A facile light managing strategy in inverted perovskite solar cells

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Abstract

A simple and facile light managing strategy has been proposed in this work to promote the light harvest in inverted perovskite solar cells (PSCs). Effective light managing structures are realized on the substrate back surface by assembling two-dimensional hexagonal closely packed (2D-HCP) SiO\textsubscript{2} nanoparticles with different diameters. The 100 nm 2D-HCP SiO\textsubscript{2} structure, which mainly benefits from the graded refractive index along the light incident route, possesses an effective reflectance reduction of more than 10% in a wide incident angle range. Consequently, the efficiency of inverted PSCs has been substantially improved from 17.24% to 19.12%.

1. Introduction

Organic-inorganic hybrid perovskite materials have become the focus of extensive academic research due to their excellent optoelectronic properties such as high carrier mobility, low exciton binding energy, bipolar charge transport and high light absorption coefficient [1–4]. Organic-inorganic hybrid perovskites were introduced in photovoltaic devices as early as 2009 reported by Miyasaka et al [5]. In the following years, the photovoltaic conversion efficiency (PCE) increased rapidly, and has now surpassed 25% [6]. Perovskite solar cells (PSCs) are mainly categorized into regular structures (n–i–p) and inverted structures (p–i–n) [7]. Inverted structures have the advantage of low temperature solution fabrication suitable for flexible substrates, negligible current density–voltage (J–V) hysteresis, and are capable of preparing efficient tandem cells, nevertheless the champion efficiency is still lower than that of n–i–p structures [8–16]. In order to further improve the efficiency, various strategies including crystallization control, defect passivation and interface engineering, etc are explored to obtain high quality perovskite films and inhibit carrier recombination [17–20]. In the optical aspect, there is still a lot of possibility to further improve the device absorption, by suppressing the reflection loss, parasitic absorption and improving optical intensity coupled inside the active layer.

Ascribed to the excellent optoelectrical properties, suitable energy levels, efficient carrier extraction and transportation, p-type organic materials such as poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), poly(bis(4-phenyl)(2,4,6-trimethylphenyl)amine) (PTAA), and 2,2',7,7'-tetakis (di-p-tolylamino)spiro-9,9'-bifluorene (Spiro-TTB) are commonly adopted and sandwiched between ITO glass and perovskite film as the hole transporting layers (HTLs) in inverted PSCs. However, organic material generally possesses a relatively low refractive index in the range ~1.35 to ~1.6, whereas this is smaller than the value of ITO (~1.64) and perovskite film (~2.55) [21–24]. Therefore, refractive index is mismatched along the light incident path, which will inevitably enhance the light reflection around the ITO/HTL/perovskite interface. Meanwhile, part of the incident light will be routinely reflected on the device surface (air/glass interface). Therefore, from the light harvest point of view, light management is extremely important to further elevate the inverted PSCs performance.
Previously, numbers of optical strategies have been involved [25–28]. Photonic structures, like grating structures formed on the buffer layer or active layer, can realize nanophotonic light trapping by diffraction [29, 30]. Plasmonic nanostructures, such as Au@Ag core–shell nanocuboids, can produce localized surface plasmon resonances, which will enhance the electric field intensity inside the active layer and finally contribute to total light intensity [31]. An antireflective layer is another universal method, for example, random scattering interface in the front or rear side, is able to significantly reduce the reflection loss in the broad spectra, and meanwhile lengthen the optical path in the active layer [32]. Recently, two-dimensional periodic structural light management layers are also successfully calculated in PSCs and realize anti-reflection and PCE enhancement [33].

In this work, we demonstrate a facile and promising strategy aiming to boost the light harvest of inverted PSCs. Two-dimensional hexagonal closely packed (2D-HCP) SiO$_2$ nanoparticle (NP) arrays with different diameters were carefully spin coated on the back surface of substrate before solar cell fabrication, this can avoid any influences on the fabrication and performance of the device. Promoted light harvest in a wide wavelength range of 300–800 nm was achieved by systematically tuning the particle size. Detailed analysis on both calculation and experimental results revealed that the light reflection in the whole wavelength range can be effectively suppressed. Consequently, the PCE of inverted PSCs with the structure of Glass/ITO/PEDOT: PSS/MAPbI$_3$/PCBM/BCP/Au has been elevated considerably from 17.24% to 19.12%. EQE spectra shows a good correlation with the reflectance in the whole wavelength range, while short-circuit current ($J_{sc}$) increases from 19.86 mA cm$^{-2}$ to 21.28 mA cm$^{-2}$. This approach provides a facile and universal way to enhance the light harvest and efficiency in PSCs. Compared with other light trapping approaches, the facile light managing structure proposed here can be directly covered on the PSCs, which can be entirely separated from the device fabrication process and avoid redundant investigation.

2. Experimental

2.1. Materials
PEDOT:PSS (Clevios AI 4083) was bought from Shanghai Zaofu New Material Technology Co., Ltd. Methylammonium iodide (MAI) was obtained from Shanghai MaterWin New Materials Co., Ltd. Dimethylsulfoxide (DMSO, 99.8%), $N,N$-dimethylformamide (DMF, 99.8%), chlorobenzene (CB), isopropyl alcohol and [6,6]-Phenyl C$_6$1 butyric acid methyl ester (PC$_6$1 BM) were purchased from Sigma-Aldrich. Lead iodide (PbI$_2$), guanidine iodide, and methylamine acetate solution (MAAc) were purchased from Xi’an Polymer Light Technology Corp. Bathocuproine (BCP, 98%) was purchased from Aladdin. The concentration of SiO$_2$ NPs latex solution is 2.5 wt%.

2.2. Device fabrication
Perovskite precursor solution was prepared by mixing 1.17 M of MAI, 0.13 M of GAI and 1.3 M of PbI$_2$ (molar ratio = 0.9:0.1:1) into a DMF and DMSO blend solvent (volume ratio = 9:1) with stirring for 12 h at room temperature, and then adding the MAAc ionic liquid at a volume ratio of 11%. ITO substrates (2.0 $\times$ 2.0 cm$^2$) were cleaned by sequential sonication of detergent, deionized water, acetone, and isopropyl alcohol. Cleaned ITO glass substrates were dried under nitrogen flow and SiO$_2$ NPs with different diameters (100 nm or 300 nm) were spin coated on the glass surface at a speed of 4000 rpm (for 100 nm particle) or 1500 rpm (for 300 nm particle) for 30 s. Then the substrates with light trapping structure were treated with ultraviolet ozone for 20 min. The PEDOT:PSS solution was filtered through polytetrafluoroethylene filters (0.45 $\mu$m) before use and then spin coated onto the ITO substrates at 4000 rpm for 30 s, followed with a thermal annealing at 130 $^\circ$C for 10 min in an ambient atmosphere. Perovskite precursor solution was spin coated on the PEDOT:PSS film at 1000 rpm for 10 s and 3500 rpm for 75 s, then the sample was annealed at 100 $^\circ$C for 5 min. The PC$_6$1 BM solution of 16 mg ml$^{-1}$ in CB was spin casted on perovskite film at 1500 rpm for 30 s. BCP solution of 1 mg ml$^{-1}$ in isopropanol was spin coated on the PC$_6$1 BM film at 4000 rpm for 30 s. Finally, a 100 nm thick Au electrode was thermally evaporated above.

2.3. Characterizations
X-ray diffraction patterns of the perovskite films were performed by a Rigaku ATX-XRD diffractometer with out-of-plane grazing incident diffraction mode, employed Cu Kα radiation ($\lambda = 1.5406$ Å) as the radiation source, while the incident angle was fixed to 1.2° and the out-of-plane exit angle was scanned from 5° to 60°. Top and cross-sectional views of scanning electron microscopy (SEM) images were obtained by using a field emission scanning electron microscope (Jeol, JSM-6700F). An AFM image of perovskite film surface morphology was measured with an atomic force microscope (NanoNavi-SPA400, Japan). PL and TRPL were characterized by a fluorescence spectrophotometer (FSS, Edinburgh instruments). Photocurrent density–voltage ($J$–$V$) curves of the devices were measured by a Keithley 2400 Digital Source Meter under
AM 1.5G illumination with a xenon-lamp-based solar simulator (HAL-320, ASAHI SPECTRA Co. Ltd, Japan), which was calibrated by Asahi Spectra Co., Ltd. The power of incident light was calibrated with a standard silicon photodiode before every measurement. And a metal mask (0.089 cm$^2$) was used to define the active area of the solar cell. The $J–V$ curves were recorded by scanning the bias voltage from 1.3 V to $-0.2$ V (reverse scan) and then $-0.2$ V to 1.3 V (forward scan), respectively, with a delay of 50 ms. EQE spectra were performed on an EQE system (CEP-2000MLQ, Bunkoukeiki Co., Ltd) in the DC mode without any voltage bias. UV–vis–NIR spectrophotometer (Cary 5000, VARIAN) was used to characterize the integral transmittance, direct transmittance, integral reflectance, and absorbance.

3. Results and discussion

The structure of the modified inverted planar PSCs is shown in figure 1. Figure 1(a) illustrates the 3D numerical model with the structure of 2D-HCP SiO$_2$ NPs/Glass/ITO/PEDOT:PSS/MAPbI$_3$/PCBM/BCP/Au, while figures 1(b) and (c) give the typical SEM images of the fabricated device and 2D-HCP SiO$_2$ NPs array, respectively.

In order to explore the light managing effect of 2D-HCP SiO$_2$ NPs array on perovskite, evaluated simulation was initially performed with the structure of 2D-HCP SiO$_2$ NPs assembled on the surface of the perovskite layer directly. Finite difference time domain algorithm calculations were performed systematically on a 2D array of hexagonal lattice structures, where a 400 nm thick perovskite active layer was used and the XY-plane was fixed at the 100 nm deep location in order to obtain a meaningful image of light electric field intensity for certain wavelength. A perfectly matched layer (PML) boundary condition is adopted along the incident direction in order to prevent nonphysical scattering at the boundary; periodic boundary condition is utilized on the XY-plane to conveniently simulate the entire array [34, 35]. The electric and magnetic field distribution in the entire domain can be achieved by analyzing the periodic structure, which in turn can describe the propagation characteristics of electromagnetic waves in the wavelength range of 300–800 nm [36–38].

The normalized 2D electric field distribution with different structures were calculated from the divergence of the Poynting vector in the XZ-plane at short (450 nm), median (580 nm) and long (700 nm) wavelengths, as shown in figure 2, in which the light illuminates from the top surface. For the reference case without 2D-HCP SiO$_2$ NPs, the electromagnetic field intensity shows obvious interferences from stacked layers at all the three wavelengths (450 nm, 580 nm and 700 nm) in figures 2(a), (d) and (g). It tends to be managed after employing 2D-HCP SiO$_2$ NPs. For 2D-HCP with 100 nm diameter, much smaller than the incident wavelength, the electric field intensity has been enhanced substantially in the whole structure. When a larger 2D-HCP structure with 300 nm diameter which is comparable to the incident wavelength was introduced, both the cavity resonances and Mie scattering effects can be identified [39, 40], as shown in figures 2(c), (f) and (i).

The detailed electric field distribution in the XY-plane in the perovskite layer with a Z-value of 300 nm was also extracted for further analysis as shown in figures 3(a)–(f) with different structures at certain
wavelengths. The features of strong photonic management arising from the resonant coupling of incident photon to a multitude of distinct modes, intuitively termed as ‘resonances’ could be clearly distinguished.

From the perspective of the electromagnetic model, when the light irradiates on the interface of the medium with different refractive index, the light trapping structure can stimulate two common optical transmission modes categorized as waveguide resonance and diffraction, respectively. The waveguide resonance can be generated under the condition of impedance matching, in turn, couple the light to the active layer. On the other hand, the trapping structure can be regarded as a grating when the eigenperiod is at a specific value and as a result, diffraction occurs when the light irradiates onto the surface. Both modes tend to confine the light into a certain area, resulting a significant enhancement of the electric field intensity in the active matrix.

For the 300 nm HCP structure with comparable size to the incident wavelength, the deviation of electric intensity is enlarged to more than 25%. As these resonances are much more dependent on the incident wavelength, the overall effect of both the cavity resonances and Mie scattering are performed differently as shown in figures 3(b), (d) and (f), and make the average electric field intensity enhanced mainly at short (450 nm) and long (700 nm) wavelengths as illustrated in figure 4. While for the small HCP structure (100 nm HCP), the deviation of electric intensity is much smaller, less than 0.1%. It is worth mentioning that as the particle size is much smaller than the incident wavelength for the 100 nm 2D-HCP SiO$_2$ NPs, the electromagnetic field distribution has been enhanced substantially and uniformly in the whole perovskite layer by a factor of 9.5% at 580 nm (increased from 0.42 to 0.46).

This is mainly benefiting from the graded refractive index along the light incident route constructed between SiO$_2$ NPs array and the air filled inside. This gradient interface makes a great contribution to suppress the interfacial Fresnel’s reflection and enlarges the electric field intensity in the whole wavelength range.

In addition, besides the perpendicular incidence mode, the 2D-HCP structure also demonstrated the excellent antireflection effect in a wide incident angle range from 0° to 30° as shown in figure 5. The average reflectance in the wavelength range of 300–800 nm for the incident angle of 0°, 15° and 30° can be reduced
Figure 3. The $XY$-plane light electric field intensity in perovskite with ((a), (c), (e)) 100 nm and ((b), (d), (f)) 300 nm 2D-HCP SiO$_2$ NPs at wavelengths of 450 nm, 580 nm and 700 nm, respectively.

Figure 4. Average electric field intensity in the whole perovskite layer with different structures at certain wavelengths.
Figure 5. Corresponding surface reflectance in terms of incident angle without (solid lines) or with 100 nm 2D-HCP structure (dashed lines).

Figure 6. (a) Measured integral transmittance, (b) haze factor on glass, (c) Integral reflectance of glass/ITO/PEDOT:PSS/perovskite structure, covered with or without the 2D-HCP SiO$_2$ NPs array with a diameter of 100 nm and 300 nm, respectively.

from 22.97%, 22.17% and 21% to 11.06%, 10.96% and 11.24% respectively after employing 100 nm HCP. Meanwhile, the reflectance fluctuation is concomitantly reduced by 85.77%. This indicates that this facile light managing strategy possesses potential practical applications in outdoor circumstance. Under the actual light irradiation condition with varying intensity and angle, the 2D-HCP structure can suppress the light reflection, moreover, a more stable power output can be achieved.

Based on the systematic calculations discussed above, discreetly engineered the 2D-HCP structure is realized on the back surface of ITO glass by spin casting SiO$_2$ NPs with different diameters before solar cell fabrication. This procedure could avoid the influences on the fabrication and performance of the device effectively. Fabrication conditions are optimized in order to realize the tightly packed single layer and match well with the ideal arrangement as shown in figure 1(c), which illustrates the typical SEM images of resulted 2D-HCP SiO$_2$ NPs.

Measured optical properties including integral transmittance and scattering factors of 2D-HCP SiO$_2$ NPs with different diameters are shown in figures 6(a) and (b). It is shown that introducing larger SiO$_2$ NPs (300 nm) results in superior optical transmission in the wavelength range comparable to the characteristic size ($\sim$300 nm to $\sim$400 nm), which is mainly ascribed to the cavity resonances and Mie scattering effects as described in the simulation section. On the other hand, an extraordinary optical transmission in the whole wavelength range ($\sim$300 nm to $\sim$800 nm) has been realized by employing small particles (100 nm). And profiting from the refractive index gradient, the average integral transmittance (from 300 nm to 800 nm) are enhanced from 89% to 94% for 100 nm 2D-HCP SiO$_2$ NPs.

The scattering spectra demonstrated that the 300 nm 2D-HCP SiO$_2$ NPs yield a significant forward-scattering mainly resulted from the photonic modulating. This indicates that more light will be diffracted into the angular deviation domain from the optical axis when the critical dimensions are comparable to the illuminating wavelengths, accordingly, extending the light path and eventually results in a higher transmittance especially in the short wavelength range [41]. While the scattering is quite limited for the smaller SiO$_2$ NPs as the diameter is much smaller than the incident wavelength.

After integrating in PSCs, the reflection spectra on the solar cell surface are evaluated as shown in figure 6(c) using the structure of 2D-HCP SiO$_2$ NPs/Glass/ITO/PEDOT:PSS/perovskite, with the light irradiated from the glass side. The difference in reflectance is still pronounced although perovskite possesses a
highlight absorption. The pristine sample without 2D-HCP structure reveals a serious reflection, the average reflectance in the UV and visible range can be higher than 30%, fundamentally limiting the promotion of solar cell efficiency ascribed to the insufficient light harvest. This partially originates from the light reflection on the device surface in the case of refractive index salutation from air to glass \[42\]. On the other hand, organic HTLs generally possess a relatively lower refractive index than ITO glass and perovskite film fabricated on both sides \[24, 43–45\]. Thereby, the refractive index is mismatched along the light penetration route, which will also enhance the light reflection around the ITO/HTL/perovskite interface. The 2D-HCP SiO$_2$ NPs array integrated above not only suppresses the refractive index mismatch on the air/glass interface, but also provides an effective management for the light, thereby more light can be coupled into the perovskite layer. Thus in the following, inverted planar PSCs with the structure of Glass/ITO/PEDOT:PSS/MAPbI$_3$/PCBM/BCP/Au are fabricated, as shown in figure 1(a). The MAPbI$_3$ active layer is prepared by a one-step method without using any antisolvent. Methylammonium acetate (MAAc) is incorporated into the perovskite precursor solution to tune the crystal growth and orientation of crystals. A high quality perovskite film with thickness of $\sim$500 nm and the grain size larger than 1 $\mu$m are achieved, which guarantees the sufficient light absorption and facilitates the efficient carrier transportation \[6, 46–49\].

Figure 7(a) illustrates the typical $J$–$V$ curves of the pristine device and the device employing SiO$_2$ NPs trapping array. Derived device photovoltaic parameters are summarized in table 1. Relatively low PCE has been achieved for the pristine device, with a $J_{sc}$ of 19.86 mA cm$^{-2}$, open voltage ($V_{oc}$) of 1064 mV, fill factor (FF) of 81.54%, and PCE of 17.24%. However, it is worth mentioning that FF with a value as high as 81.54% has been succeeded in the pristine device, taking into consideration the low series resistance ($R_s$) and high shunt resistance ($R_{sh}$) as illustrated in table 1. This provides compelling evidence of a high quality pristine device with adequately annihilated carrier recombination loss. This is probably benefiting from the high quality of perovskite film with large grain size and a uniform surface, and promoted interface performance with a mitigated carrier recombination process. In addition, FF changes slightly in devices involved in this work, which comes from the reasonable fluctuations although all solar cells are fabricated with the identical
Table 1. Detailed photovoltaic parameters of the pristine device and devices with SiO$_2$ NPs trapping array in different diameters.

| SiO$_2$ NPs diameter (nm) | $J_{sc}$ (mA cm$^{-2}$) | $V_{oc}$ (mV) | FF (%) | PCE (%) | $R_{sh}$ (Ω cm$^2$) | $R_{s}$ (Ω cm$^2$) |
|--------------------------|-------------------------|----------------|--------|---------|---------------------|------------------|
| Pristine                 | 19.86                   | 1064           | 81.5   | 17.24   | 4316.5              | 2.32             |
| 100                      | 21.28                   | 1064           | 84.4   | 19.12   | 14 163.2            | 1.99             |
| 300                      | 20.99                   | 1064           | 80.9   | 18.09   | 3573.8              | 2.75             |

Process in one-pot. Device PCE is improved substantially by employing 2D-HCP, and mainly originates from the enhanced $J_{sc}$. The $V_{oc}$ presents an almost identical value and the FF is still larger than 80% although with a small fluctuation, which implies that the deposition process of SiO$_2$ NPs array on the back surface of substrate before device preparation has negligible influences on device quality. Device PCE has been elevated from 17.24% to 19.12% after assembling 100 nm 2D-HCP SiO$_2$ NPs. In addition, 36 devices with identical fabrication conditions were fabricated in order to assess the reproducibility. Corresponding average and statistical parameters with narrow distributions are collected in figure 7(b), denoting a good reproducibility of the device. Figure 7(c) shows the corresponding EQE spectra and integrated $J_{sc}$ of devices. The integrated $J_{sc}$ values are 19.24, 20.63, and 20.09 mA cm$^{-2}$, respectively, matching well with the values from the $J$–$V$ characterization.

In the EQE spectra of the pristine device, two obvious shoulders can be distinguished around the wavelengths of 450 nm and 700 nm. This shows a good correlation with the high reflectance in the same region as depicted in figure 6(c). This finding permits the conclusion that the strong light reflection in the pristine device is responsible for the EQE response drop. These drops can be compensated by utilizing SiO$_2$ NPs array, delivering an enhanced EQE response as high as ~80% in the wide range from 400 nm to 750 nm. In order to quantitatively evaluate the enhancement of the EQE response.

4. Conclusion

In summary, a facile and effective strategy has been demonstrated aiming to enhance the light harvest and the efficiency of PSCs by employing the SiO$_2$ NPs trapping array. Both calculation and experimental results verified the feasibility of suppressing light reflection. Moreover, a single layer of SiO$_2$ NPs trapping array in the diameter of 100 nm provides the prominent light harvest by coupling more light in the whole wavelength range. Therefore, device efficiency has been elevated from 17.24% in the pristine device to 19.12% in the champion device, mainly originating from the improved $J_{sc}$ from 19.86 mA cm$^{-2}$ to 21.28 mA cm$^{-2}$. This strategy also provides a convenient and versatile method to enhance the light harvest in perovskite solar cells or tandem devices irrelevant to materials employed.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Conflicts of interest

There are no conflicts to declare.

Contributions of each author

Y W and H P contributed equally to this work; Y W conducted most of fabrication process, H P performed most of calculation works of devices; Q H and Y D developed the basic concept; Y X and M H carried out the
optical measurement; G H helped SEM measurement and analyzed the data; Q H (2018YF1500104), Y Z (B16027), X Z (61674084) and Y D (61874061) directed the project; all authors discussed the results and revised the paper.

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