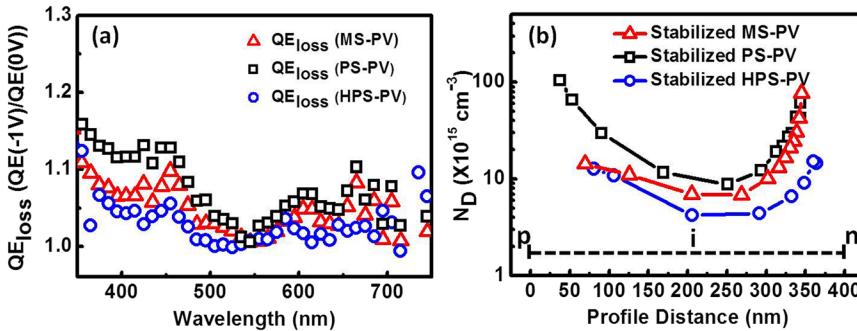


FIG. 5. (a) The normalized  $QE_{loss}$  ( $QE(-1 \text{ V})/QE(0 \text{ V})$ ) of MS-, PS-, and HPS-PV devices as a function of wavelength. (b) The depth profile of bulk-defect densities in the intrinsic a-Si:H layer retrieved from DLCP measurements for MS-, PS-, and HPS-PV devices at  $120$  °C after  $10^4$ -s exposure with a light irradiance of  $6$  Sun.

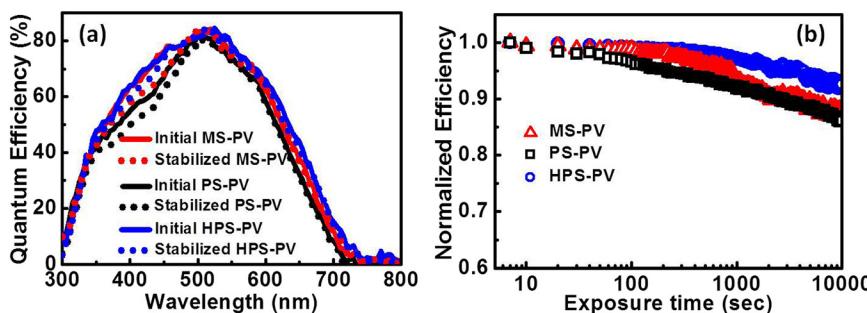


Photo-degradation of a-Si:H PV devices resulting from the Staebler-Wronski effect (SWE) is the major obstacle to the practical use of PV devices because the induced metastable defects during light-soaking enhances the recombination of photogenerated carriers. Figure 6(a) shows the QE spectra of the three PV cells before (initial state) and after (stabilized state) the light-soaking process. These cells demonstrate very slight photo-degradation in the green-red wavelengths range after the light-soaking. On the other hand, obvious photo-degradation occurs to all the three cells in the range of 350–500 nm. However, HPS-PV cell shows a higher resistance to photo-degradation than the MS- and PS-PV cells. Figure 6(b) shows the conversion efficiency of the PV cells subjected to light exposure of 6-sun irradiance at 60 °C as a function of the light exposure time. According to the figure and Table I, the conversion efficiency of the HPS-PV cell decreases by about 7.0% after the light exposure of 10<sup>4</sup> s, and the MS- and PS-PV cells show a decrease in the conversion efficiency of 12.5% and 13.6%, respectively. In combination of the hybrid plasmonic-structured electrode and the ICP-a-Si:H p-i-n multilayer of low defect density, the HPS-PV cell clearly demonstrates a high efficiency and a high stability.

In summary, the a-Si:H thin film solar cell, which consists of the hybrid plasmonic-structured AZO/Au-NPs/FTO electrode and the a-Si:H active layers of low-defect density, has a high photovoltaic efficiency and stability. The hybrid plasmonic light-trapping structure exhibits a broadband light harvest, a high conversion efficiency of 10.1% and a slight photo-degradation as small as 7% after light-soaking. The good photovoltaic properties result from the introduction of the conformal AZO ultra-thin layer between the active layers and the Au-NPs dispersed FTO substrate. The AZO layer encourages the growth of the a-Si:H layers of low defect density and, thus, improves the interface properties between the a-Si:H active layers and the electrode. As a result, the plasmonic a-Si:H solar cell has an enhanced efficiency in the green-red band and a high resistance to photo-degradation in the ultraviolet-blue wavelengths regime. This multi-functional light-trapping electrode structure has a great application potential for the a-Si:H thin film solar cell technology, in which devices of high performance and high stability are extremely desirable.

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FIG. 6. (a) The QE variation of MS-, PS-, and HPS-PV devices at initial state and stabilized state, (b) the conversion efficiency degradation of MS-, PS-, and HPS-PV devices as a function of exposure time with a light irradiance of 6-Sun at 60 °C.

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