Ordered array structures for efficient perovskite solar cells

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Funding information
Key University Science Research Project of Jiangsu Province, Grant/Award Number: 17KJA430013; National Natural Science Foundation of China, Grant/Award Number: 51772197; Priority Academic Program Development of Jiangsu Higher Education Institutions

Abstract
In the past few years, metal halide perovskite solar cells (PSCs) have attracted widespread attention in the photovoltaic field, and their power conversion efficiency has rapidly exceeded 25%. Among the adopted effective strategies to realize the high efficiency, the introduction and utilization of ordered array structure in the PSCs are particularly prominent, which can simultaneously enhance light harvesting efficiency and carrier transport properties by manipulating the light paths (reflection, scattering, extraction, and propagation) and constructing direct charge transport paths. The full utilization of sunlight contributes to high short-circuit current density ($J_{sc}$), and strong carriers transport is favorable for high charge collection and reducing charge recombination. Moreover, for flexible PSCs, ordered array structured device can be greatly relaxed during bending, thereby suppressing the formation of cracks in PSCs. Herein, this article focuses on the ordered array structure design, reviews the recent research progress of the ordered array structures in PSCs, and provides some promising perspectives for further improvement.

KEYWORDS
ordered array structure, perovskite, solar cell

1 | INTRODUCTION

Metal halide perovskites (ABX$_3$, A = CH$_3$NH$_3^+$ (MA$^+$), CH(NH$_2$)$_2^+$ (FA$^+$), Cs$^+$, and so on; B = Pb$^{2+}$, Sn$^{2+}$, and so on; X = Cl$^-$, Br$^-$, I$^-$) are considered to be one class of the most promising candidate materials for constructing high-efficiency solar cells owing to their excellent optical and electrical properties. 1-8 Generally, perovskite solar cells (PSCs) are composed of a transparent conductive oxide (TCO) substrate, an electron-transporting layer (ETL), a metal halide perovskite light absorber, a hole-transporting layer (HTL), and a top electrode layer. Among them, the metal halide perovskite layer is used to absorb photons to produce photogenerated carriers, and the carrier transport layer (CTL, including ETL and HTL) is used to extract and transfer photogenerated carriers. In just several years, the PSCs have expanded dramatically and the power conversion efficiency (PCE) has exceeded 25%, 9 which is comparable to the silicon solar cells. The efficiency loss of PSCs mainly originates from the recombination at the imperfect interface and the underutilization of incident light. 10,11 To realize the theoretical PCE of 30%, efficient light harvesting and strong carriers transport are essential. Full use of sunlight can produce high short-circuit current density ($J_{sc}$), and strong carriers transport can improve charge collection and reduce charge recombination.12,13

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Engineering Reports. 2020;2:e12319.
https://doi.org/10.1002/eng2.12319
Nowadays, many strategies have been adopted to enhance the light-harvesting efficiency by reducing light reflection, enhancing light scattering and resonance in PSCs. Among them, designing an ordered array structure with various novel architectures (such as nanorods, nanosheets, nanocones, hemisphere, and so on) into the perovskite absorber layer, CTL or TCO substrate is considered to be a powerful strategy to improve the performance of PSCs (Figure 1). Their dimensions are similar to or smaller than the incident light wavelength, and thus possess the various optical characteristics related to size, shape, and composition. It increases internal scattering, reduces light loss caused by front surface reflection and escape of unabsorbed light, and causes a surface plasmonic resonance effect to capture the incident light, thereby effectively improving light harvesting efficiency. Meanwhile, the ordered array structures have also been found to provide a direct carrier transport pathway, which facilitates charge transport and thus reduces charge recombination. More importantly, the ordered array structures have open channels and large surface to volume ratio, allowing perovskite penetrate into the CTL layer and improving the interfacial contact area between perovskite and CTL, which also helps to carriers transport. In summary, compared to their regular nanostructures or planar films-based devices, the ordered array structured devices not only enhance light harvesting efficiency by improving the light scattering and reducing the light reflection, but also improve the transport properties by constructing direct charge transport paths. Moreover, for flexible PSCs, ordered array structured devices can be greatly relaxed during bending, thereby suppressing the formation of cracks in different layers of PSCs. Therefore, constructing an ordered array structure is an effective strategy that can further improve the performance of PSCs.

This article reviews the recent research progress of the ordered array structure in PSCs. We systematically introduce the design and application of the ordered array structure in the TCO substrate, CTL, and perovskite layer (Table 1). In particular, several representative and effective strategies for constructing ordered array structure are emphasized, and the internal mechanisms for improving their performance are highlighted. Finally, we provide some perspectives for the construction of ordered array structure to further improve the efficiency of PSCs.

2 ORDERED ARRAY STRUCTURES FOR TCO SUBSTRATE

TCO substrate is one of the basic components of PSCs, which plays an important role in the extraction and transportation of photogenerated carriers. In addition, in almost all PSCs, the incident light must pass through the substrate and then absorbed by the perovskite film, which means that the influence of TCO substrate on light also plays an obvious role in the performance of PSCs. Therefore, constructing an ordered array structure for TCO substrate should be an effective strategy to improve the performance of PSCs. More importantly, the ordered array structured substrates may influence the crystal growth, thereby affecting the morphology of the upper film, which may help the upper layer to form an ordered array structure. For example, Luo et al fabricated an inverted planar heterojunction PSCs on corrugated indium tin oxides (ITO) substrate, and the post-deposited Poly[bis(4-phenyl)(4-butylphenyl)amine] (poly-TPD) HTL and perovskite bottom surface both show typical corrugated structure similar to the ITO substrate (Figure 2A). Under such conditions, the absorption of the corrugated device was greatly improved compared to the planar device, which indicated that the corrugated device had an enhanced light harvesting. Thus, a 15% enhancement of $J_{sc}$, 11% enhancement of $V_{oc}$, and 14% enhancement of fill factor (FF) are achieved in the corrugated device (Figure 2B). Similarly, Huang and co-workers fabricated the PSCs on etched nanosphere array structured fluorine-doped tin oxide (FTO) substrates (NAFS) (Figure 2C). The post-deposited TiO$_2$ ETL exhibited a similar periodic hemisphere structure, and the MAPbI$_3$ perovskite film was
deposited uniformly on the TiO₂ ETL. Based on this structure, the average reflectance of NAFS-based device exhibited a 22.16% improvement compared to planar structured device, indicating that more incident light was absorbed by the NAFS-based device (Figure 2D). This trend was also proved by 3D finite-difference-time-domain (FDTD) simulation (Figure 2E,F). Besides, the fast decay and slow decay lifetime of the nanopatterned FTO substrate (NPFS)-based sample were decreased from 7.16 to 6.59 nanoseconds and from 51.78 to 25.15 nanoseconds, respectively, compared to planar structure-based sample, which also proved that the NPFS-based device had a faster electron transport and lower carrier recombination. Eventually, the $J_{sc}$ increased from 19.27 mA cm$^{-2}$ for planar structure-based device to 21.92 mA cm$^{-2}$ for NAFS-based device, and the corresponding PCE increased from 14.21% to 17.85%. In addition, to further obtain higher light transmittance and maintain good electrical conductivity, our group designed one dimensional antimony-doped tin oxide (ATO) nanorod arrays as a conductive mesoporous layer for PSCs. The ATO substrate with suitable Sb doping ratio greatly enhanced the transmission (up to 90%) and resulted in a higher maximum theoretical current density than that of FTO substrate (Figure 2G). Based on the better transmission of ATO substrate and higher quality of perovskite film, the $J_{sc}$ of the optimized device reached 23.8 mA cm$^{-2}$ and corresponding PCE reached 20.1% (Figure 2H,I).

3 | ORDERED ARRAY STRUCTURES FOR ETL

The n-type ETL is also an important part of PSCs, which is mainly used to transport electrons from the perovskite absorber layer and hinder the backflow of electrons. Constructing ordered array structure in ETL has caught extensive
attention, and a large number of materials based on different dimensions (1D, 2D, 3D) have been investigated. In this section, we will review the ordered array structured ETLs according to the dimensions of ETL layer.

### 3.1 1D ordered array structured ETL

1D ordered array (nanorod, nanowire, nanocone, and so on) structured ETLs are considered as direct electron transport paths, and can provide large surface-to-volume ratio and high surface reactivity. Thus, they exhibit a better electrical and optical performance compared to bulk structured ETLs. Controlling the synthesis process of ETL is a key point to obtaining high-quality and high-ordered 1D arrays. Among the various synthesis methods, hydrothermal method is the most common method for preparing 1D ordered array structured ETL, and the length and diameter of 1D arrays can be adjusted by controlling the reaction time and precursor (concentration and additives). For instance, Li et al. fabricated TiO₂ nanocone arrays by a disodium ethylenediamine tetraacetate assisted hydrothermal process. Chen et al. grown CdS nanorod arrays through a hydrothermal process. Peng synthesized ZnO nanorod arrays by using an aqueous solution containing \( \text{Zn(NO}_3\text{)}_2 \cdot 6\text{H}_2\text{O}, (\text{CH}_2)_6\text{N}_4 \) and polyethyleneimine. Due to the advantages of 1D arrays, these ETLs have achieved considerable performance. In addition, it is worth mentioning that Ahmad and co-workers used a unique and scalable physical vapor deposition at oblique angles (PVD-OAD) method to fabricate TiO₂ nanocolumnar, and the thickness can be tuned via controlling the deposited-angle (Figure 3A). After that, they further obtained a nanoforest structured TiO₂ via the PVD-OAD process. The perovskite film deposited on the optimal TiO₂ nanoforest has a great enhancement in the absorbance (up to 90%) due to the reduced transmittance and reflectance. Moreover, the TiO₂ nanoforest-based device exhibited an order of magnitude higher recombination resistance than that of the reference device. The improvement was attributed to the fewer trap sites induced by the TiO₂-nanoforest arrays. Finally, a PCE of 16.8% can be obtained with optimal TiO₂ nanoforest while the pristine device only delivered a PCE of 14.86%.

Doping engineering is an effective strategy to improve the charge mobility and conductivity and tune the energy level of ETL materials. This is more conducive to the carriers transfer from the absorber layer to the CTL layer and then to the electrode layer. So far, various elements (such as Cs, Sn, Cd, and so on) have been doped into the ETL layers to further improve their electrical properties. For instance, Li et al. added the tantalum chloride to hydrothermal precursors solution to prepare Ta-doped TiO₂ nanorods, and the doping phenomenon was confirmed by the X-ray diffraction (XRD) patterns (Figure 3B) and the elemental analysis of transmission electron microscopy (TEM) (Figure 3C). By optimizing doping solubility, the conduction band (CB) of optimal Ta-doped TiO₂ had a 0.05 eV downshift compared to the pristine TiO₂. The fast decay lifetime of perovskite film deposited on optimal Ta-doped TiO₂ nanorods reduced from 9.63 to 5.37 nanoseconds compared to pristine TiO₂ nanorods sample (Figure 3D). These results suggested that Ta-doped TiO₂ had more effective electron injection and hole blocking capabilities. Then, the optimal Ta-doped TiO₂ nanorod array-based device obtained a high PCE of 19.11%, represented a 10% improvement compared to the pristine device. Amassian’s group used conjugated polyelectrolyte polyethyleneimine (PEI) as a dipole layer and a selective polymer capping layer to obtain high aspect ratio electron-rich nitrogen-doped ZnO (N-ZnO) nanowires (Figure 3E). The N-ZnO nanowires provided larger surface contact with perovskite film and perovskite film deposited on it exhibited fewer pinholes. Besides, due to the N-doping and the PEI-coating, the CB of the N-ZnO has a 0.44 eV upshift compared to the pristine ZnO, which is more matched to the CB of the perovskite layer. Based on these advantages, the optimal device delivered a PCE of 16.12% with a \( J_{\text{sc}} \) of 21.5 mA cm\(^{-2}\), while the pristine device only delivered a PCE of 14.41% with a \( J_{\text{sc}} \) of 20.5 mA cm\(^{-2}\).

In addition to the doping engineering, modifying the surface of 1D ETL arrays to form a heterostructured ETL is another effective strategy to improve device performance. The core layer of heterostructured ETL can transfer the photogenerated electrons more quickly and the shell layer can block the backflow of electrons, thereby reducing the recombination between the electrons in the CB of ETL and the holes in the valence band of perovskite absorber. Besides, the shell layer may form a well-matched interface with the perovskite film and maintain better stability when in contact with the perovskite. For instance, Amassian and co-workers fabricated a heterostructured ZnO/TiO₂ core-shell metal oxide arrays for PSCs. The vertically aligned ZnO nanorods were first deposited on FTO substrate, and then TiO₂ shell was decorated by a two-step hydrothermal process. The core-shell structure enhanced the total surface area and provided a superior surface for a better contact interface between the ETL and perovskite film. Simultaneously, the core-shell structure greatly enhanced the light-harvesting. Then the core-shell-based device had an external quantum efficiency (EQE) enhancement of approximate 20% compared to the pristine device at the light wavelength range of 300 to 800 nm (Figure 3F), thereby enhancing \( J_{\text{sc}} \) from 16 to 20 mA cm\(^{-2}\). Finally, the optimal ZnO/TiO₂ core-shell-based device delivered an improved PCE of 15.4% (Figure 3G). Besides, Yu et al fabricated a ZnO/SnO₂ core-shell structured ETL via a
hydrothermal process. The ZnO/SnO\textsubscript{2} device maintained over 90\% of its initial PCE after 25 days, which was more stable than pure ZnO-based (maintain 10\%) or TiO\textsubscript{2}-based device (maintain 70\%) (Figure 3H). The well-matched interface between SnO\textsubscript{2} and perovskite film and lower defects were believed to be the main reasons for improving PSCs stability. Schmuki et al treated the surface of the TiO\textsubscript{2} nanorods with TiCl\textsubscript{4} and PC\textsubscript{61}BM. The treated device exhibited a PCE of 19.5\%, while the pristine device exhibited a PCE of 14.2\%.

3.2 | 2D ordered array structured ETL

Similar to 1D structured ETL, 2D structured ETL is also a promising candidate for optoelectronic devices due to their excellent optical and electronic properties. Compared to 1D materials, 2D materials have higher carrier mobility and are easier to modify for flexible devices. So far, nanosheet arrays are the main 2D structure, and various different materials (such as SnO\textsubscript{2}, SnS\textsubscript{2}, In\textsubscript{2}S\textsubscript{3}, and so on) are used to construct 2D structured ETL. For instance, Yang et al first prepared an In\textsubscript{2}S\textsubscript{3} nanosheet array-based ETL by a low temperature chemical bath deposition process. By optimizing the reaction
time, the optimal device exhibited the lowest photoluminescence (PL) intensity and obtained a highest PCE of 18.22%, which was higher than that of the reference TiO$_2$ ETL-based device (15.70%). Ma et al. fabricated SnS$_2$ nanosheet arrays as ETL by a low temperature hydrothermal method. The proper homogeneously grown SnS$_2$ nanosheet arrays contributed to more effective electron collection and the perovskite particles embedding, and was confirmed by scanning electron microscopy (SEM) images and PL measurements. Finally, the optimal device achieved a champion PCE of 13.63% with a high $J_{sc}$ of 23.70 mA cm$^{-2}$. Fang et al designed a SnO$_2$ nanosheet arrays-based ETL for PSCs, and optimized the preparation time to obtain the best efficiency. Then, they further doped Y into SnO$_2$ to improve the electron transfer capability, and the corresponding PCE improved from 13.38% to 17.29% with significantly reduced hysteresis compared to pristine SnO$_2$ ETL-based device.

3.3  3D ordered array structured ETL

Ordered honeycomb or inverse-opal structured ETL films constructed by colloidal lithography are the common 3D structured ETLs for PSCs. In addition to increasing surface area, promoting light collection and facilitating carrier transportation like other dimensional structured ETL, it also contributes to perovskite to crystallize into large domains and provides the effect of subwavelength grating. In 2015, Snaith’s group first introduced a honeycomb structured TiO$_2$ into PSCs by using a double templating approach (Figure 4A,B). The TiO$_2$ height can be changed by changing the PS microsphere size and TiO$_2$ precursor concentration. After optimizing the TiO$_2$ height and size, the optimal honeycomb structured device exhibited a higher shunting resistance ($4.1 \times 10^9$ Ω) compared to reference device ($1.8 \times 10^4$ Ω), which indicated that the recombination path was significantly hindered in the honeycomb structured device. Then, a 32% improvement of PCE was achieved. In 2016, Park et al used an inverse-opal structured TiO$_2$ ETL for enhancing light harvesting efficiency of PSCs (Figure 4C). Compared to the planar TiO$_2$-based device (~8.5%), the device with inverse-opal

![Figure 4](image-url)
TiO$_2$ exhibited a lower spectral reflectance (~7.7%) (Figure 4D). The reduced reflectance induced a higher light harvesting efficiency, and was confirmed by FDTD simulation. Eventually, the $J_{sc}$ improved by 5.3% and PCE improved by 10.4% by changing the planar TiO$_2$ ETL to the inverse-opal TiO$_2$ ETL. After that, in order to further improve the device performance, Wang et al designed a nanowire-coated, branched macroporous structured TiO$_2$ ETL for PSCs (Figure 4E). This structure combined the advantages of both 1D nanowires and honeycomb structures, resulting in a remarkable PCE of 20.1%, which was a noticeable increase of 16% compared to TiO$_2$ nanowire ETL-based device (PCE = 17.3%). Besides, ordered nanotube arrays were also employed for ETL, which resemble the inverse opal structure with deeper grooves. Unlike the random nanotubes fabricated by hydrothermal methods, anodic self-organization processes, and others. Ho et al obtained an extremely ordered TiO$_2$ spherical nanotube array through a photoresist template process (Figure 4F), which simultaneously ensured the perovskite material filling and the efficient electron transmission. Moreover, compared to planar structured TiO$_2$ ETL, the perovskite on optimal ordered TiO$_2$ nanotube arrays displayed enhanced absorbance over the entire response spectrum range due to the stronger light trapping capability of TiO$_2$ nanotube arrays. Thus, compared to planar device, the $J_{sc}$ of nanotube array-based device enhanced from 12.89 to 21 mA cm$^{-2}$ and the corresponding PCE boosted from 6.75% to 14.13%.

Ordered 3D plant (flower, tree and cluster) shaped array is another effective 3D structure for ETL. Compared with 1D array, it has more complex morphology and therefore exhibits a higher specific surface area, which further increases the light harvesting and promote the electron transport. In 2015, Wang's group reported a 3D treelike TiO$_2$ nanowire structured ETL, the length of TiO$_2$ trunk varies from 600 nm to 1.7 $\mu$m. The optimal 3D TiO$_2$ treelike structure produced a PCE of 9.0%, which was higher than the reported PSCs based on 1D TiO$_2$ nanorods or nanotubes at the time. After that,
Choi et al designed a 3D flower-shaped TiO₂ ETL by solvent-assisted molding (Figure 5A). The light harvesting efficiency increased by 5.2% over the light response range due to the reduced reflectance and enhanced absorption compared to planar reference device (Figure 5B). Moreover, the shunt resistance of 3D flower-shaped TiO₂-based device increased from 14 957.26 Ω to 33 540.58 Ω, meaning that better electron transport in 3D flower-shaped TiO₂-based device. Then, the PCE of the 3D flower-shaped TiO₂-based devices were boosted from 14.33% to 15.96%. In addition, Lee et al applied selectively grown TiO₂ nanorod cluster arrays as ETL. It provided direct electron transport paths and exhibited more efficient PL quenching. The shorter PL lifetime (52.8 nanoseconds) than that based on planar TiO₂ film (182 nanoseconds), indicating that the TiO₂ nanorod cluster array-based device have a stronger electron extraction capability and a lower charge recombination rate than the planar TiO₂ film-based device. Besides, it also provided a higher diffuse transmittance and exhibited an increased optical path through the active layer, thereby allowing higher light absorption of the active layer. Finally, the optimal device delivered an improved $J_{sc}$ of 23.13 mA cm⁻² and PCE of 19.86%. Moon et al introduced a similarly functioning 2D photonic crystal array structured ETL by using nanosphere lithography method, and boosted the PCE to 18.7%, which was 13% higher than the planar TiO₂ ETL.

Unlike the above structures, some other interesting ordered array structures (grating and helices, and so on) are also employed to prepare ETL to improve device performance. For example, Park et al fabricated helical TiO₂ arrays of different radius and pitch by an oblique-angle electron beam evaporation process (Figure 5C). Although the helical TiO₂ array had longer pathway compared to 1D vertical nanorod array, but it showed higher electron extraction efficiency due to the larger contact area between TiO₂ ETL and perovskite. Our group first designed a grating-patterned TiO₂ ETL to modulate the optical transmission and absorption in PSCs (Figure 5D). When irradiated by the same incident light, the absorption intensity of the grating TiO₂-based device was much higher than that of the planar TiO₂-based device (Figure 5E). The absorption enhancement mainly located closer to the TiO₂ scaffold, while only a standing wave was formed in planar TiO₂-based device (Figure 5F). Finally, an enhanced PCE of 18.6% was achieved for grating TiO₂-based device.

### 4 ORDERED ARRAY STRUCTURES FOR PEROVSKITE LAYER

Perovskite, as the light absorbing layer, is the most important part of PSCs. Its light collection ability, charge dynamic and defects are closely related to the performance of PSCs. To obtain more efficient performance, the researchers also tried to construct an ordered array structure in the perovskite layer. In 2016, Fan et al reported an inverted nanocone (i-cone) array structured perovskite film by a facile molding process for PSCs (Table 1). When illuminated under the light, the i-cone array structured device exhibited a lower reflectance than the planar-based device, which suggested a great enhancement in transmittance and was also confirmed by FDTD simulation. Thus, compared to the planar-based device, the $J_{sc}$ of i-cone-based device exhibited an improvement from 13.9 to 19.2 mA cm⁻², and the corresponding PCE increased from 6.3% to 10.2%. This result demonstrated the potential of perovskite arrays for designing high performance device. After that, Su and co-workers fabricated an ordered microporous perovskite via a polystyrene (PS) microsphere template (Figure 6A), and the average visible transmittance (AVT) can be tuned from 20% to 45% with different PS diameters. Almost all ordered microporous perovskite films had a smaller AVT than that of the normal perovskite film (40.6%), which mean that the optimal microporous perovskite layer had better light-harvesting ability. Besides, the time constants and $R_{rec}$ of the optimal pristine microporous perovskite was 225.58 nanoseconds and 172.2 Ω cm², which were larger than that of the pristine planar perovskite (110 nanoseconds, 76.89 Ω cm²) (Figure 6B,C). Longer lifetime and larger $R_{rec}$ proved that microporous perovskite had enhanced charge transport and reduced recombination. Eventually, compared to the reference device, the $J_{sc}$, $V_{oc}$, and PCE of the optimal device increased to 31%, 34%, and 109%, respectively. Unlike the general light harvesting or carrier transport effect, Dauskardt et al developed a special compound solar cell array with scaffold to address the perovskite stability issue (Figure 6D). Through optimizing scaffold size, the fracture energy of compound solar cell (~12.1 J m⁻²) was several orders of magnitude larger than that of planar device (~0.37 J m⁻²), indicating that the special array structured compound solar cell had much better mechanical stability and service lifetimes. Moreover, when exposed to the condition of 85°C and 85% relative humidity, the encapsulated array devices maintained 60% of initial PCE for 6 weeks, which was much better than the encapsulated planar devices. These results demonstrated that the design of array structured compound solar cell can effectively improve device stability.

In addition to the bulk perovskite arrays, constructing an ordered array at the perovskite surface via a template during the perovskite crystallization process is also a simple and efficient route to improve the PSCs performance. Compared to the bulk perovskite arrays, the array structures at the perovskite interface exert less damage to perovskite film, and still retains the advantages of the array structure including high light-harvesting ability and superior carries dynamic.
| Devices | $V_{oc}$ (V) | $J_{sc}$ (mA cm$^{-2}$) | FF (%) | PCE (%) | Improvement (%) | Reference |
|---------|-----------|-----------------|------|-------|----------------|---------|
| FTO/ATO nanorods array/TiO$_2$/MAPbI$_3$/Spiro/Ag | 1.1 | 23.81 | 76 | 20.13 | 13 | 21 |
| FTO/bi-TiO$_2$/perovskite/Spiro/Au | 0.95 | 19.2 | 64 | 11.7 | 108.9 | 14 |
| FTO/Y-SnO$_2$ nanosheets/MAPbI$_3$/Spiro/Au | 1.08 | 22.55 | 71 | 17.29 | 29.2 | 30 |
| Corrugated substrates/ITO/poly-TPD/PCBM/C$_{60}$/BCP/Ag | 1.08 | 19.7 | 63 | 13 | 49.4 | 32 |
| Etched FTO/TiO$_2$/MAPbI$_3$/Spiro/Ag | 1.06 | 23.81 | 71 | 17.85 | 20.3 | 33 |
| FTO/TiO$_2$ nanocones array/MAPbI$_3$/Spiro/Ag | 0.92 | 19.5 | 62 | 11 | 6.7 | 26 |
| FTO/ITO/Cds nanorods array/MAPbI$_3$/Spiro/MoO$_3$/Ag | 0.89 | 18.77 | 50 | 8.36 | 73.6 | 57 |
| FTO/ZnO nanorods array/TiO$_2$/Spiro/Au | 0.96 | 21.5 | 70 | 16.12 | 19.8 | 51 |
| FTO/SnO$_2$ nanorods array/MAPbI$_3$/P3HT/Au | 0.96 | 21.13 | 78 | 19.11 | 10.1 | 50 |
| FTO/c-TiO$_2$/TiO$_2$ nanocolumns array/MAPbI$_3$/Spiro/Au | 1.03 | 22.54 | 78 | 18.22 | 16 | 66 |
| Perovskite microcell (ITO/TiO$_2$/C$_{60}$/perovskite/PTAA/Ag) | 1 | 17.6 | 69 | 12.7 | - | 81 |
| FTO/TiO$_2$/patterned perovskite/Spiro/Au | 1.11 | 23.11 | 77 | 19.71 | 18.0 | 82 |
| FTO/TiO$_2$/patterned perovskite/Spiro/Au | 1.09 | 23.62 | 77 | 19.80 | 29.4 | 83 |
| FTO/C-TiO$_2$/M-TiO$_2$/columnar compressed perovskite/spiro/Au | 1.05 | 23.24 | 78 | 19.16 | 15.8 | 84 |
| Perovskite NP array based top cell (4T tandem with Si) | 1.18 | 19.8 | 79 | 18.5 | 46.8 | 85 |
| Perovskite top cell on a textured SHJ bottom cell (2T tandem with Si) | 1.788 | 19.5 | 73 | 25.52 | - | 86 |
| FTO/NiO nanowall-DEA/Perovskite/PCBM/Ag | 1.11 | 21.54 | 80 | 19.16 | 50.0 | 87 |
| ITO/NiO flakes-perovskite/PCBM-ZnO/Ag | 1.08 | 22.76 | 78 | 19.10 | 57.7 | 88 |
| ITO/NiO nanorods-NaPA/Perovskite/PCBM-BCP/Ag | 1.08 | 23.23 | 80 | 20.05 | 20.6 | 89 |
| ITO/porous PEDOT:PSS/MAPbI$_3$/PCBM/Bphen/Al | 1.07 | 22.14 | 74 | 16.5 | 7.6 | 90 |
| ITO/hexagonal prism-like CuSCN array/MAPbI$_3$/C$_{60}$/Bphen/Ag | 0.90 | 19.19 | 64 | 11.05 | - | 91 |
| inverted hemispherical architecture PDMS array/TiO$_2$/MAPbI$_3$/Spiro/Au | 1.1 | 23.29 | 74 | 19 | 17.2 | 92 |
| moth-eye Structured PDMS array/ITO/(FAPbI$_{3.97}$)(MAPbBr$_{0.03}$)/C$_{60}$/BCP/Cu | 1.05 | 25.11 | 79 | 20.93 | 21 | 93 |
| hierarchical pyramid PDMS array/TiO$_2$/MAPbI$_3$/Spiro/Ag | 1.033 | 20.87 | 65 | 14.01 | 6.8 | 15 |
| inverted cone PDMS array/ITO/ZnO/MAPbI$_3$/Spiro/Ag | 0.98 | 19.3 | 69 | 13.14 | 8.9 | 17 |
| inverse micro-cone PDMS array/ITO/SnO$_2$/MAPbI$_3$/Spiro/Ag | 1.09 | 22.97 | 61 | 14.83 | 8.9 | 94 |
For instance, through a facile imprinting process, Song’s group applied a diffracted-grating structure on perovskite layer for efficient PSCs. Commercial optical discs (CD/DVD) were used as initial masters, and the width and height of grating perovskite can be controlled by selecting different types discs (Figure 7A). The perovskite film with grating can utilize different order diffracting light to realize more efficient light collection, thus inducing a much higher light harvesting efficiency (over 90%) compared to planar structure perovskite (80%). The optical simulation about the per unit absorption also confirmed the effect of diffraction grating on optical enhancement (Figure 7B). Simultaneously, the carrier lifetime of perovskite film deposited on FTO substrate was 36.72 nanoseconds and 15.49 nanoseconds for planar and DVD-imprinted perovskite film, respectively. This result suggested that the perovskite with diffraction grating structure can extract photo-induced electrons and reduce electron-hole recombination more effectively. Benefiting from the better light-harvesting and carrier dynamic, the DVD-imprinted device showed a PCE of 19.71% with enhanced $V_{oc}$ (1.111 V), $J_{sc}$ (23.11 mA cm$^{-2}$), and FF (76.75%), which exhibited a 18% PCE increment compared to the planar perovskite device. Thereafter, Song’s group further constructed a micro-structure pattern mimicking whispering-gallery as light trapping structure for light harvesting at the perovskite interface by a similar imprinted process. Finally, the optimal device exhibited a PCE of 19.8%, which was 29.4% higher than that of planar perovskite device. In addition, Park et al also designed a pressure-induced perovskite porous structure on perovskite surface for PSCs. A polyurethane stamp was applied to press on the intermediate-state perovskite film and the pressure had great impact on the crystallization of perovskite film. As shown in 2D grazing-incidence wide-angle diffraction (GI-WAXD) pattern, the (110) plane diffraction peak of pristine perovskite film showed an isotropic intensity distribution along the azimuthal angle due to the random orientation of MAPbI$_3$ crystals (Figure 7C). While the anisotropic ring-pattern with an intensified region in the in-plane and out-of-plane directions was presented in porous perovskite (Figure 7D), which exhibited a preferred orientation of the crystal structure. The preferred oriented perovskite grain and high light absorption induced by the perovskite array provided better carrier dynamic. This was also confirmed by the time-resolved PL (TRPL), where the carrier lifetime extended from 28.2 nanoseconds (pristine planar perovskite) to 40 nanoseconds (pristine porous perovskite). As a result, compared to the planar device, the PCE of the optimal device boosted from 16.54% to 19.16% (Figure 7E).
Except for the single-junction PSCs, ordered perovskite arrays can also be applied in tandem solar cells. Zaccaria and co-workers replaced the traditional compact perovskite film by perovskite nanopillar for higher light trapping and better charge separation in four-terminal perovskite/c-Si tandem solar cells. When just focus on the PSC part, the conversion efficiency of nanopillar structured device was 46.8% higher than the planar structured device. This extraordinary enhancement was due to the higher $J_{sc}$ (15.3-19.8 mA cm$^{-2}$) and $V_{oc}$ (0.7-1.18 V), which were attributed to the stronger light coupling through waveguide-like modes and the faster carrier transport in the nanopillar structure. Finally, the optimal tandem solar cells obtained a PCE of 27%, which was 21% relatively higher than that from a planar perovskite tandem solar cell. Ballif et al designed an all-textured perovskite/c-Si tandem solar cell, the improved optical system induced by the ordered texture structure was also the main reason for its performance improvement. The reflections occurring in planar perovskite part induced a current loss of 3.14 mA cm$^{-2}$, while it only reduced by 1.64 mA cm$^{-2}$ in textured device. The reflection losses in the fully textured monolithic tandem were below 2%. Eventually, the optimal device obtained a high PCE of 25.2%.

**5 | ORDERED ARRAY STRUCTURES FOR HTL**

The p-type HTL is used to transport holes from the perovskite absorber layer and hinder holes backflow from the electrode. Unlike conventional structured (n-i-p, the ETL prepared on the substrate) PSCs, inverted structured (p-i-n, the HTL prepared on the substrate) PSCs provide the opportunity to design ordered array structured HTL layer. Among the p-type materials, nickel oxide (NiO$_x$) arrays (such as nanowalls, nanosheets, and nanoflakes) are commonly used due to their appropriate energy band, high hole mobility ($10^{-5}$-$10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$), good stability, and easy-to-control preparation process. For instance, Ko et al fabricated a 3D NiO nanowall by a hydrothermal process (Figure 8A). After optimizing the hydrothermal time, the NiO nanowall-based device exhibited a lower PL intensity, smaller $R_{ct}$, and higher $R_{rec}$ compared to control nanocrystals film-based device, suggesting that the NiO nanowall had better charge transport ability (Figure 8B,C). Besides, the hole trap density of NiO decreased from $7.48 \times 10^{15}$ cm$^{-3}$ (nanocrystals film) to $4.51 \times 10^{15}$ cm$^{-3}$.
(nanowell array), which directly indicated that the nanowell structured NiO was more effective to passivate the trap states density at the NiO/perovskite interface. Thus, an enhanced PCE of 19.16% was obtained with improved $V_{oc}$ of 1.109 V, $J_{sc}$ of 21.54 mA cm$^{-2}$, and FF of 80.23%. Hong et al fabricated a nanoporous NiO$_x$ (np-NiO$_x$) by a co-precipitation process. The optimized NiO$_x$ thin film exhibited an interconnected nanoporous network with numerous interconnecting nanowalls, and the corresponding device exhibited a high PCE of 19.10%. In addition, low temperature synthesized NiO$_x$ array architecture was also an effective structure for flexible devices, which can effectively reduce reflection loss and release stress/strain. Huang et al designed a NiO$_x$ nanopillar array (NaPA) on a flexible conductive substrate by a glancing angle deposition method (Figure 8D). During the deposition process, the electrode temperature was well-controlled at room temperature. As shown in UV-vis spectra, the NiO NaPA exhibited higher transmittance and lower diffuse reflectance than that of NiO compact layer (CL) (Figure 8E), which was attributed to the light confining induced by NaPA structure. Besides, the carrier lifetime of perovskite on NiO$_x$ NaPA (56 nanoseconds) was much smaller than that on NiO$_x$ CL (73 nanoseconds), meaning that the NaPA-based device had better charge-carriers dynamic. Eventually, the flexible PSCs based on NaPA delivered an enhanced PCE (from 14.64% to 17.23%) with an improved $J_{sc}$ (from 20.72 to 22.23 mA cm$^{-2}$) (Figure 8F). Moreover, the NaPA-based flexible devices retained over 80% of their initial PCE value after bending at a curvature radius of 10 mm for 500 times, while the CL-based flexible devices only retained 56% of their initial PCE (Figure 8G).
Except for the NiO\textsubscript{x} arrays, some other HTLs arrays also been researched for inverted structured PSCs. Zeng and co-workers fabricated porous PEDOT:PSS arrays by using a polystyrene (PS) nanosphere template.\textsuperscript{90} The porous Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate)dryre-dispersiblepellets\] (PEDOT:PSS) effectively enhanced the crystallization and morphology of perovskite film, which was confirmed by the SEM images and XRD pattern (Figure 8H). The large interface contact area between PEDOT:PSS and high quality perovskite induced more efficient carrier transport and lower trap-assisted recombination. Thus, the device based on porous PEDOT:PSS exhibited a higher EQE intensity compared to planar PEDOT:PSS among the wavelength range from 300 to 800 nm (Figure 8I). Finally, the optimal porous PEDOT:PSS-based device obtained a PCE of 17.32%, while the planar-PEDOT:PSS-based device only showed a PCE of 15.33%. Xu et al designed different CuSCN nanostructures (hexagonal prism-like, pyramid-like, and nanowire structure) for inverted structured PSCs.\textsuperscript{91} Those results showed that the quality of perovskite film had a strong bond with the thickness and morphology of nanostructured HTL. Among the three CuSCN nanostructures, perovskite film deposited on the 3D hexagonal prism-like structure exhibited a compact and pin-hole free morphology. After optimizing the thickness of 3D hexagonal prism-like structured CuSCN, the optimal device delivered a PCE of 11.40%.

6  |  ORDERED ARRAY STRUCTURES OUTSIDE THE SUBSTRATE

Except for the active layers in the PSCs, the ordered array structures can also be constructed outside the substrate. The ordered array structured films outside the substrate can be used as an antireflective layer, which can efficiently enhance the light harvesting in the devices by enhancing transmission and reducing the surface reflection. For example, Yu et al fabricated a hemispherical architecture (HSA) and an inverted hemisphere architectured (IHSA) polydimethylsiloxane (PDMS) transparent antireflective layer by a soft-imprint lithography patterning method (Figure 9A).\textsuperscript{92} After laminating the micro-grating structured PDMS layer on the bare glass surface, the optical transmittance/reflectance enhanced/reduced from 87.3%/8.8% (bare glass) to 88.8%/7.5% (HSA-PDMS) and 89.2%/6.4% (IHSA-PDMS) (Figure 9B,C). The difference was attributed to the different diffraction along the array direction of micro-grating structure, and the conclusion was also confirmed by the FDTD simulation. Eventually, compared to the reference device, the optimized device’s $J_{sc}$ increased from 19.91 to 23.29 mA cm$^{-2}$, and the corresponding PCE promoted from 16.2% to 19.0%. Choi et al designed a similar moth-eye structured PDMS film for efficient PSCs.\textsuperscript{93} By optimizing the PDMS film size, the 300 nm periodic nano-structured substrate displayed significantly enhanced light transmittance (from 92% to 96%). Finally, the $J_{sc}$ of the optimized device was 5.4% higher than that of the bare glass substrate based device, and the corresponding PCE reached $\sim$21%.

In addition, water repellent of the outer layer is also very important for PSCs, which can improve the stability of the device. However, simple nanostructured antireflective layer is not sufficient to achieve water repellent properties. An excellent water repellent layer requires a high static contact angle (CA) ($>150^\circ$) and a low roll-off angle (ROA) ($<10^\circ$) with long-term stability. In order to obtain the advantages of antireflection and water repellent at the same time, Park’s group fabricated a micro-pyramid structured PDMS film (Figure 9D).\textsuperscript{15} The pyramid structured PDMS film can maintain a high CA of $\sim$157$^\circ$ and a low ROA of $\sim$7$^\circ$ over 1 month under ambient conditions (Figure 9E), indicating that the pyramid structured PDMS film had a super hydrophobic feature. Meanwhile, the device’s PCE improved by 6.8% due to its anti-reflection effect. Fan et al designed a nanocone array structured PDMS film as an antireflective and water repellent layer for efficient PSCs (Figure 9F,G).\textsuperscript{17} The micro-cone array structure allowed light propagation surrounding the cone structure, which led to a higher antireflection, thereby reducing the light reflectivity of 8%. Moreover, it possessed a water CA of 155$^\circ$ and dust particles can be easily moved by rolling a water droplet across the surface (Figure 9H), which reflected the excellent self-cleaning performance of the micro-cone array structure. Based on the advantages, the optimized PSCs obtained an enhanced $J_{sc}$ (from 17.7 to 19.3 mA cm$^{-2}$) and PCE (from 12.06% to 13.1%), as well as an excellent self-cleaning property. Kang et al attached an inverse micro-cone array PDMS film outside the glass substrate.\textsuperscript{94} Through optimizing the textured master molds, the optimized device exhibited a $J_{sc}$ increment of 7.0 and a PCE increment of 8.9% with a good hydrophobicity.

7  |  SUMMARY AND OUTLOOK

This article has highlighted the efficient and stable PSCs based on ordered array structures and discussed the respective advantages of different ordered array structures. The proper ordered array structure is vital to light harvesting and
carriers transport in PSCs, which directly affects the efficiency and stability of PSCs. As shown in the article, the ordered array structures have the following advantages: ordered array structure improves the light scattering and reduces the light reflection, thereby enhancing light harvesting efficiency; ordered array can work as direct charge transport paths, which improves the transport properties; ordered array structured device can be greatly relaxed when bending, which suppresses the formation of crack in different layers of PSCs. Therefore, the PSCs with ordered array structures exhibit a high efficiency and outstanding stability. The highest efficiencies of ordered array structured single-junction PSCs and tandem solar cells reach 21% and 27%, respectively.

With a better understanding of ordered array structure, introducing the ordered array structures with more suitable gap, length, or size into PSCs can further balance carrier transport and light collection, and is expected to further improve the efficiency of perovskite devices. In addition, the current research on ordered arrays is mainly focused on small-area perovskite devices. It is relatively easy to design an array structure on a small-area substrate. How to design ordered arrays on large-area perovskite device in a cheaper and simpler way requires further research. Meanwhile, it should...
be noted that improving long-term stability of the device and reducing the material toxicity of perovskite is also the focus of future research on perovskite devices. More effective strategies need to be explored to reduce the Pb toxicity and isolate the oxygen and water in the work environment without compromising light harvesting, thereby ensuring the future commercialization and industrialization of PSCs. For example, combining ordered array structure with appropriate defect passivation layer, especially the passivation at the grain boundaries of perovskite film; forming stronger chemical bonds to trap Pb ions to ensure that Pb cannot escape easily. These areas have not been well studied, and we believe these are promising ways to further realize efficient and stable ordered array structured PSCs.

ACKNOWLEDGMENTS
We acknowledge the support from the National Natural Science Foundation of China (51772197), Key University Science Research Project of Jiangsu Province (17KJA430013), and the Priority Academic Program Development (PAPD) of Jiangsu Higher Education Institutions.

PEER REVIEW INFORMATION
Engineering Reports thanks Yaoguang Rong, John I. B. Wilson, and other anonymous reviewers for their contribution to the peer review of this work.

PEER REVIEW
The peer review history for this article is available at https://publons.com/publon/10.1002/eng2.12319.

CONFLICT OF INTEREST
The authors declare no potential conflict of interest.

DATA AVAILABILITY STATEMENT
Data sharing not applicable to this article as no datasets were generated or analysed during the current study.

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**How to cite this article:** Cao F, Wang M, Sun H, Tian W, Li L. Ordered array structures for efficient perovskite solar cells. *Engineering Reports*. 2020;2:e12319. [https://doi.org/10.1002/eng2.12319](https://doi.org/10.1002/eng2.12319)