Enhancing the Efficiency of Perovskite Solar Cells through Interface Engineering with MoS$_2$ Quantum Dots

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Abstract: The interface of perovskite solar cells (PSCs) determines their power conversion efficiency (PCE). Here, the buried bottom surface of a perovskite film is efficiently passivated by using MoS$_2$ quantum dots. The perovskite films prepared on top of MoS$_2$-assisted substrates show enhanced crystallinity, as evidenced by improved photoluminescence and a prolonged emission lifetime. MoS$_2$ quantum dots with a large bandgap of 2.68 eV not only facilitate hole collection but also prevent the photogenerated electrons from flowing to the hole transport layer. Overall promotion leads to decreased trap density and an enhanced built-in electric field, thus increasing the device PCE from 17.87% to 19.95%.

Keywords: perovskite; solar cell; MoS$_2$; quantum dots; interface

1. Introduction

Perovskite solar cells (PSCs) have been widely explored in recent years due to their low cost and high efficiency [1]. Notably, inverted-structure devices have shown negligible hysteresis and high stability due to the employment of undoped hole transport materials [2]. However, their power conversion efficiency (PCE) still lags that of normal structure devices, mainly due to the energy and carrier losses at the interface between the hole transport layer (HTL) and the perovskite active layer [3]. Although replacing the typical poly(3,4-ethylenedioxythiophene) (PEDOT:PSS) with poly[bis (4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA) can significantly suppress interface losses [4], there is still a large number of voids and defects at the buried bottom surface of the perovskite layer. This is caused by the hydrophobicity of PTAA film, which serves as a substrate affecting the growth of perovskites. Degradation is more likely to happen at trap centers than inside the crystal under illumination, an electric field, and humidity conditions. Thus, buried interfacial voids and defects will serve as decomposition centers that accelerate device efficiency decline [5].

The interface between the HTL and the perovskite layer determines the hole transfer rate and hole collection efficiency [6], which can be significantly improved by enhancing the coupling between layers [7]. Moreover, interface engineering with various materials, including polymers [8–10], large organic cations [11,12], fullerene derivatives [13,14], carbon dots [15,16], and quantum dots (QDs) [17], can also block the undesired charge carrier flow, passivate surface traps, and improve the interface contact.

Molybdenum disulfide (MoS$_2$), with high charge carrier mobilities and a tunable energy band structure, is a promising low dimensional material used in optoelectronic applications. The lattice of MoS$_2$ and MAPbI$_3$ (MA, methylamine) perovskites match well with each other, so they can form high-quality heterojunctions via epitaxy growth [18].
being beneficial for reducing non-radiative recombination. MoS\textsubscript{2} has been used mainly as a hole-transporting layer or dopant in these layers \cite{19,20}. However, it is difficult to adjust the chemical properties of most MoS\textsubscript{2} used in photoelectric devices \cite{21}. Meanwhile, stacking 2D MoS\textsubscript{2} reduces interface contact and easily causes a void at the interface.

Rich chemistry and size effects make QDs unique 0D materials with huge possibilities to tune their electronic properties and enhance device interface contact. Leyla et al. utilized MoS\textsubscript{2} QDs with reduce graphene oxide (RGO) hybrids as hole transport layers (HTLs) and active buffer layers. The final PCE of the normal structure PSCs with MoS\textsubscript{2} quantum dot/graphene hybrids reached 20.12\% \cite{22}. Here, we engineer the interface between perovskite and PTAA using MoS\textsubscript{2} QDs, which passivates the buried defects, facilitates interface contact, promotes the perovskite crystal quality, prevents reverse carrier flow, and enhances hole collection. The MoS\textsubscript{2} QDs are capped with trioctyl-phosphine oxide (TOPO).

With optimized MoS\textsubscript{2} QD layers, the PCE of perovskite solar cells increases from 17.87\% to 19.95\%, demonstrating that MoS\textsubscript{2} QDs are indeed promising materials for passivating the buried defects of solution-processed perovskite optoelectronic devices.

2. Materials and Methods

Lead iodide (PbI\textsubscript{2}), lead bromide (PbBr\textsubscript{2}), formamidinium iodide (FAI), methylamium bromide (MABr), cesium iodide (CsI), and poly[bis(4-phenyl)(2,4,6-triMethylphenyl)aMine] (PTAA) were purchased from Xi’an Polymer Light Technology Corp (China). Bathocuproine (BCP) and (6,6)-phenyl C\textsubscript{61}-butyric acid methyl ester (PCBM) were purchased from Luminescence Technology Corp (Taiwan, China). N, N-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), tri-n-octylphosphine (TOP), N-methylpyrrolidone (NMP) and sulfur were purchased from Sigma-Aldrich (Sigma-Aldrich, St. Louis, MO, USA). Ammonium molybdate tetrahydrate and cysteine were purchased from Sinopharm Chemical Reagent limited corporation (Beijing, China). Chlorobenzene (CB) was purchased from J&K Scientific Corp (Beijing, China). Silver was purchased from commercial sources with high purity.

The MoS\textsubscript{2} QDs were synthesized using the hydrothermal method. Ammonium molybdate tetrahydrate (80.4 mg) and cysteine (20 mg) were dissolved in 10 mL NMP, heated, and stirred at 70 ℃ for 20 min. Sulfur (19.0 mg) was added into 2.5 mL TOP in a glove box before being stirred and heated at 70 ℃ until it was fully dissolved. Then, a 333 µL sulfur solution was added to the ammonium molybdate tetrahydrate NMP solution. Stirring and heating continued through the addition of 30 µL HCl (0.1 M) until the solution was transparent. The mixture was transferred to a Teflon reactor and heated continuously at 240 ℃ for 24 h. After cooling down to room temperature in the reactor, the obtained solution was poured into centrifuge tubes for centrifugation. The first-time centrifugation precipitate containing sulfur and bulk MoS\textsubscript{2} was abandoned. The solution was then mixed with ethanol and centrifuged at 12,000 rpm for 20 min. The target product was obtained and dispersed in DMF for device fabrication.

Perovskite precursor was prepared by dissolving 506.9 mg PbI\textsubscript{2}, 80.7 mg PbBr\textsubscript{2}, 172.2 mg FAI, and 22.4 mg MABr in 1 mL of 4:1, v/v proportion dimethylformamide (DMF) and dimethyl sulfoxide (DMSO) mixture. The solution was heated at 70 ℃ until it became pellucid. Next, 42.4 µL as-prepared CsI (1.5M) DMSO solution with a concentration of 1.5 M was added to the as-mixed solution, which was left on a hotplate at 70 ℃ for 30 min. At this point, the precursor solution was ready for perovskite film preparation.

ITO substrates were cleaned using isopropyl alcohol, acetone, chloroform, acetone, and isopropyl alcohol in sequence, and all were treated with ultrasound for 10 min. Then, the ITO substrates were blow-dried with a dry nitrogen flow and treated with a vacuum plasma cleaner for 15 min. The PTAA (1.5 mg mL\textsuperscript{-1}, in toluene) solution was spin-coated onto the pre-cleaned ITO substrates at 4000 rpm for 30s and heated at 100 ℃ for 10 min. After cooling down, 30 µL MoS\textsubscript{2} QDs in different concentrations were spin-coated on PTAA layers at 4000 rpm for 8 s. Immediately, 70 µL perovskite precursor solution was dropped on the prewetted substrates. A two-stage spin-coating procedure was used for perovskite coating, 1000 rpm for 10 s and 6000 rpm for 20 s. During the second spin-coating
step at 6000 rpm, 120 µL chlorobenzene was quickly dropped to the center of the wet film. After annealing at 100 °C for 10 min and cooling down to room temperature, PCBM (20 mg/mL) solution was dropped on the perovskite and spin-coated at 1500 rpm for 30 s and annealed at 100 °C for 10 min. Then, a 40 µL BCP (0.5 mg/mL in isopropanol) solution was dynamically dropped on the surface of the PCBM layer at a speed of 4000 rpm for 40 s. Finally, an Ag electrode was fabricated on the BCP layer using thermal evaporation depositing. The active area could be confirmed by the ITO and Ag electrode overlapping region of 0.047 cm$^{-2}$.

X-ray diffraction (XRD) patterns were measured with a D8 Advance-Bruker AXS X system (Bruker AXS, Karlsruhe, Germany). Transmission electron microscopy (TEM) images were taken with a JEM 2100F TEM (Japan Electronics Co., Ltd, Japan). X-ray photoelectron spectroscopy (XPS) spectra were characterized with ESCALAB 250 (Thermo Fisher Scientific, Waltham, MA, USA). A previously reported method was referred to for the characterization of the interface [6]. Scanning electron microscope (SEM) images were taken using a Hitachi SU8000 SEM (Hitachi Limited, Tokyo, Japan). Fourier-transform infrared spectroscopy (FTIR) was conducted using a Nicolet IS50 FT-IR (Thermo Fisher Scientific, Waltham, MA, USA). Current density–voltage (J-V) and space-charge-limited current (SCLC) curves were measured using a Keithley 2400 Source Meter (Tektronix, Beaverton, OR, USA). All the current density-voltage measurements were conducted using a solar simulator (Class AAA solar simulator, Newport Technologies, Inc., Irvine, CA, USA) External quantum efficiency (EQE) curves were measured using a Crowntech QTest Station 1000 AD (Crowntech, INC., Indianapolis, IN, USA). Capacitance–voltage (C-V) curves were tested at a constant frequency of 100 kHz. Electrochemical impedance spectroscopy (EIS) was measured using an Admiral electrochemical workstation (Admiral Instruments, Tempe, AZ, USA) from 10 Hz to 1000 kHz frequency with 0.89 V bias voltage in dark conditions.

3. Results and Discussion

3.1. Characterization of MoS$_2$ QDs

The MoS$_2$ QDs were prepared hydrothermally, wherein the deionized water was replaced by NMP. Due to the strong stripping ability of NMP, the formation of bulk MoS$_2$ can be suppressed during the synthesis [23]. Tri-n-octylphosphine (TOP) was oxidized to Tri-n-octylphosphine oxide, capping the MoS$_2$ QDs and further restricting the size of the quantum dots.

A transmission electron microscopy (TEM) image is given as Figure 1a, showing the morphology of MoS$_2$ QDs. The uniformly dispersed MoS$_2$ QDs indicates their excellent polydispersity in DMF. The high-resolution TEM image (Figure 1b) shows the interplanar spacing of 2.26 Å of a near-spherical particle, which is assigned to the (103) plane of MoS$_2$. The X-ray photoelectron spectroscopy (XPS) spectra were characterized to determine the chemical states and the element composition of the as-prepared MoS$_2$ QDs. The peaks of Mo, S, P, and C are clearly observable in the survey spectrum (Figure 1c). Figure 1d shows characteristic peaks at 162.05 eV and 160.9 eV, which are relevant to S 2$p_{1/2}$ and S 2$p_{3/2}$ orbitals, respectively. Two strong peaks located at 228.05 eV and 231.35 eV in Figure 1e are assigned to Mo 3$d_{5/2}$ and Mo 3$d_{3/2}$ orbitals, respectively. The higher valance resulted from the partial oxidation of MoS$_2$ QD with the remaining oxygen in the Teflon-lined stainless-steel autoclave during the reaction [24,25]. Moreover, the atomic ratio of S to Mo is calculated as ~1.89. In addition, the P element from TOP can also be detected from the XPS, as shown in Figure 1g,h. The P 2$p$ and the O 1$s$ signal prove the formation of the oxidation product of TOP. As shown in the Fourier transform infrared (FTIR) spectrum of MoS$_2$ QDs (Figure 1f), the four dominating peaks at
3347, 2928, 2856, and 1706 cm\(^{-1}\) can be observed, corresponding to NH\(_2\), CH\(_2\), CH\(_3\), and C=O groups, respectively. The peaks at 1117 and 1014 cm\(^{-1}\) are assigned to P=O [26,27], indicating that the oxidation product of TOP serves as the surface ligand of MoS\(_2\) QDs, which is in line with the XPS results.

![Figure 1.](image)

(a) TEM image, (b) high-resolution TEM images, (c) FTIR spectrum of the MoS\(_2\) QDs. (d) XPS survey spectrum of the MoS\(_2\) QDs. (e) High-resolution peak-fitting XPS spectrum of Mo 3d. (f) High-resolution peak-fitting XPS spectrum of S 2p. (g) High-resolution peak-fitting XPS spectrum of O 1s. (h) High-resolution peak-fitting XPS spectrum of P 2p.
Figure 2 shows the UV-visible absorption and photoluminescence (PL) spectra of MoS$_2$ QDs. The PL peak and absorption peak are located at 518 nm and 343 nm, respectively. The band gap of MoS$_2$ QDs can be calculated from the absorption spectrum according to Lambert–Beer’s law, which is described as:

$$(ahv)^2 = BK^2 (hv - E_g)$$  \hspace{1cm} (1)$$

where $B$ and $K$ are proportionality constants, $a$ is the absorption coefficient, $E_g$ is the band gap energy, and $hv$ is the energy of incident photons. Based on the quantum confinement effect [28], the $E_g$ of the as-prepared MoS$_2$ QDs is about 2.68 eV.

![Absorption and PL spectra of MoS$_2$ QD colloid.](image)

**Figure 2.** Absorption and PL spectra of MoS$_2$ QD colloid.

### 3.2. Characterization of Perovskite Films

To determine the influence of MoS$_2$ QDs on perovskite films, the top-view SEM images shown in Figure 3 were compared. It was noted that the film with MoS$_2$ QD modification exhibits more homogeneous grains, and some light-colored particles are distributed on top of the films, which can be observed as PbI$_2$ by X-ray diffraction (XRD) patterns in Figure 4a [6].

![SEM images of perovskite films.](image)

**Figure 3.** SEM images of perovskite films. (a) The control perovskite film. (b) Perovskite film treated with 0.075 mg/mL MoS$_2$ QDs. (c) Perovskite film treated with 0.10 mg/mL MoS$_2$ QDs. (d) Perovskite film treated with 0.15 mg/mL MoS$_2$ QDs. Scale bar: 1 µm.
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The XRD patterns of the perovskite films deposited on the PTAA and PTAA/MoS2 QD substrate are shown in Figure 4a. The perovskite films both present characteristic diffraction peaks [29]. There are no perovskite peak position shifts, indicating that the Mo or S atom did not diffuse into the perovskite crystal and enable doping. Compared with the control film, the perovskite film deposited on the PTAA/MoS2 QD substrate suggests a peak at 12.5° belonging to (001) crystal planes of PbI2, which is consistent with the SEM analysis [30–32]. Moreover, the increased intensity of the peaks corresponding to perovskites evidently indicates a higher quality of perovskite films and crystal-oriented growth. The promoted peak intensity can be attributed to the interaction between MoS2 QDs and perovskites. MoS2 QDs served as a growth template and induced crystal growth [33]. The FTIR spectra of the perovskite film with MoS2 QDs and without are shown in Figure 4c. The peak shifts from 1175 to 1191 cm−1, which overlaps P=O peaks. Moreover, there is a new peak at 1096 cm−1 belonging to P=O. Compared to the QD FTIR spectrum, the peak shifts from 1014 cm−1 to 1096 cm−1.

To determine the chemical effects of MoS2 QD modification, the buried interfaces were characterized using XPS [6]. The XPS results of the MoS2-QD-modified perovskite and control film are shown in Figure 4d. According to the XPS spectra of the Pb 4f region, the binding energy decreased from 136.58 eV and 141.45 eV to 136.40 eV and 141.28 eV, respectively. The lower binding energy of Pb could be ascribed to the strong interaction between the QD modification and Pb through the Lewis acid–base interaction. The XPS results confirm the interaction between the MoS2 QD layer and perovskite films, indicating increased electron density [34]. The XPS results confirm the interaction between the MoS2 QD layer and perovskite films.

To further characterize the effects of MoS2 QDs on the photophysical properties of perovskites, we observed the steady-state PL spectra of the perovskite films with and without MoS2 QDs, as shown in Figure 4b. It is noted that the PL peak blue-shifts from 761 nm to 759 nm with the assistance of MoS2 QDs, indicating a lower density of trap states [35,36]. Meanwhile, the perovskite film with MoS2 QDs presents a much higher PL intensity, which
means less nonradiative recombination losses [37]. Combined with the XPS results, the Pb 4f peaks shift toward the lower binding energy, demonstrating that the suppressed nonradiative recombination in the perovskite films is related to the interaction between the QDs and perovskites, which is responsible for the reduced traps and improved PL.

To evaluate the effects of MoS2 QDs on the carrier dynamics, time-resolved photoluminescence (TRPL) measurement was employed. As shown in Figure 5a, TRPL curves were fitted using a bi-exponential function described as:

\[ I(t) = I_0 + A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) \]

where \( \tau_1 \) and \( \tau_2 \) represent the fast decay time constant and the slow time constant, respectively, and \( A_1 \) and \( A_2 \) are the amplitudes of the fast and slow decay processes [38,39]. The average carrier lifetime (\( \tau_{ave} \)) was acquired through equation \( \tau_{ave} = \tau_1 f_1 + \tau_2 f_2 \).

![Figure 5](image_url)

Figure 5. (a) TRPL curves of perovskite films deposited on the substrates with and without MoS2 QDs. (b) The PL spectra of perovskite films with and without MoS2 QDs, where the schematic diagram of the test detail is given as an inset.

The fitting relevant parameters aresummarized in Table 1. The proportion of the fast decay component \( \tau_1 \) representing the surface trap-assisted nonradiative recombination decreased from 52% to 40.5%. Moreover, the prolonged average carrier lifetime (398.8 ns) compared with that of the control film (175.3 ns) further demonstrates the promoted photophysical properties of suppressing the nonradiative recombination processes.

| Samples       | \( \tau_1 \) (ns) | \( f_1 \) (%) | \( \tau_2 \) (ns) | \( f_2 \) (%) | \( \tau_{ave} \) (ns) |
|---------------|------------------|--------------|------------------|--------------|---------------------|
| Without MoS2  | 11.7             | 52.6         | 354.6            | 47.4         | 175.3               |
| With MoS2     | 11.5             | 40.5         | 662.5            | 59.5         | 398.8               |

We also noted the formation of PbI2 on the surface and at the grain boundaries. The PL peaks were compared to further understand the influence of PbI2 in Figure 5b. When the PCBM film as electron transport layer (ETL) was deposited on top of the perovskite films, the sample with QDs showed significantly weaker PL, demonstrating that carriers were extracted more efficiently the pure perovskite film. It is reported that the PbI2 located at the surface of the perovskite film could heal the halide vacancy forming type-I band alignment and reduce the trap state density [36,40–43].

3.3. Perovskite Solar Cell Performances

Typical perovskite solar cells (PSCs) were fabricated with the structure of ITO/PTAA/perovskite/PCBM/BCP/Ag, as shown in Figure 6a. A cross-section SEM of the device is shown in Figure 6b.
The current density–voltage (J–V) curves of the devices are provided in Figure 7a, while the device parameters are summarized in Table 2. As expected, due to the QDs’ passivation effect, the $V_{oc}$ (open circuit voltage) and $J_{sc}$ (short circuit current) were enhanced from 1.05 to 1.08 V and 23.25 mA/cm$^2$ to 23.79 mA/cm$^2$, respectively. Consequently, a relatively superior performance with a power conversion efficiency (PCE) from 17.87% to 19.95% was achieved. Figure 7c shows the statistic efficiency distribution. The current densities integrated from the incident photon to converted electron spectra (IPCE) increased from 21.67 to 22.15 mA/cm$^2$ with the MoS$_2$ modification, which is consistent with the increased $J_{sc}$. The enhancement of IPCE (Figure 7b) in the short wavelength part could be attributed to the MoS$_2$ QDs at the HTL interface acting as an interface passivator. Furthermore, the increase in long wavelength conversion may be related to the passivation effect of lead iodide at the electron collection side.

![Figure 6](image_url)

Figure 6. (a) A schematic diagram of the PSCs structure. (b) Cross-sectional SEM image of the control device.

![Figure 7](image_url)

Figure 7. (a) J–V curves of control device and devices treated with MoS$_2$ QDs at different concentrations. (b) IPCE measurement of control device and device treated with MoS$_2$ QDs. (c) Measurement for PCE of devices with and without MoS$_2$, where 22 cells were collected for each batch. (d) Capacitance–voltage curves of control device and device treated with MoS$_2$ QDs. (e) Nyquist plots of control device and device treated with MoS$_2$ QDs. Inset: An equivalent circuit model of the devices. (f) Dark J–V curves of control device and device treated with MoS$_2$ QDs.
To further investigate the influence of MoS\(_2\) QD modification on the carrier extraction/transport in devices, dark capacitance–voltage curves were measured, as shown in Figure 7d. The Mott–Schottky relationship can be described as follows:

\[
\frac{1}{C^2} = 2(V - V_{bi} - kT/e) / (\varepsilon\varepsilon_0eN_D)
\]

where \(V_{bi}\) is the voltage of the built-in electric field, \(V\) is the applied voltage, \(C\) is the capacitance, \(k\) is the Boltzmann’s constant, \(T\) represents the temperature, and \(N_D\) is the donor density. The fitting results show that the \(V_{bi}\) of the device with MoS\(_2\) QDs increased from 0.84 V to 0.86 V. The enhanced driving force to separate photogenerated carriers accounts for the higher \(V_{oc}\) [44,45].

Electrochemical impedance spectroscopy (EIS) under 0.85 V bias voltage and dark conditions was performed to further explore the charge transfer and recombination progress [1,46]. Nyquist plots of devices with and without MoS\(_2\) are presented in Figure 7e. An equivalent circuit model is depicted as an inset where series resistance (Rs) and recombination resistance (Rrec) represent the resistance corresponding to the behavior of carrier transport and recombination, respectively. The plot for devices with MoS\(_2\) QDs clearly shows a larger diameter of the semicircle than that of the control device, indicating a larger recombination resistance and chemical capacitance, which corroborates the suppression of carrier recombination and an improvement in transfer and extraction efficiency. This phenomenon can be ascribed to the passivation of the defects and band alignment at the interface. Dark \(J-V\) curves were measured to confirm the buffer function of MoS\(_2\) QD modification. As shown in Figure 7f, current leakage of the device was reduced by nearly more than one order of magnitude with the introduction of MoS\(_2\) QDs. The remarkable decrease in the current leakage demonstrates that MoS\(_2\) QDs as a buffer layer prevent the photogenerated electrons from flowing to the hole transport layer [47–49].

To further individually clarify the effect of PbI\(_2\) and MoS\(_2\), we adopted the mechanical exfoliation method to remove the PbI\(_2\) on the top surface of the perovskite films [50]. The films without PbI\(_2\) SEM images are shown in Figure 8a–c. Based on the treated films, we fabricated hole-only devices with and without MoS\(_2\).

**Table 2.** Relevant device parameters obtained from \(J-V\) curves.

| Samples         | \(J_{sc}\) (mA/cm\(^2\)) | \(V_{oc}\) (V) | FF (%) | PCE (%) |
|-----------------|--------------------------|----------------|--------|---------|
| 0 mg/mL MoS\(_2\) | 22.89                    | 1.05           | 74.35  | 17.87   |
| 0.075 mg/mL MoS\(_2\) | 23.30                    | 1.06           | 73.81  | 18.23   |
| 0.100 mg/mL MoS\(_2\) | 23.79                    | 1.08           | 77.64  | 19.95   |
| 0.125 mg/mL MoS\(_2\) | 23.99                    | 1.07           | 72.07  | 18.50   |

**Figure 8.** Cont.
Figure 8. (a,b) The films after mechanical exfoliation treatment without PbI2 morphology. (c) The film with MoS2 and PbI2. Scale bar in the SEM images is 3 μm. (d) SCLC measurement device treated with MoS2, with and without PbI2. Inset: the hole-only devices structure diagram.

Subsequently, space-charge-limited current (SCLC) measurements were carried out to estimate the trap densities by employing hole-only device configurations (ITO/PTAA/Perovskite/Spiro-OMeTAD/Ag). According to the SCLC mode, the defect state density is determined by the following equation [51,52]:

\[ n_t = \frac{2e\epsilon_0 V_{TFL}}{eL^2} \]  

(4)

where \( n_t \) is trap density; \( V_{TFL} \) represents trap-filled limited voltage; \( e \) is elementary charge; and \( \epsilon \) and \( \epsilon_0 \) are the permittivity of perovskite films and the vacuum permittivity, respectively.

Figure 8d suggests that the \( V_{TFL} \) for the optimized device is 0.70 V compared to the control device’s 0.81 V. Without PbI2, the defect density is reduced from \( 9.6 \times 10^{15} \) to \( 8.3 \times 10^{15} \), which could individually prove the MoS2 effects. The reduced trap density demonstrates that the introduction of MoS2 QDs can effectively suppress carrier recombination, which is consistent with the prolonged carrier lifetime and improved device performance. Furthermore, the films with MoS2 and PbI2 show the least defective state density, which is \( 6.9 \times 10^{15} \). These results prove the synergistic passivation of MoS2 and PbI2.

Electron-only devices were fabricated, and the trap state density for electron transfer was calculated in Figure 9b. The defect state density decreased from \( 1.25 \times 10^{16} \text{ cm}^{-3} \) to \( 0.82 \times 10^{16} \text{ cm}^{-3} \). As shown in the schematic diagram of band energy (Figure 9a), the PbI2 on the electron transfer layer side could facilitate the extraction of electrons and reduce the defect state density. Thus, we assume that the MoS2 on the buried interface and the PbI2 on the top interface promote hole and electron transfer, respectively.
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Figure 9. (a) Schematic diagram of band energy. (b) SCLC measurement of control device and device treated with MoS$_2$. Inset: the electron-only devices structure diagram. (c–f) The statistics of $V_{oc}$, $J_{sc}$, FF, and PCEs of 20 devices for each fabrication condition.

We introduced 5% excess PbI$_2$ and reduce the 5% proportion PbI$_2$ in the precursor on the base of the component proportion in the manuscript and then respectively fabricated devices, namely excessive PbI$_2$ and insufficient PbI$_2$. The devices with 5% excessive PbI$_2$, insufficient PbI$_2$ and control device performances are compared in the Figure 10.
Which indicates that a proportion of the control devices are best without modification. Conceptualization, W.Z. (Wei Zhang) and X.Z.; Data curation, Z.L., T.G., C.W. 

Table 3. Relevant device parameters obtained from J-V curves.

| Samples      | $J_{sc}$ (mA/cm$^2$) | $V_{oc}$ (V) | FF (%) | PCE (%) |
|--------------|----------------------|--------------|--------|---------|
| 0 mg/mL MoS$_2$ | 24.07                | 1.085        | 76.39  | 19.95   |
| 0.075 mg/mL MoS$_2$ | 23.25                | 1.063        | 73.64  | 18.20   |
| 0.100 mg/mL MoS$_2$ | 22.35                | 1.049        | 71.01  | 17.29   |
| 0.125 mg/mL MoS$_2$ | 21.42                | 1.072        | 72.51  | 16.32   |

Based on the above experiment and analysis, a conclusion could be drawn: the MoS$_2$ QDs could passivate the defect through the interaction without PbI$_2$, while the PbI$_2$ induced by MoS$_2$ reduced the trap density further. Simply adding excessive PbI$_2$ could not achieve higher efficiency and lower trap density. The box plot statistics of $V_{oc}$, $J_{sc}$, FF, and PCEs of 20 devices for each fabrication condition are shown in Figure 9c–f. The MoS$_2$-modified device without PbI$_2$ showed lower defect density. In a word, the PbI$_2$ induced by MoS$_2$ QDs and MoS$_2$ both play a role in passivation. Under synergistic effects, the current density and open voltage were both improved.

4. Conclusions

In summary, we employed MoS$_2$ QDs as interface passivating agents to improve the performance of perovskite solar cells. It was found that MoS$_2$ QDs can improve perovskite crystallinity and passivate interfacial defects, which prolongs carrier lifetimes and suppresses carrier recombination. Moreover, due to the wide bandgap of 2.6 eV, the MoS$_2$ QDs served as a buffer layer, reducing the shunting paths. Benefitting from the suppressed nonradiative recombination and reduced leakage current, the device performance of PSCs improved from 17.87% to 19.95% after MoS$_2$ QD modification. This work provides an exploration of QDs for PSC applications, specifically for perovskite film modification and buried interface passivation.

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**References**

1. Yang, Z.; Yu, Z.; Wei, H.; Xiao, X.; Ni, Z.; Chen, B.; Deng, Y.; Habisreutinger, S.N.; Chen, X.; Wang, K.; et al. Enhancing Electron Diffusion Length in Narrow-Bandgap Perovskites for Efficient Monolithic Perovskite Tandem Solar Cells. *Nat. Commun.* **2019**, *10*, 4498. [CrossRef] [PubMed]
2. Kumar, N.; Lee, H.B.; Hwang, S.; Kang, J.W. Large-Area, Green Solvent Spray Deposited Nickel Oxide Films for Scalable Fabrication of Triple-Cation Perovskite Solar Cells. *J. Mater. Chem. A* **2020**, *8*, 3357–3368. [CrossRef]
3. Sandhu, S.; Saharan, C.; Buruga, S.K.; Kumar, S.A.; Rana, P.S.; Nagajothy, P.C.; Mane, S.D. Micro Structurally Engineered Hysteresis-Free High Efficiency Perovskite Solar Cell Using Zr-Doped TiO2 Electron Transport Layer. *Ceram. Int.* **2021**, *47*, 14665–14672. [CrossRef]
4. Shao, Y.; Yuan, Y.; Huang, J. Correlation of Energy Disorder and Open-Circuit Voltage in Hybrid Perovskite Solar Cells. *Nat. Energy* **2016**, *1*, 15001. [CrossRef]
5. Chen, S.; Dai, X.; Xu, S.; Jiao, H.; Zhao, L.; Huang, J. Stabilizing Perovskite-Substrate Interfaces for High-Performance Perovskite Modules. *Science 2021*, *373*, 902–907. [CrossRef] [PubMed]
6. Yang, X.; Luo, D.; Xiang, Y.; Zhao, L.; Anaya, M.; Shen, Y.; Wu, J.; Yang, W.; Chiang, Y.H.; Tu, Y.; et al. Buried Interfaces in Halide Perovskite Photovoltaics. *Adv. Mater. 2021*, *33*, e2006435. [CrossRef]
7. Dong, W.; Zhang, X.; Yang, F.; Zeng, Q.; Yin, W.; Zhang, W.; Wang, H.; Yang, X.; Kershaw, S.V.; Yang, B.; et al. Amine-Terminated Carbon Dots Linking Hole Transport Layer and Vertically Oriented Quasi-2D Perovskites through Hydrogen Bonds Enable Efficient Leds. *ACS Nano* **2022**, *16*, 9679–9690. [CrossRef]
8. Kim, K.M.; Ahn, S.; Jang, W.; Park, S.; Park, O.O.; Wang, D.H. Work Function Optimization of Vacuum Free Top- Electrode by Pedot:Pss/Pi Interaction for Efficient Semi-Transparent Perovskite Solar Cells. *Sol. Energy Mater. Sol. Cells* **2018**, *176*, 435–440. [CrossRef]
9. Jia, X.; Wu, N.; Wei, J.; Zhang, L.; Luo, Q.; Bao, Z.; Li, Y.-Q.; Yang, Y.; Liu, X.; Ma, C.-Q. A Low-Cost and Low-Temperature Processable Zinc Oxide-Polyethylenimine (Zno:Pei) Nano-composite as Cathode Buffer Layer for Organic and Perovskite Solar Cells. *Org. Electron.* **2016**, *38*, 150–157. [CrossRef]
10. Zhang, X.; Lin, H.; Huang, H.; Reckmeier, C.; Zhang, Y.; Choy, W.C.; Rogach, A.L. Enhancing the Brightness of Cesium Lead Halide Perovskite Nanocrystal Based Green Light-Emitting Devices through the Interface Engineering with Perfluorinated Ionomer. *Nano Lett.* **2016**, *16*, 1415–1420. [CrossRef]
11. Degani, M.; An, Q.; Albaluejo-Siguan, M.; Hofstetter, Y.J.; Cho, C.; Paulus, F.; Granzini, G.; Vaynzof, Y. 23.7% Efficient Inverted Perovskite Solar Cells by Dual Interfacial Modification. *Sci. Adv.* **2021**, *7*, eabj7930. [CrossRef] [PubMed]
12. Zhang, X.; Sun, C.; Zhang, Y.; Wu, H.; Ji, C.; Chuai, Y.; Wang, P.; Wen, S.; Zhang, C.; Yu, W.W. Bright Perovskite Nanocrystal Films for Efficient Light-Emitting Devices. *J. Phys. Chem. Lett.* **2016**, *7*, 4602–4610. [CrossRef] [PubMed]
13. Shao, S.; Abdu-Aguye, M.; Qu, L.; Lai, L.-H.; Liu, J.; Adjakatse, S.; Jahani, F.; Kamminga, M.E.; Brink, G.H.T.; Palstra, T.T.M.; et al. Elimination of the Light Soaking Effect and Performance Enhancement in Perovskite Solar Cells Using a Fullerene Derivative. *Energy Environ. Sci.* **2016**, *9*, 2444–2452. [CrossRef]
14. Wang, Y.; Wu, T.; Barbadou, J.; Kong, W.; Cui, D.; Chen, H.; Yang, X.; Han, L. Stabilizing Heterostructures of Soft Perovskite Semiconductors. *Science 2019*, *365*, 687–691. [CrossRef]
15. Litvin, A.P.; Zhang, X.; Ushakova, E.V.; Rogach, A.L. Carbon Nanoparticles as Versatile Auxiliary Components of Perovskite-Based Optoelectronic Devices. *Adv. Funct. Mater. 2021*, *31*, 200768. [CrossRef]
16. Zhang, X.; Zeng, Q.; Xiong, Y.; Ji, T.; Wang, C.; Shen, X.; Lu, M.; Wang, H.; Wen, S.; Zhang, Y.; et al. Energy Level Modification with Carbon Dot Interlayers Enables Efficient Perovskite Solar Cells and Quantum Dot Based Light-Emitting Diodes. *Adv. Funct. Mater.* **2020**, *30*, 1910530. [CrossRef]
17. Zheng, X.; Troughton, J.; Gasparini, N.; Lin, Y.; Wei, M.; Hou, Y.; Liu, J.; Song, K.; Chen, Z.; Yang, C.; et al. Quantum Dots Supply Bulk- and Surface-Passivation Agents for Efficient and Stable Perovskite Solar Cells. *Joule* **2019**, *3*, 1963–1976. [CrossRef]
18. Tang, G.; You, P.; Tai, Q.; Yang, A.; Cao, J.; Zheng, F.; Zhou, Z.; Zhao, J.; Chan, P.K.L.; Yan, F. Solution-Phase Epitaxial Growth of Perovskite Films on 2D Material Flakes for High-Performance Solar Cells. *Adv. Mater.* **2019**, *31*, e1807689. [CrossRef]
19. Hu, W.; Jin, X.; Li, A.; Liu, C.L.; Wang, X.F. Efficiency Improvement of Inverted Perovskite Solar Cells Enabled by Ptaa/Mos2 Double Hole Transporters. *Nanotechnology* **2022**, *33*, 335202. [CrossRef]
20. Capasso, A.; Matteucci, F.; Najafi, L.; Prato, M.; Buha, J.; Cinà, L.; Pellegrini, V.; di Carlo, A.; Bonaccorso, F. Few-Layer MoS2 flakes as Active Buffer Layer for Stable Perovskite Solar Cells. Adv. Energy Mater. 2016, 6, 1600920. [CrossRef]

21. Kakavelakis, G.; Paradisonos, I.; Faci, B.; Generosi, M.; Papachatzakis, M.; Maksudov, T.; Najafi, L.; Del Rio Castillo, A.E.; Kisseoglu, G.; Stratakis, E.; et al. Extending the Continuous Operating Lifetime of Perovskite Solar Cells with a Molybdenum Disulfide Hole Extraction Interlayer. Adv. Energy Mater. 2018, 8, 1702287. [CrossRef]

22. Najafi, L.; Taheri, B.; Martin-Garcia, B.; Bellani, S.; di Girolamo, D.; Agresti, A.; Oropesa-Nunez, R.; Pescetelli, S.; Vesce, L.; Calabro, E.; et al. Mos2 Quantum Dot/Graphene Hybrids for Advanced Interface Engineering of a Ch3NH3PbI3 Perovskite Solar Cell with an Efficiency of over 20. ACS Nano 2018, 12, 10736–10754. [CrossRef] [PubMed]

23. Jawaid, A.; Nepal, D.; Park, K.; Jespersen, M.; Qualle, A.; Mirau, P.; Drummey, L.F.; Vaia, R.A. Mechanism for Liquid Phase Exfoliation of Mos2. Chem. Mater. 2016, 28, 337–348. [CrossRef]

24. Zhu, X.; Xiang, J.; Li, J.; Feng, C.; Liu, P.; Xiang, B. Tunable Photoluminescence of Mos2 Quantum Dots Passivated by Different Functional Groups. J. Colloid Interface Sci. 2018, 511, 209–214. [CrossRef] [PubMed]

25. Chen, J.; Li, S.; Ma, T.; Wu, D.; Zhao, Y.; Wang, C.; Zhao, D.; Li, X. Managing Lead Leakage in Efficient Perovskite Solar Cells with Phosphate Interlayers. Adv. Mater. Interfaces 2022, 9, 2200570. [CrossRef]

26. Lu, X.; Korgel, A.B.A.; Johnston, K.F. High Yield of Germanium Nanocrystals Synthesized from Germanium Dioxide in Solution. Chem. Mater. 2005, 17, 6479–6485. [CrossRef]

27. Chen, S.T.; Zhang, X.L.; Zhang, Q.H.; Tan, W.H. Trioctylphosphine as Both Solvent and Stabilizer to Synthesize Cds Nanorods. Nanoscale Res. Lett. 2009, 4, 1159–1165. [CrossRef]

28. Dong, H.F.; Tang, S.S.; Hao, Y.S.; Yu, H.Z.; Dai, W.H.; Zhao, G.F.; Cao, Y.; Lu, H.T.; Zhang, X.J.; Ju, H.X. Fluorescent Mos2 Quantum Dots: Ultrasonic Preparation, up-Conversion and Down-Conversion Bioimaging, and Photodynamic Therapy. ACS Appl. Mater. Interfaces 2016, 8, 3107–3114. [CrossRef]

29. Sadhu, A.; Rai, M.; Salim, T.; Jin, X.; Tan, J.M.R.; Leow, S.W.; Ahmed, M.G.; Magdassi, S.; Mhaisalkar, S.G.; Wong, L.H. Dual Role of Cu-Chalcogenide as Hole-Transporting Layer and Interface Passivator for P–I–N Architecture Perovskite Solar Cell. Adv. Funct. Mater. 2021, 31, 2103807. [CrossRef]

30. Zhu, H.; Liu, Y.; Eickemeyer, F.T.; Pan, L.; Ren, D.; Ruiz-Preciado, M.A.; Carlsen, B.; Yang, B.; Dong, X.; Wang, Z.; et al. Tailored Amphiphilic Molecular Mitigators for Stable Perovskite Solar Cells with 23.5% Efficiency. Adv. Mater. 2020, 32, 1907757. [CrossRef]

31. Jiang, X.; Zhang, J.; Ahmad, S.; Tu, D.; Liu, X.; Jia, G.; Guo, X.; Li, C. Dion-Jacobson 2d-3d Perovskite Solar Cells with Improved Efficiency and Stability. Nano Energy 2020, 75, 104892. [CrossRef]

32. Luo, C.; Zhao, Y.; Wang, X.; Gao, F.; Zhao, Q. Self-Induced Type-I Band Alignment at Surface Grain Boundaries for Highly Efficient and Stable Perovskite Solar Cells. Adv. Mater. 2021, 33, e2103231. [CrossRef] [PubMed]

33. Zheng, X.; Hou, Y.; Bao, C.; Yin, J.; Yuan, F.; Huang, Z.; Song, K.; Liu, J.; Troughton, J.; Gasparini, N.; et al. Managing Grains and Interfaces Via Ligand Anchoring Enables 22.3%-Efficiency Inverted Perovskite Solar Cells. Nat. Energy 2020, 5, 131–140. [CrossRef]

34. Leng, X.; Wan, K.; Wang, H.; Wu, G.; Zhang, X.; Liu, Y.; Han, B.; Wang, B.; Zhang, Y.; Zhou, H.; et al. Adenosine Triphosphate Disodium Modified Hole Transport Layer for Efficient Inverted Perovskite Solar Cells. ChemNanoMat 2022, 8, e202200041. [CrossRef]

35. Shao, Y.; Xiao, Z.; Bi, C.; Yuan, Y.; Huang, J. Origin and Elimination of Photocurrent Hysteresis by Fullerene Passivation in Ch3NH3PbI3 Planar Heterojunction Solar Cells. Nat. Commun. 2014, 5, 5784. [CrossRef]

36. Jiang, X.; Zhang, J.; Liu, X.; Wang, Z.; Guo, X.; Li, C. Deeper Insight into the Role of Organic Ammonium Cations in Reducing Surface Defects of the Perovskite Film. Angew. Chem. 2022, 134, e202115663. [CrossRef]

37. Liu, X.; Zhang, Y.; Shi, L.; Liu, Z.; Huang, J.; Yun, J.S.; Zeng, Y.; Pu, A.; Sun, K.; Hameiri, Z.; et al. Exploring Inorganic Binary Alkaline Halide to Passivate Defects in Low-Temperature-Processed Planar-Structure Hybrid Perovskite Solar Cells. Adv. Energy Mater. 2018, 8, 1801838. [CrossRef]

38. Chen, J.; Zhao, X.; Kim, S.; Park, N. Multifunctional Chemical Linker Imidazoleacetic Acid Hydrochloride for 21% Efficient and Stable Planar Perovskite Solar Cells. Adv. Mater. 2019, 31, 1902902. [CrossRef]

39. Wu, Z.; Jiang, M.; Liu, Z.; Jamshaid, A.; Ono, L.K.; Qi, Y. Highly Efficient Perovskite Solar Cells Enabled by Multiple Ligand Passivation. Adv. Energy Mater. 2020, 10, 1903696. [CrossRef]

40. Chen, Q.; Zhou, H.; Song, T.-B.; Luo, S.; Hong, Z.; Duan, H.-S.; Dou, L.; Liu, Y.; Yang, Y. Controllable Self-Induced Passivation of Hybrid Lead Iodide Perovskites toward High Performance Solar Cells. Nano Lett. 2014, 14, 4158–4163. [CrossRef]

41. Wang, L.; McCleese, C.; Kovalsky, A.; Zhao, Y.; Burda, C. Femtosecond Time-Resolved Transient Absorption Spectroscopy of Ch3NH3PbI3 Perovskite Films: Evidence for Passivation Effect of Pbi2. J. Am. Chem. Soc. 2014, 136, 12205–12208. [CrossRef] [PubMed]

42. Jiang, Q.; Chu, Z.; Wang, P.; Yang, X.; Liu, H.; Wang, Y.; Yin, Z.; Wu, J.; Zhang, X.; You, J. Planar-Structure Perovskite Solar Cells with Efficiency Beyond 21%. Adv. Mater. 2017, 29, 1703852. [CrossRef] [PubMed]

43. Li, Y.; Li, W.; Xu, Y.; Li, R.; Yu, T.; Lin, Q. Interfacial Engineering of Perovskite Solar Cells with Evaporated Pbi2 Ultrathin Layers. ACS Appl. Mater. Interfaces 2021, 13, 53282–53288. [CrossRef] [PubMed]

44. Zhang, F.; Guan, S.; Ye, M.; Xu, K.; Guo, W.; Liu, X.Y.; Xu, H. Chemical Decoration of Perovskites by Nickel Oxide Doping for Efficient and Stable Perovskite Solar Cells. ACS Appl. Mater. Interfaces 2018, 10, 36841–36850. [CrossRef]
45. Wang, L.; Yan, Z.; Qiu, J.; Wu, J.; Zhen, C.; Tai, K.; Jiang, X.; Yang, S. Interface Regulation Enables Hysteresis Free Wide-Bandgap Perovskite Solar Cells with Low Voc Deficit and High Stability. *Nano Energy* 2021, 90, 106537. [CrossRef]

46. Cao, Q.; Li, Y.; Zhang, H.; Yang, J.; Han, J.; Xu, T.; Wang, S.; Wang, Z.; Gao, B.; Zhao, J.; et al. Efficient and Stable Inverted Perovskite Solar Cells with Very High Fill Factors Via Incorporation of Star-Shaped Polymer. *Science* 2021, 7, eabg0633. [CrossRef]

47. Duan, J.; Zhao, Y.; Yang, X.; Wang, Y.; He, B.; Tang, Q. Lanthanide Ions Doped CsPbBr3 Halides for Htm-Free 10.14%-Efficiency Inorganic Perovskite Solar Cell with an Ultrahigh Open-Circuit Voltage of 1.594 V. *Adv. Energy Mater.* 2018, 8, 1802346. [CrossRef]

48. Zhang, J.; Liang, W.S.; Yu, W.; Yu, S.W.; Wu, Y.L.; Guo, X.; Liu, S.Z.; Li, C. A Two-Stage Annealing Strategy for Crystallization Control of Ch3Nh3Pbi3 Films toward Highly Reproducible Perovskite Solar Cells. *Small* 2018, 14, 1800181. [CrossRef]

49. Sandhu, S.; Singh, R.; Yoo, K.; Kumar, M.; Lee, J.-J. Effect of Binary Additives in Mixed 2d/3d Sn-Based Perovskite Solar Cells. *J. Power Sources* 2021, 491, 229574. [CrossRef]

50. Chen, S.; Liu, Y.; Xiao, X.; Yu, Z.; Deng, Y.; Dai, X.; Ni, Z.; Huang, J. Identifying the Soft Nature of Defective Perovskite Surface Layer and Its Removal Using a Facile Mechanical Approach. *Joule* 2020, 12, 2661–2674. [CrossRef]

51. Qi, X.; Wang, J.; Tan, F.; Dong, C.; Liu, K.; Li, X.; Zhang, L.; Wu, H.; Wang, H.-L.; Qu, S.; et al. Quantum Dot Interface-Mediated CsPbBr2 Film Growth and Passivation for Efficient Carbon-Based Solar Cells. *ACS Appl. Mater. Interfaces* 2021, 13, 55338–55346. [CrossRef] [PubMed]

52. Cai, Y.; Zhang, Z.; Zhou, Y.; Liu, H.; Qin, Q.; Lu, X.; Gao, X.; Shui, L.; Wu, S.; Liu, J. Enhancing the Efficiency of Low-Temperature Planar Perovskite Solar Cells by Modifying the Interface between Perovskite and Hole Transport Layer with Polymers. *Electrochim. Acta* 2018, 261, 445–453. [CrossRef]