

Colorful Efficient Moiré-Perovskite Solar Cells

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Light harvesting is crucial for thin-film solar cells. To substantially reduce optical loss in perovskite solar cells (PSCs), hierarchical light-trapping nanoarchitectures enable absorption enhancement to exceed the conventional upper limit and have great potential for achieving state-of-the art optoelectronic performances. However, it remains a great challenge to design and fabricate a superior hierarchical light-trapping nano-architecture, which exhibits extraordinary light-harvesting ability and simultaneously avoids deteriorating the electrical performance of PSCs. Herein, colorful efficient moiré-PSCs are designed and fabricated incorporating moiré interference structures by the imprinting method with the aid of a commercial DVD disc. It is experimentally and theoretically demonstrated that the light harvesting ability of the moiré interference structure can be well manipulated through changing the rotation angle (0°-90°). The boosted short-circuit current is credited to augment light diffraction channels, leading to elongated optical paths, and fold sunlight into the perovskite layer. Moreover, the imprinting process suppresses the trap sites and voids at the active-layer interfaces with eliminated hysteresis. The moiré-PSC with an optimized 30° rotation angle achieves the best enhancement of light harvesting (28.5% higher than the pristine), resulting in efficiencies over 20.17% (MAPbI₃) and 21.76% ((FAPbI₃)_{1-x} (MAPbBr₃)_x).

1. Introduction

Organic-inorganic hybrid perovskite solar cells (PSCs) have emerged as superior thin-film photovoltaic devices^[1a–If] due to their excellent optoelectronic properties including large absorption coefficients, long carrier diffusion lengths, and large grain sizes with less grain boundaries and defects,^[2a–2e]

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as well as low production costs compared to silicon solar cells. Recently, the power conversion efficiency (PCE) of PSCs has increased to a certificated efficiency of 25.5%.^[3] In parallel with the development of lighter, thinner and more efficient PSCs, many efforts have been made to reduce materials usage (especially less lead) and production cost for future mass production. Although perovskite is an excellent optoelectronic material, the PCE is still lower than its theoretical limiting value.^[4] Particularly, there are 35% light loss (Figure 1a) including 14% absorbed by the conductive substrates (FTO, ITO), 4% reflected by the glass substrate surface, 2% lost at the functional layers (electron transfer layer, hole transfer layer, and electrode layers), and 15% escaping from the PSCs.^[5] Effective photon management is expected to solve this issue and holds the promise to realize highefficiency thin PSCs at low cost. On the one hand, light-trapping nanostructures fold light into the absorber layer with

elongated optical paths and thus increase the photocarriers generation,^[6a,6b] resulting in improved photocurrent^[7a,7b] and PCE.^[8a–8c] On the other hand, the incorporated nanostructure usually induces voids, defects, and trapped states in the photo-voltaic devices, which deteriorates the electrical properties of the devices through photocarriers trapping, recombination, and shunting.

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Figure 1. Schematic of moiré-PSCs imprinted by a commercial DVD disc. a) Schematic of the PSCs with effective light absorption. A large portion of light is wasted in perovskite photovoltaic devices without efficient light-trapping structures. b) Schematic of the PSCs with dual diffraction gratings imprinted by a PDMS stamp and a commercial DVD disc. c) Principle of moiré-PSCs with different intersection angles for high light-harvesting efficiency. d) AFM image of the perovskite film with the D-PVK grating structure of $\approx 0.75 \,\mu$ m period, $\approx 0.25 \,\mu$ m width, and $\approx 0.1 \,\mu$ m height. e,f) Beam images of moiré perovskite film with dual diffraction gratings, diffraction observed at 532 nm (e: side view and f: front view). It has three sets of diffraction beams and diffraction spots.

Herein, we report a colorful moiré interference structured PSC (moiré-PSC) with simultaneously enhanced optical and electrical responses. The moiré interference structure is shaped by coupling two shallow diffraction grating structures, stacking the diffraction grated electron transport layer (D-TiO₂) and perovskite active layer (D-PVK) made by the micro-nano imprinting via a commercial DVD disc as a nano-grating stamp. Our design delivers three advantages: 1) In contrast to the high cost of a Si template fabricated by focused ion beam milling or electron beam lithography covering only small areas, and the high risk of damage during imprinting, the DVD template is trivially accessible and has low cost; 2) it boosts the light harvesting and thus the short-circuit current and PCE are enhanced; 3) the moiré interference structure allows for thinner semiconductor films, hence less lead in the device. The depth of the shallow grating in the PVK active layer is ≈100 nm, which ensures a small material loss.

The moiré interference structure shows extraordinary light management ability by increasing the high-order diffraction channels and naturally elongating the optical paths, and therefore it reduces the reflection loss, which demonstrates advantages over the single and independent light-trapping structures.^[9a-9c] Moreover, the imprinting process improves the crystallinity and interface morphology of the PVK so that defects and voids are reduced. We experimentally and theoretically investigate the optical and electrical responses of PSC devices after incorporating the moiré interference structure and the influence of rotation angle (from 0° to 90°) of the moiré structure on its light harvesting ability. The moiré-PSCs with 30° rotation angle exhibit the champion efficiency up to 20.17% for MAPbI₃ and 21.76% for (FAPbI₃)_{1-x}(MAPbBr₃)_x, respectively.

2. Results and Discussion

In nature, a moiré pattern, formed by the interference of two periodic structures with a twist and/or a difference in lattice constant, including repetitive gratings, dot arrays, or grids,^[10a–10c] is a fantastic phenomenon which has been widely applied in optical interferometry, image processing, artistic and textile design.^[11] We invent the moiré-PSCs by the micro-nano imprinting method using a commercial DVD as a nano-grating stamp, as depicted in Figure 1b (see the Section 4 for the details of the fabrication process). The moiré interference structure results from the stack of the diffraction grating based electron transport layer (D-TiO₂) and perovskite active layer (D-PVK). Through adjusting the rotation angle of the moiré interference structure (Figure 1c), the ability of light harvesting and trapping in the moiré-PSCs are



highly tunable. Meanwhile, the hierarchical nano-structure facilitates charge extraction and reduces surface recombination and shunt current due to the induced large surface area and suppressed defects and voids at the interface during the micro-nano imprinting process. The moiré perovskite film imprinted by optical DVD (as depicted in Figures S1 and S2, Supporting Information) gains the moiré interference structure of $\approx 0.75 \ \mu m$ period, $\approx 0.25 \ \mu m$ line width, and $\approx 0.1 \ \mu m$ height, which is confirmed by both AFM and scanning electron microscopy (SEM) images (Figure 1d and Figure S3, Supporting Information). The thicknesses of the pristine perovskite film and perovskite film (MAPbI3 and $(FAPbI_3)_x(MAPbBr_3)_{1-x}$ films) with moiré interference structure are clearly observed in the cross-sectional SEM images (Figures S3 and S4, Supporting Information). The X-ray diffraction peaks of the moiré-perovskite films become strong and sharp (Figure S5, Supporting Information). This demonstrates that the crystallinity of the moiré perovskite films is improved by the micro-nano imprinting method, which is strongly related to the open-circuit voltage (V_{oc}) of the PSCs.^[12]

In order to investigate the optical property of the moiréperovskite film for enhancing light harvesting, a laser beam was vertically incident upon the moiré-perovskite films, and the images of the diffracted laser beams are captured using a digital camera. The moiré-perovskite film has three sets of diffraction beams and diffraction spots, respectively (Figure 1e,f). According to the diffraction principle (see Supporting Information), the two sets of diffraction beams are attributed to the DVD diffraction grating because the corresponding first-order diffraction angle of the imprinted perovskite film is 46.7° at a wavelength of 532 nm (Figures S6 and S7, Supporting Information). However, another new set of diffraction light and spot is induced by the mutual coupling of the dual diffraction gratings in the moiré interference structure. Consequently, the diffraction image confirms that the moiré pattern has been successfully constructed by the stacked D-TiO₂ and D-PVK. Additionally, Figure S8, Supporting Information, shows that the light spots of the moiré patterns are located further away from the center of the spots as the rotation angles increasing from 0° to 90°, which indicates that the moiré diffraction pattern is highly dependent on or tunable with the rotation angle, causing the change of light harvesting efficiency (LHE).

To understand the formation of the moiré pattern, we employed a video camera to intuitively record the transformation of the moiré pattern on the substrate (see Video S1–S2). The width and period of moiré pattern have been modified significantly with the increasing rotation angle, and the maximum rotation angle of the moiré pattern is 90° due to its central symmetry. In brief, schematic diagram of the moiré pattern formation is shown:

$$W_{\rm m} = \frac{d_1 d_2}{\sqrt{d_1^2 + d_2^2 - d_1 d_2 \cos \theta^2}} \tag{1}$$

where $W_{\rm m}$ is the period of formed moiré patterns, d_1 and d_2 are the spaces of the different diffraction gratings, θ (θ + $n*90^{\circ}$, n = 0, 1, 2...) is the rotation angle between the diffraction

gratings. When the same period for both diffraction gratings is adopted $(d_1 = d_2 = d)$,

$$W_m = \frac{d}{2\sin\theta/2} \tag{2}$$

According to Equation (2), the change of the moiré period depends on the rotation angle, as shown in Figure 2a,b. Figure 2c exhibits the trend of moiré period versus rotation angles, which decreases sharply from 0° to 30° and changes mildly when the rotation angle goes beyond 30°. For PSCs, it is challenging to trap light in a wide wavelength range employing single period nano-architecture. Fortunately, the prepared moiré interference structure shows great ability to regulate the incident light over broad wavelengths. Figure 2d clearly shows that the short wavelength incident light (400 nm) through the perovskite moiré interference structure can be easily diffracted when the rotation angle is from 0° to 90° , while the long wavelength incident light (800 nm) can only be diffracted when the rotation angle is less than 55°. Consequently, broad incident light (400-800 nm) can be well modulated by the perovskite moiré interference structure with rotation angles ranging from 5° to 55° . This further indicates that the rotation angle greatly influences on the moiré diffraction and LHE.

To optimize the rotation angle for achieving the best LHE,^[13] we utilize FDTD simulations to calculate reflection (RI) (Figure S9, Supporting Information) and absorption (Figure S10, Supporting Information) coefficients as a function of rotation angles. Figure 2e shows that a moiré interference structure with an optimal rotation angle of 30° gains the strongest absorption. Furthermore, we measure the reflectance (R), absorbance (A) and LHE of the moiré-perovskite film (Figure 2f and Figure S11, Supporting Information) to prove the diffraction effect of the moiré interference structure on the light harvesting of the perovskite film. The moiré-perovskite film has both lower reflectance and lower transmittance (Figure S12, Supporting Information) than the pristine, thus achieves higher absorbance and LHE (especially for long wavelengths λ > 700 nm). Particularly, the number of diffraction channels of the moiré-perovskite film reaches its maximum value at the rotation angle of 30° and more sunlight is guided along the perovskite active layer via total internal reflections (Figure 2g and Figure S13, Supporting Information).

The $4n^2$ classical absorption limit of a solar cell could be surpassed by improving the anti-reflection coating and coupling more light into the moiré structure as guided waves. It is well known that a substantial fraction of photons cannot enter the solar cell due to the reflection at the top of the solar cell. In addition to the Yablonovitch $4n^2$ limit, an alternative practical $4n^2$ limit corrected for considering the reflection loss at the top surface is also calculated here.^[14] The $4n^2$ limit with the reflection loss $R(\lambda)$, that is, $4n^2(\lambda)(1-R(\lambda))$, describes the amount of light entering the absorber layer, and is significantly below the $4n^2$ classical absorption limit (Figure 2h). The external quantum efficiency (EQE) values of the moiré PSC within the whole visible light range are close to the practical $4n^2$ limit with the reflection loss. Particularly, the absorption of the moiré-PSC ranging from 400 nm to 500 nm has exceeded the average value of the practical $4n^2$ limit, suggesting that the moiré structure









Figure 2. Optical characterization of moiré perovskite film. a) Image of moiré pattern caused by the interference of two periodic structures. b) Preparation of moiré PSCs by rotating the two diffraction gratings from 0° to 90°. The moiré period depends on the rotation angle between the two gratings. c) Phase diagram of moiré periods versus rotation angles (from 0° to 90°) and schematic diagram of the moiré pattern formation (inset (c)). The moiré period decreases with the increment of the rotation angle. d) Diagram of rotation angles versus incident wavelengths for moiré diffraction. The rotation angles from 5° to 55° ensure that the moiré light diffraction covers broadband incident wavelengths (400–800 nm). e) Calculated optical absorption versus rotation angles from 0° to 70° under two incident wavelengths (500 and 700 nm). f) Measured reflection and LHE of moiré perovskite film. The moiré perovskite film improves light harvesting efficiency and enhances light trapping at the range of long wavelengths (λ > 700 nm). g) Transmitted optical power (dB) relative to unpolarized incident beam (532 nm) into moiré-PSCs for 30° rotation angle. h) EQE, Yablonovitch 4n² limit, and 4n² limit considering the reflection loss of the PSCs. EQE of moiré-PSCs has been enhanced at the range of long wavelengths (λ > 700 nm) due to the improved LHE of the coupling effect of moiré diffraction gratings.

excites the guided waves and elongates the optical paths. The corresponding integrated short-circuit current density (J_{sc}) of the PSCs from the EQE spectrum is shown in Figure S14, Supporting Information. Also, the EQE of the moiré-PSC has been enhanced within the range of long wavelengths ($\lambda > 700$ nm) where the perovskite material absorption is weak.

Furthermore, we utilize the FDTD method to simulate spatial profile of the optical absorption inside the PSCs with the device configurations of glass/FTO/compact-TiO2/meso-TiO₂/perovskite (pristine and moiré interference structure)/ Spiro-OMeTAD. Figure 3a-d illustrates the spatial profile of the time averaged electromagnetic energy density with respect to the xz-plane at the wavelengths of 500, 600, 700, and 750 nm for the pristine, D-TiO₂ PSC, D-perovskite PSC, and the moiré-PSC, respectively. It is clearly shown that the moiré-PSC has strong absorption at the grating troughs and peaks, while absorption of the pristine one is concentrated close to the TiO₂ layer forming a fringe pattern due to the constructive and destructive interferences between the forward and backward propagating waves. Regarding the pristine PSC, longwavelength light (>700 nm) will propagate through the perovskite layer and cannot contribute to the short-circuit current. For the moiré-PSC, differently, most of the light is effectively trapped within the perovskite layer due to the increased diffraction channels and elongated optical paths. Besides, the electromagnetic energy density distribution for the different planes is shown in Figure S15a,b, Supporting Information. To visualize the influence of the grating rotation on the field distribution, we present the difference between the electromagnetic energy distribution between a 30° moiré-PSC and a 20° moiré-PSC in the xz-plane for a wavelength of 700 nm, as shown in Figure S15 c, Supporting Information. An enhancement of the energy density inside the absorbing PVK troughs is observed for the 30° moiré-PSC.

On the other hand, for the electrical properties, the prepared moiré interference structure helps the extraction of the photoinduced electrons in the perovskite film, which is demonstrated by the photoluminescence characterization as depicted in Figures S11 and S16, Supporting Information. In Figure S11, Supporting Information, the steady-state PL intensity (at \approx 780 nm) of the moiré-perovskite film on TiO₂-FTO glass is lower than those of pristine PVK, D-TiO₂+F-PVK, and F-TiO₂+D-PVK, which suggests that the moiré interference structure facilitates the charge-carrier extraction. Moreover, the time-resolved photoluminescence decay of perovskite films on fluorine-doped tin-oxide (FTO) glass is measured (Figure S16, **ADVANCED** SCIENCE NEWS

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Figure 3. 3D FDTD simulations for light absorption of PSCs incorporating different light-trapping structures. The grating period is 750 nm. The spatial profile of the electromagnetic energy density with respect to xz- plane at the wavelengths of 500, 600, 700, and 750 nm: a) The pristine, b) the D-TiO₂, c) the D-perovskite, d) the moiré-perovskite films. The detailed parameters of the moiré-perovskite films include 25 nm compact-TiO₂ layer on FTO, 180 nm meso-TiO₂ incl. 80 nm corrugation at the meso-TiO₂/PVK interface, 330 nm PVK incl. 80 nm corrugation at the meso-TiO₂/PVK interface and 100 nm corrugation at the PVK/Spiro-OMeTAD interface, 300 nm Spiro-OMeTAD including the 100 nm corrugation at PVK/Spiro interface. The moiré-perovskite films achieve the strongest absorption caused by the coupling effect of moiré diffraction grating compared with other samples (pristine, D-TiO₂, D-perovskite films).

Supporting Information). We fit the data with a global bi-exponential model and the detailed fitted PL lifetimes^[15a-15c] (τ_1 , τ_2 , and τ) are collected in Table S1, Supporting Information.

$$Y = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) + \gamma_0$$
(3)

where A_1 and A_2 are the relative amplitudes and τ_1 and τ_2 are the lifetimes for the fast and slow recombination.^[16] The average lifetimes of D-TiO₂+F-PVK, F-TiO₂+D-PVK, and moiré-PVK are 60.13, 52.58, and 24.69 ns, respectively, which are much lower than that of the pristine (84.47 ns). The PL quenching rate is expected to originate from the charge-carrier extraction across the interface. The moiré-PVK film exhibits the lowest average lifetime due to the well crystallinity and PL coupling to the resonance mode of the optical cavity. Consequently, the hierarchical

moiré interference structure shows much better ability to enhance PL quenching than the single grating structure.

In order to investigate the influence of the moiré interference structure on the electrical responses of the PSCs, the moiré-PSCs are constructed based on a typical PSC configuration consisting of a FTO substrate/compact TiO₂/meso-TiO₂/ perovskite/Spiro-OMeTAD/Au. Due to the optical interference, the imprinted moiré-PSCs show a colorful visual effect (**Figure 4**a). The current density-voltage (*J*–*V*) characteristics of the moiré-PSCs under AM 1.5G illumination with the light intensity of 100 mW cm⁻² are measured, as presented in Figure 4b. The corresponding photovoltaic parameters, including short-circuit current density (*J*_{sc}), open-circuit voltage (*V*_{oc}), fill factor (FF), and PCE are listed in Table S2 and Figure S17, Supporting Information. For MAPbI₃, the champion





Figure 4. Photoelectric performances of the moiré-perovskite solar cells. a) The visual effect of colorful PSCs. b) J-V characteristics of moiré-PSCs with MAPbI₃ and (FAPbI₃)_{1-x}(MAPbBr₃)_x perovskite active layers. For MAPbI₃, the PCE of moiré-PSCs (20.17%) is higher than that of the pristine (17.42%). For (FAPbI₃)_{1-x}(MAPbBr₃)_x the PCE of moiré-PSCs (21.76%) is higher than that of the pristine (19.11%). c) J-V hysteresis of the PSCs. The moiré-PSCs show a negligible photocurrent hysteresis. d,e) J-V characteristics of PSCs with MAPbI₃ for experimental measurement and theoretical fitting. The m_i recombination coefficient has been obtained. f) V_{oc} versus light intensity for the PSCs. g,h) J-V hysteresis measurement of PSCs under the 480 and 650 illumination with the light intensity of 100 mW cm⁻². i) Current density at maximal output power point and corresponding PCE.

PSCs (moiré-PSCs) exhibits I_{sc} of 23.74 mA cm⁻², V_{oc} of 1.104 V, FF of 76.95%, and PCE of 20.17%, which is much better than those of the pristine (PCE: 17.42%, J_{sc} : 21.77 mA cm⁻², V_{oc} : 1.086 V, FF: 73.65%). The PCE of moiré-PSCs (20.17%) is higher than that of the pristine (17.42%), PSCs with D-TiO₂ (18.61%), and PSCs with D-PVK (19.34%). Among all photovoltaic parameters, the I_{sc} also shows a linear improvement, increasing from 21.77 mA cm^{-2} , 22.66 mA cm^{-2} , 23.15 mA cm^{-2} to 23.74 mA cm^{-2} . The current density can be remarkably improved after introducing the moiré interference structure by accelerating electron collection, suppressing defects and voids^[17a,17b] and efficiently enhancing light harvesting. In order to further demonstrate the effect of the moiré interference structure on another perovskite system, $(FAPbI_3)_{1-x}(MAPbBr_3)_x$ has been employed. In Figure 4b, the moiré interference structure as a spatial light modulator also shows the excellent light harvesting ability, resulting in a higher PCE (21.76%) than that of the pristine (19.11%). The improved V_{oc} of the moiré-PSC results from the imprinting treated perovskite films due to the photon population with the focusing effect of the moiré interference structure, fewer surface defects induced recombination^[18a,18b] and lower voids and pinholes. In Figure S18, Supporting Information, *I*-V and EQE curves of the moiré-PSCs with different rotation angles are characterized, and the effect of moiré interference by rotation angles (pristine, 10°, 20°, 30°, 40°, 60°, 80°) on the efficiency of moiré PSCs (17.42%, 19.31%, 19.59%, 20.17%, 19.23%, 18.96%, 18.41%) are revealed. The nearly best PCE of the moiré-PSC at 30° rotation angle agrees with the optical simulation results (Figure 2e,g). Subsequently, we measure the I-V curves of the plane and moiré PSCs with the same perovskite crystallinity (Figure S19, Supporting Information). In Figure S20, Supporting Information, the plane PSC after the imprinting process exhibits J_{sc} of 21.90 mA cm⁻², V_{oc} of 1.095 V, FF of 76.43%, and PCE of 18.32%, which is higher than those of the pristine. It is concluded that the imprinting process improves the electrical properties of PSCs due to efficiently decreasing defects and voids, as well as enhancing crystallinity in the perovskite active layer.^[6a,18a] The effect of the moiré interference structure on the optical and electrical properties of PSCs is summarized in Figures S21 and S22, Supporting Information. Compared to the pristine one, the moiré-PSC achieves the enhancement of LHE (28.5%), photocurrent (9.1%), and

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efficiency (15.8%). Photovoltaic parameters of moiré-PSCs with 20 cells in total are measured, as shown in Tables S3 and S4, Supporting Information. The statistic PCE histogram of the moiré-PSCs along with the Gaussian fitting is illustrated in Figure S23, Supporting Information, which demonstrates that the high-efficient moiré-PSCs have well reproducibility.

Interestingly, the optimized moiré-PSC exhibits a negligible photocurrent hysteresis (Figure 4c) and stable $V_{\rm oc}$. In Figure 4d,e, and Figure S24, Supporting Information, I-V curves of MAPbI₃ PSCs are analyzed by the modified detailed balance model.^[19] For reverse and forward scanning modes, the recombination coefficients m_i of moiré-PSCs is $2.25 \times 10^{12} \text{ s}^{-1} \text{ cm}^{-3}$ and $2.86 \times 10^{12} \text{ s}^{-1} \text{ cm}^{-3}$, respectively. In comparisons to the moiré-PSCs, the pristine devices have higher recombination coefficients (2.83 \times $10^{12}~s^{-1}~cm^{-3}$ and $5.69 \times 10^{12} \text{ s}^{-1} \text{ cm}^{-3}$). Also, the reduced voids and pinholes by the imprinting fabricated moiré-PSCs can be confirmed by the increased shunt resistances. Then, we test the device response under different light intensities (Figure S25, Supporting Information). Both the pristine and moiré-PSCs show a linear I_{sc} versus light intensity relations (Figure S26, Supporting Information). The relation between J_{sc} and P is described by the formula of $J_{sc} \propto PS$, S of 0.966 is observed for the pristine PSC and S of 0.988 is observed for the moiré PSC, respectively. As S is closer to 1, it indicates that all free carriers are swept out and collected at the electrodes with ignorable recombination and accumulation. The relation between the V_{oc} and light intensity is also plotted (Figure 4f). The pristine shows a slope of 1.81 $k_{\rm B}T/q$, while the moiré-PSC shows a much smaller slope (1.54 $k_{\rm B}T/q$), where $k_{\rm B}$ is the Boltzmann constant, T is the temperature, and q is the electric charge. It is known that the deviation of the slope from $k_{\rm B}T/q$ implies a defect-assisted recombination in the devices.^[20] The moiré-PSC with a smaller slope demonstrates fewer trap sites in the film, which results in the smaller nonradiative recombination loss and a higher Voc. In order to clarify the physical origin for the eliminated hysteresis and improved $V_{\rm oc}$ by the imprinting process, the J-Vhysteresis of PSCs under monochromatic illumination at 480 and 650 nm with a light intensity of 100 mW cm⁻² is measured (Figure 4g,h). For the pristine PSC under the 480 nm illumination, a pronouncedly large Voc difference between the forward and reverse scanning modes and giant Jsc loss undoubtedly confirm the dominated surface recombination at the PVK-TiO₂ interface where light absorption is strong (see Figure 3a). The surface recombination has been significantly reduced during the imprinting process.

We also employ electro-chemical impedance spectroscopy (EIS) to investigate the mechanism of carrier recombination in PSCs. The Nyquist plots for PSCs are measured in the dark at a bias voltage of 0.8 V, as shown in Figure S27, Supporting Information. The large semicircle is attributed to the recombination resistance ($R_{\rm rec}$) in literature reports,^[21a,21b] and a higher $R_{\rm rec}$ is associated with a lower carrier recombination. The moiré-PSCs achieves higher $R_{\rm rec}$ (82.1 k Ω) and lower carrier recombination than other samples (PSC with D-PVK: 75.7 k Ω , PSC with D-TiO₂: 70.4 k Ω , the pristine: 63.1 k Ω). The results indicate that the moiré pattern can improve charge collection with reduced occurrences of trap-assisted recombination at the interface. Ultimately, the stabilized power output at the maximum

power point (MPP) for the best performing devices (Figure 4i) has been extracted from the J–V curves, which confirm the reliability of the moiré-PSC parameters. A stabilized output efficiency of 19.68% is obtained at the MPP.

3. Conclusion

We construct the moiré interference structure as a spatial light modulator in PSCs by imprinting diffraction grating structures on both the electron transport layer and perovskite active layer. Most importantly, we propose the dual grating formed moiré interference structure to flexibly manipulate light harvesting through changing the rotation angle from 0° to 90°. Compared to the single and independent optical structures, the moiré interference structure offers more diffraction channels leading to increased optical path lengths and enhanced light absorption at the perovskite active layer. Accordingly, two moiré-PSCs with the champion efficiencies of 20.17% for MAPbI₃ and 21.76% for $(FAPbI_3)_{1,x}(MAPbBr_3)_x$ are achieved. The results suggest that the moiré interference structure is a promising light management strategy for fabricating high-performance photovoltaic devices and photodetectors without degrading their electrical properties, especially for the potential application of perovskite/silicon tandem PSCs. The imprinting method based on a commercial DVD template makes the fabrication facile, lowcost, and scalable for various photoelectric conversion devices. Our approach is practical and easy to be generalized for the fabrication of high-efficiency, low-cost, and large-area PSCs, which opens new avenues for next-generation highly-efficient photovoltaics.

4. Experimental Section

Materials: TiO₂ paste (30NR-D) was purchased from Dyesol. Methylammonium iodide, methylammonium bromide, PbI₂ (99.999%), PbBr₂ (99.999%), CsI (99.999%), *N*,*N*-dimethylformamide (99.8%, anhydrous), dimethyl sulfoxide (99.8%, anhydrous), and Spiro-OMeTAD were purchased from Xi'an Polymer Light Technology Corp. 4-*tert*-butyl pyridine (Sigma-Aldrich), γ butyrolactone, and acetonitrile were purchased from Sigma-Aldrich. Li-TFSI was purchased from TCI Tokyo Chemical Industry.

Preparation of Grating-Patterned PDMS Film by DVD Disc: The DVD optical discs were soaked in ethanol and cleaned by ultra-sonication in ethanol for 15 min until the films are removed and rinsed with ethanol. The masters were kept in an oven at 75 °C for 1 h. After drying, a commercial PDMS solution was cast on the disk layer, followed by a degassing and curing process at 70 °C for 4 h in a vacuum oven. Then, the grating-patterned PDMS film was peeled off of the DVD disk.

Perovskite Solar Cells Fabrication: FTO (fluorine-doped tin oxide)coated glass (NSG, ≈15Ω) were washed by sonication with deionized water, ethanol, and acetone, and treated by O₃/ultraviolet for 15 min. To obtain a compact TiO₂ layer onto the FTO, TiCl₄ (99.9%) solution was diluted to precursor solutions with 0.2 м. Then, the treated FTO substrates were immersed into the TiCl₄ precursor solution and kept in an oven at 75 °C for 1 h. Then all substrates were washed with deionized water and ethanol, and dried at 100 °C in air for 1 h. To form a TiO₂ mesoporous scaffold with a grating pattern, 50 µL diluted TiO₂ nanoparticle solution (Dyesol, 30NR-D, 0.135 g mL⁻¹ in ethanol) was spin-coated at 3000 rpm onto a FTO substrate with a compact TiO₂ layer, and the as-prepared PDMS film was attached on the deposited film. After baking at 100 °C for 5 min, the PDMS film was removed,



leaving the imprinted pattern on the TiO₂ layer. For MAPbI₃, 30 μ L 1.3 m CH₃NH₃I and PbI₂ (molar ratio = 1:1) were mixed into dimethyl sulfoxide and γ butyrolactone (volume ratio = 3:7). The CH₃NH₃PbI₃ precursor solution was deposited onto FTO/TiO₂ substrates by spin-coating process at 4000 rpm for 30 s. Toluene (120 μ L) was dropped onto the substrate toward the end of spin coating. For (FAPbI₃)_{1x}(MAPbBr₃)_{xr}, the perovskite film was made by the two-step method, as described elsewhere.^[22] The imprint process with a DVD as a template was finished under 50 °C (I) and 100 °C (II) for 5 and 30 min. 35 μ L of Spiro-OMeTAD solution was spin-coated on the layer at 3000 rpm for 30 s. The Spiro-OMeTAD in 1 mL chlorobenzene, 28.8 μ L of 4-*tert*-butyl pyridine (Sigma-Aldrich), and a solution of 520 mg of Li-TFSI (TCI) in 1 mL acetonitrile (Sigma-Aldrich). Finally, an 80 nm Au layer was deposited on Spiro-OMeTAD layer by thermal evaporation.

Characterization: The morphologies of the perovskite films were evaluated by the field-emission SEM (JEOL, JSM-7500F, Japan). Absorption spectra were obtained with an UV-vis spectrophotometer (Perkin Elmer Lambda 750). Current density-voltage characteristics of the PSCs under 1 sun illumination were measured in ambient conditions using a programmable Keithley 2400 source meter under AM 1.5G solar irradiation at 100 mW cm² (Enli Tech, Class AAA solar simulator). EIS characteristics of the cells were measured by an electrochemical analyzer (CHI 660D, Chenhua Instruments Co., Shanghai) in the dark at room temperature. The EQE was measured by illumination with monochromatic light (Enli Tech, Class AAA solar simulator, QE-mini).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that supports the findings of this study are available in the supplementary material of this article.

Keywords

DVD disc, light diffraction, micro-nano imprinting, moiré interference structure, perovskite solar cells

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