Tailoring the Interfacial Chemical Interaction for High-Efficiency Perovskite Solar Cells

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Supporting Information

ABSTRACT: The ionic nature of perovskite photovoltaic materials makes it easy to form various chemical interactions with different functional groups. Here, we demonstrate that interfacial chemical interactions are a critical factor in determining the optoelectronic properties of perovskite solar cells. By depositing different self-assembled monolayers (SAMs), we introduce different functional groups onto the SnO2 surface to form various chemical interactions with the perovskite layer. It is observed that the perovskite solar cell device performance shows an opposite trend to that of the energy level alignment theory, which shows that chemical interactions are the predominant factor governing the interfacial optoelectronic properties. Further analysis verifies that proper interfacial interactions can significantly reduce trap state density and facilitate the interfacial charge transfer. Through use of the 4-pyridinecarboxylic acid SAM, the resulting perovskite solar cell exhibits striking improvements to the reach the highest efficiency of 18.8%, which constitutes an ~10% enhancement compared to those without SAMs. Our work highlights the importance of chemical interactions at perovskite/electrode interfaces and paves the way for further optimizing performances of perovskite solar cells.

KEYWORDS: Perovskite solar cells, interfacial chemical interactions, trap states passivation, coordination bond, self-assembly monolayer, energy level alignment

The advent of perovskite solar cells has rapidly jumped into the hot seat of academic research and industrial interest due to the recent surges in device performances and the potential low-cost solution-based manufacturing.1−6 The perovskite structure of the active layers can be described as ABX3, where the A is a cation, e.g. methylammonium (MA+), formamidinium (FA+), Cs+, etc., B is a metal cation, e.g., Pb2+, Sn2+, etc., and the X is an anion, e.g., I−, Br−, Cl−, SCN−, etc.2,4,7 One of the most striking features distinguishing the perovskite solar cells from other PV materials is its mixed ionic–covalent bonding perovskite structures, which enables the combined advantages of both solution processability and extraordinary optoelectronic properties, such as high dielectric constant,5,9 large absorption coefficient,10 long carrier lifetime,11 high tolerance for trap states,12 and more.

The device architecture of perovskite solar cells is typically a layer-by-layer configuration with a perovskite layer sandwiched between electrodes.13−16 Interfacial layers are applied between the perovskite and electrode layers in order to facilitate charge transport and collection and suppress interfacial charge recombination.13,14,17 The theory of energy level alignment18,19 was established as the guiding rule for the design of interfacial layers in solar cells.20,21 Electron transporting layers (ETLs) with low work functions are favored to align with the conduction band of perovskite (~3.75 eV for MAPbI3−xClx) for efficient electron charge carrier extraction22 and likewise for hole transporting layers (HTLs) at the anode (valence band ~5.31 eV for MAPbI3−xClx).13 A variety of efficient ETLs have been developed in recent years, including organic materials (e.g., [6,6]-phenyl-C61-butyric acid methyl ester (PC61BM),23 poly [(9,9-bis(3′- (N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN),24 polyethylenimine ethoxylated (PEIE),13 etc.25), metal oxides (TiO2,26 SnO2,20 ZnO,27 etc.), and more. Strong electron coupling is observed between these ETLs and the perovskite layer, which enable efficient electron carrier extraction and high device performance.13 Aside from the energy level alignment, it should be noted that different chemical interactions between the perovskite film and the interfacial layers can form due to the ionic nature of the perovskite.28,29 The formation of different surface chemical interactions or the different chemical terminations can dramatically change the electron density and energy level structures at the surface of the perovskite crystals, as studied in previous works.30−32 Therefore, chemically tailoring the interactions at the perovskite/electrode interfaces...
should also play a critical role in the interfacial optoelectronic properties but unfortunately has been rarely explored. The self-assembled monolayer (SAM) has been demonstrated as an effective strategy for fine-tuning the interfacial optoelectronic properties by bonding the perovskite layer to the electrode substrates.\[27,37]\ In using SAMs, the substrate work function can be adjusted due to the permanent dipole moment resulting from the ordered molecular alignment, which has been widely studied in organic electronics.\[36]\ Most importantly, the deposition of SAMs on substrates offers diverse linkages between the substrates and the ionic perovskite films. Although a variety of SAMs have been developed to enhance device performances of perovskite solar cells, the effects of chemical interactions between perovskite films and the substrates were neglected, as limited by the difficulty in distinguishing the role of chemical interactions from other contributing factors, e.g., energy level alignment and surface induced morphological variations.\[27,37]\ 

In this work, we systematically studied the independent role of interfacial chemical interactions for highly efficient perovskite solar cells. The SAMs with different terminal groups were anchored onto a SnO2 ETL to form different chemical interactions with the perovskite film. The chemical structures and schematic diagram for the various interactions are shown in the Figure 1a. van der Waals interactions with benzoic acid (BA) and dipolar interactions with the 4-pyridinecarboxylic acid (PA), 3-amipropionic acid (C3), 4-aminobenzoic acid (ABA), and 4-cyanobenzoic acid (CBA) were explored. In contrast to the energy level alignment theory, an opposite trend was observed in device performance of perovskite solar cells using different SAMs, which showed that interfacial chemical interactions play a dominant role in determining the interfacial optoelectronic properties and device performance (device structure shown in Figure 1a and b). The best device performances were obtained with the deposition of PA, and the highest device performance was able to reach 18.8% for a single scan and a steady state power output of 17.7%, composing an approximate 10% improvement compared to devices without SAMs. Further analysis showed that the enhanced device performances was a result of proper chemical interactions that reduced trap state density and facilitated interfacial charge transfer. Our results highlight the importance of chemical interactions for achieving highly efficient perovskite solar cells, which should be taken into consideration for future designs.

Results and Discussion. The deposition of different SAMs on the SnO2 surfaces was confirmed by the existence of N elements as shown in the X-ray photoelectron spectroscopy (XPS) in Figure S1. Compared to the bare SnO2 and BA-SAM modified SnO2 surfaces, obvious N 1s signals appeared at a binding energy of ~399.5 eV with PA, CBA, ABA, and C3 modification. The deposition of different SAMs formed different functional groups on the SnO2 surfaces. It has been reported that substrates properties can significantly influence the morphologies of perovskite films processed from the one-step solution method due to the surface induction effect.\[27,37]\ In the one-step solution process, the precursors, e.g., MAI and PbI2 are evenly mixed and casted onto the substrates. Crystallization is controlled by the nucleation of the perovskite films on the substrate, which can be affected by the substrate properties. Since the film morphology has been demonstrated to significantly influence the device performances of perovskite solar cells, it is necessary to exclude the influences of morphological surface variations in order to judiciously evaluate the role of chemical interactions on device performance. Here, the two-step solution process was adopted for the perovskite film formation to avoid this effect. In a two-step solution process, the PbI2 and MAI precursors are casted sequentially, and the crystallization is governed by the interdiffusion of PbI2 and MAI. An SEM image of the perovskite film on the pristine SnO2 substrate is shown in Figure 1c. The perovskite film exhibits grain sizes of several hundred of nanometers, which are closely packed to form a flat films without any pin-holes.\[26]\ Figure S2 shows the surface morphology of the polycrystalline perovskite films on different SAM-modified SnO2 substrates. The morphology of these perovskite films on different SAM-modified SnO2 substrates are shown to be similar in terms of surface morphology, e.g., crystal grain sizes and flat pinhole free textures. The X-ray diffraction (XRD) pattern of perovskite on SnO2 is shown in Figure 1d, where the diffraction peaks at 14.1° and 28.6° are attributed to the <110> and <220> faces of the perovskite structure. The tiny peak at 12.7° results from a slight excess of PbI2, the existence of which has been demonstrated to have a positive effect for grain boundary passivation.\[26]\ Figure S3 shows the XRD patterns of perovskite films on different SAM-modified SnO2 substrates. As can be seen, the perovskite films exhibit similar XRD peak positions and intensities. These results indicate the similar crystalline properties of perovskite film regardless of different substrate properties. It is thus safe to conclude that the perovskite film morphology shows little
dependence on substrate surface properties with different SAM modification. As a result, the changes in device performances can be independently attributed to the variations in interfacial optoelectronic properties of the perovskite solar cells.

The work function of the interfacial layer is critical for achieving optimal electrical contact at interfaces of perovskite solar cells. To distinguish the influence of different surface terminations on the performance of organic and perovskite solar cells with or without PA modification, we fabricated perovskite solar cells with the device architecture depicted in Figure 2a. For example, the PVSKSCs with a CBA modification show an opposite trend compared to that observed in the work function variation (Figure 2a). For example, the PVSKSCs with a CBA modification show an opposite trend compared to that observed in the work function variation (Figure 2a).

Table 1. Device Performance of Organic and Perovskite Solar Cells with Different SAMs Modification

<table>
<thead>
<tr>
<th>SAMs</th>
<th>$E_f$ (eV)</th>
<th>$J_{sc}$ (mA/cm$^2$)</th>
<th>$V_{oc}$ (V)</th>
<th>PCE* (%)</th>
<th>FF</th>
<th>$J_{sc}$ (mA/cm$^2$)</th>
<th>$V_{oc}$ (V)</th>
<th>PCE* (%)</th>
<th>FF</th>
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<tr>
<td>none</td>
<td>4.21</td>
<td>16.64</td>
<td>0.72</td>
<td>4.96</td>
<td>0.52 ± 0.21</td>
<td>0.414</td>
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<tr>
<td>CBA</td>
<td>4.29</td>
<td>16.82</td>
<td>0.60</td>
<td>4.51</td>
<td>4.14 ± 0.26</td>
<td>0.447</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>PA</td>
<td>4.17</td>
<td>16.52</td>
<td>0.77</td>
<td>7.16</td>
<td>6.87 ± 0.19</td>
<td>0.563</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BA</td>
<td>4.16</td>
<td>16.24</td>
<td>0.77</td>
<td>7.33</td>
<td>6.95 ± 0.22</td>
<td>0.586</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C3</td>
<td>4.07</td>
<td>16.85</td>
<td>0.78</td>
<td>7.80</td>
<td>7.67 ± 0.05</td>
<td>0.593</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>ABA</td>
<td>4.05</td>
<td>16.80</td>
<td>0.80</td>
<td>8.00</td>
<td>7.84 ± 0.11</td>
<td>0.595</td>
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</table>

*The PCE values outside of the parentheses are the best, and those in parentheses are the averaged values with deviations.

The work function of the interfacial layer is critical for achieving optimal electrical contact at interfaces of perovskite solar cells. The SAM-modified SnO$_2$ ETL shows a high device performance of 17.19%, with a $V_{oc}$ of 1.06 V, $J_{sc}$ of 21.65 mA/cm$^2$, and FF of 0.749, which is similar to previous reports. However, the PVSKSCs with the different SAM modifications show an opposite trend compared to that observed in the work function variation (Figure 2a). For example, the PVSKSCs with a CBA modification show a higher PCE of 18.27% compared to that of the control device, regardless of the increased work function. On the other hand, PVSKSCs with ABA show an obvious drop in device performance (16.50%) compared to that without any SAM, despite our expectation that the low work function at the interface should improve the device performance compared to that of bare SnO$_2$. Also, the PVSKSCs with the C3 show no enhancement in device performance compared to that on bare SnO$_2$. Here, we attribute the abnormal performance variations to the strong chemical interactions between the terminal functional groups of the SAMs and the ionic perovskite layers, where an intimate contact formed between the SAM and perovskite film induces a change of interfacial properties. For example, an amino or the cyano group can form bipolar bonds to the Pb$^{2+}$ ion by sharing the electron pair with the empty orbitals of Pb$^{2+}$. Actually, strong interactions between these functional groups, e.g., pyridine, have been shown to immobilize metal oxide nanoparticles with modified surfaces. Hanamura et al. and Gong et al. independently simulated the effects of different terminations on the surfaces of...
perovskite crystals. They concluded that the optoelectronic properties of the perovskite grain boundaries varied substantially with different terminations, e.g., the MA⁺ rich or the I⁻ rich grain surface. The interfacial structures between different terminations of perovskite and TiO₂ surfaces were also simulated to dramatically influence device performances.²² In our case, modified SnO₂ surfaces have different chemical groups to interact with the surfaces of the perovskite films. The optoelectronic property changes are induced at the surface of perovskite grains adjacent to the SnO₂ substrates. Also, the origin of reduced work function with SAMs relied on their permanent dipole moment, which have different electron density distributions. The coordination of SAMs with perovskite might affect the electron density distribution of SAMs and, thus, the work function. Thereafter, a device variation contrary to work function change of SnO₂/SAM is observed.

Remarkably, PVSKSCs on PA modified SnO₂ substrates show the most striking improvements compared to the devices with bare SnO₂ ETL layer, with a best PCE of 18.77%, V_OC of 1.10 V, J_SC of 22.03 mA/cm², and FF of 0.774. This device performance is among the highest achieved for planar heterojunction perovskite solar cells.¹³ Notably, a significant passivation effect was observed through use of pyridine according to a previous report.⁴⁹ As such, a similar passivation effect can be expected at the interface of SnO₂ and perovskite. Figure 2c shows the external quantum efficiency (EQE) of the PVSKSCs. As can be seen, the maximum EQE is ~94% and represents one of the highest in literature,⁴⁵ indicating the efficacy of optimal interfacial chemical interactions for efficient photon-to-electron conversion. The integrated J_SC (21.76 mA/cm²) from the EQE spectra matches well to that of the measured J_SC (22.03 mA/cm²) from I–V measurements. Optical simulations were carried out to calculate the internal quantum efficiency of the perovskite solar cells.⁴⁶,⁴⁷ Detailed information about the optical simulations can be found in the Supporting Information. As shown in Figure 2d, the simulated EQE of the perovskite solar cells (or the light absorbing fraction of perovskite within the whole device) shows a similar profile to that of the measured EQE. The appearance of peaks and valleys in the EQE spectra ranging from 550 to 800 nm can be attributed to the optical microcavity or the optical interference effects as proven by the optical intensity distribution of the whole device (Figure S4). The IQE is obtained by dividing the experimental EQE with the simulated EQE, which depicts the absorption fraction of the perovskite layer in the device. As can be seen, the calculated IQE spectra is over 90% in almost the whole absorption region of the perovskite solar cells. This result verifies the efficacy of the designed device architecture with PA modification for highly efficient internal photon-to-electron conversion.

One of the issues with perovskite solar cells is the hysteresis problem,⁴⁸,⁴⁹ where I–V characteristic curves can vary with different scanning directions, e.g., the forward scanning (FW, from negative bias to positive bias) or the reverse scanning (RS, from positive bias to the negative bias). The existence of hysteresis makes it difficult to accurately evaluate the device performance of perovskite solar cells. Alternatively, the steady output power has been regarded as a more reliable method to precisely characterize the true device efficiency of perovskite solar cells.⁴⁵ The hysteresis phenomenon is also observed in our case, which is shown in Figure 2e. The forward scan of perovskite solar cells without SAMs shows an efficiency of 15.6% and improved to 17.2% with deposition of PA. It is hard to determine whether the hysteresis issue is alleviated with PA, because both the forward and reverse scan device efficiencies were improved. By applied a bias voltage near the maximum power output point, we recorded the variation of the photocurrent density to measure the steady state output of the perovskite solar cells under AM 1.5 G 1 sun intensity. As shown in Figure 2f, the output curves of perovskite solar cells without any surface modification show a steady output power of 15.8 mW/cm², with bias of 0.88 V and a photocurrent of ~17.9 mA/cm², while for the PVSKSCs with PA, the steady output show the steady state output power of 17.7 mA/cm², with the bias of 0.88 V and the photocurrent of 20.1 mA/cm². These results clearly verify the improvement of device performance with PA modification and confirm the influence of interfacial chemical interactions on device performance.

We also deposited the BA onto the SnO₂ surface in order to form an aromatic rich surface. No chemical interaction was present except for a weak van der Waals force formed between the substrates and perovskite, which is even weaker than that with the pristine SnO₂. The device performance shows an appreciable improvement with a PCE of 18.11%. The improved device performance can be attributed to the lowered work function according to the energy level alignment theory.

Furthermore, we applied the SAMs to organic solar cells, where the interaction between the active layers and the substrates is only van der Waals type. As such, the influences of chemical interactions can be excluded in organic solar cells. The state-of-the-art poly[(4,8-bis-(5-(2-ethylhexyl)thiophen-2-yl)-benzo[1,2-b:4,5-b’]dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-b]thiophene)-2-carboxylate-2–6-diyl)];[6,6]-phenyl-C71-butyric acid methyl ester (PTB7-Th:PC71BM) composite film was selected as the active layer, and MoO₃/Ag was adopted as the anode (Figure 3a). The I–V
characteristic curves of organic solar cells with or without different SAM modification are depicted in Figure 3b, and the device parameters are summarized in Table 1. As can be seen, the organic solar cells with bare SnO2 as an ETL show poor efficiency of 4.95%, with a low $V_{OC}$ of 0.72 V and a low FF of 0.41. With the BA, PA, C3, and ABA modification, the device performance show significant improvement with the highest efficiency up to 8.00%. The $V_{OC}$ is significantly increased to 0.77–0.80 V, and the FF also shows appreciable improvement with BA, PA, C3, and ABAs, owing to the lowered work function of SnO2 as revealed by UPS measurements. With the CBA modification, the device PCE and $V_{OC}$ dropped remarkably, which is consistent with the higher work function of the CBA modified surfaces. The device performance variations conform to the work function changes with different SAMs, which abides to the energy level alignment theory. The sharp contrast in the device performance variations of perovskite solar cells and organic solar cells with different SAMs further stresses the importance of chemical interactions on interfacial properties for perovskite solar cells.

Device performances are closely correlated to charge dynamics in perovskite solar cells. Photoluminescence (PL) properties of the perovskite film provides a direct observation of photocarrier dynamics.11,22,40,50 As shown in Figure 4a, the steady state PL intensity of perovskite on ABA is mostly annihilated compared to the perovskite films without any SAM. The PL intensity of perovskite films on the C3 or BA shows higher than that on ABA, but still lower compared to that without SAM deposition. Little PL quenching is observed on perovskite films deposited on PA or the CBA compared to that on pristine SnO2 substrate. Figure 4b shows the transient PL of perovskite on SnO2 with different SAM modification. As shown, transient PL shows an obvious biexponential decay behavior. The fast decay PL species ($\tau_1$) can be attributed to quenching via trap states or interfacial charge transfer, and the longer PL species ($\tau_2$) is attributed to the bimolecular recombination via the light emission.\textsuperscript{12,40,50} It can be seen that the perovskite film on the bare SnO2 shows the longest lifetime of 150 ns ($\tau_1$), and the PL lifetimes are sequentially shortened to 62, 75, 83, 92, and 99 ns with deposition of C3, ABA, BA, PA, and CBA. The PL quenching and shortened PL lifetime with deposition of any SAM can be attributed to the enhanced electrical coupling (or faster charge transfer) between the perovskite and the SnO2 ETL or the increased interfacial trap states. However, the two PL quenching mechanisms can lead to significantly different carrier fates, where charge recombination occurs with traps but photocurrent is generated with interfacial charge transfer. To distinguish the two mechanisms, photocarrier dynamics in a real device were monitored via transient photovoltage decay measurements.

During the operation of a real device, photogenerated carriers, either free carriers or excitons, are mostly separated at the interfaces of Spiro-OMeTAD/perovskite or perovskite/ SnO2, resulting in strong PL annihilation by charge transfer at the interfaces. The separated charges can either be recombinant via surface trap states or be swept out of device for electrical power. In this scenario, the technique of transient photovoltage (TPV) can provide a clear picture of photocarriers dynamics during device operation.\textsuperscript{50,55} For the TPV measurement, the devices were operated at open-circuit voltage condition (a 6 MΩ resistance was applied) at a light intensity of 1 sun, and a laser pulse was generated to perturb the device and cause a rapid decay of photovoltage. The carrier lifetime is estimated to be the decay time at which the intensity become 1/e of the initial intensity. As shown in Figure 4c, the decay of transient photovoltage exhibits monoeponential decay. The carrier lifetime without SAMs is 4.1 $\mu$s. With deposition of BA, PA, and CBAs, the carrier lifetime is significantly improved to 5.4, 6.6, and 7.6 $\mu$s, respectively. With deposition of C3, the carrier lifetime decreases to 3.1 $\mu$s and becomes even shorter (1.5 $\mu$s).
with ABA. Since the deposition of SAM at the interface has little influence on the bulk properties of perovskite film, we attributed the change in the carrier lifetime to the modification of the surface or interface properties with SAM layer. Due to the open-circuit condition, all of the photocarriers either in the perovskite bulk or transferred at the interfaces can only be recombined via trap states. Thus, long carrier lifetimes indicate reduced trap states in real perovskite devices, and it is concluded that the deposition of CBA, PA, and BA-SAM can significantly reduce the surface trap states. Combining the reduced carrier lifetimes from transient PL measurements and the little changes in the PL intensity of the perovskite films with CBA and PA modification, we can see that the electronic coupling between the perovskite and SnO$_2$ layers were enhanced via deposition of these SAMs. In the case of ABA and C3, it seems that the trap state density at the interface was enhanced via deposition of these SAMs. In the case of ABA, the interaction of perovskite and SnO$_2$ substrate was increased according to the reduced carrier lifetimes from transient PL measurements, which also contribute to the reduced PL intensity. These results confirm that the chemical interactions at the electrode surfaces have a dramatic influence on the photocarrier dynamics and the interface trap states.

A schematic diagram is drawn in Figure 4d to show the carrier dynamics and the mechanism of enhanced device performance with deposition of PA. As illustrated, the photogenerated carriers can be relaxed to ground state via the light emission and trap state recombination. To generate the photocurrent, the photocarriers need to be collected by the electrode via charge transfer at the perovskite/electrode interfaces. The separated photocarriers need to bypass the interfacial recombination via the surface trap states in order to contribute to the output power. With the PA deposition, the surface trap states were suppressed as evidenced by the enhanced TPV decay time. The charge transfer at the perovskite/SnO$_2$ is enhanced due to the lowered work function and the improved interaction of perovskite and SnO$_2$ substrate with PA deposition, as evidenced by the shortened PL lifetime. The improved electronic coupling and suppressed interfacial traps contributed to the improved device performance.

**Conclusion.** In summary, we demonstrated that the interfacial chemical interactions between perovskite and charge transporting layers are important for device performance of perovskite solar cells. The ionic–covalent nature of perovskite films makes it easy to form various chemical interactions with different chemical groups, which can significantly influence interfacial optoelectronic properties, e.g., the surface trap state density. Through delicate control of the interfacial chemical interactions via self-assembled monolayers, we achieved highly efficient perovskite solar cells with device efficiencies reaching 18.8%, showing a 10% improvement compared to the that without SAM modification. We stress that the interfacial chemical interactions are an important consideration in designing highly efficient charge transporting layers for the future development of highly efficient perovskite solar cells.

**REFERENCES**


**ACKNOWLEDGMENTS**

This work was financially supported by a grant from The Air Force Office of Scientific Research (AFOSR, grant number: FA2386-15-1-4108, Program Manager: Dr. Charles Lee), UCLA Internal Funds, UC-Solar Program (Fund Number MRPI 328368), and the Enli Tech (in Taiwan) for donating the EQE measurement system to UCLA. We also like to acknowledge Dr. Yajie Jiang and Prof. Anita Ho-Baillie for assistance in optical modelling.

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**Notes**

The authors declare no competing financial interest.

**ASSOCIATED CONTENT**

* Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.6b04015.

Details regarding the experiments along with the supporting figures (PDF)


