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Harnessing Surface Dipole for $CsPbI_3$ Perovskite Solar Cells with Poly(3-hexylthiophene)

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The efficient functioning of perovskite solar cells largely depends on the interaction between perovskite halide materials and the hole-transport layer poly(3-hexylthiophene) (P3HT). However, a high rate of nonradiative recombination often hampers this interaction, leading to poor performance of the solar cells. We have developed a technique to modify the interface using a long-chain alkyl halide molecule called *n*-hexyl trimethylammonium bromide to address this issue. This modification technique significantly improves hole extraction, leading to an impressive open-circuit voltage of 1.14 V and a power conversion efficiency of 15.8% for inorganic perovskite $CsPbI_3$ with P3HT as a dopant-free hole-transport layer. This breakthrough can pave the way for developing more efficient and sustainable solar cells.

1. Introduction

Perovskite solar cell (PSC) efficiency has approached 83% of the Shockley–Queisser limit (theoretical limit) in its single junction and over 75% in tandem architectures.[1–3] However, the stability of PSCs is the chink in the armour for their market introduction.[4,5] Inorganic perovskite $CsPbI_3$ is one of the most stable compositions reported in the field of PSCs.[6] Though, there has been a lot of work dedicated to its crystallization and phase stability for $CsPbI_3$.[7–12] However, the $CsPbI_3$-based devices need precise improvement. The most efficient devices, with record efficiencies of over 21%, are only stable for up to a few hundred hours in operational conditions.[13,14] In contrast, the most stable device, which holds a record in stability with a T_{S80} lifetime of around 51,000 hours, has 17% efficiency.[6] The studies suggest room for improvement in the $CsPbI_3$ interface to make the devices more efficient by interfacial engineering.[6,11–15]

The perovskite absorber determines the working device's stability, and the other layers are equally crucial for the best performance and stability.[16,17] Generally, PSCs contain two essential charge extraction layers known as selective contacts, such as the electron-transport layer (ETL) and the hole-transport layer (HTL). The organic molecule spiro-OMeTAD (2,2′,7,7′-tetrakis (*N*,*N*-di-*p*-methoxyphenyl-amine)-9,9′-spirobifluorene) is one of the widely used HTLs in n–*i*–p device structures and is considered as the standard for the most efficient devices reported so far.[18] However, spiro-OMeTAD has several disadvantages for commercialization; for example, other than high cost, it is susceptible to high temperatures, needs time-consuming oxygen soaking, has film deposition issues, and requires dopants that may trigger perovskite layer degradation.[19–21] However, several efforts have been invested in improving its properties, which include changing the doping methods (e.g., CO_2 bubbling) and engaging dopants themselves.[22–24] However, the other physical properties of spiro-OMeTAD, such as temperature sensitivity and high costs, are still a problem. Hence, P3HT can be used as an alternative HTL to spiro-OMeTAD. The P3HT is comparatively low cost, easy to fabricate, and does not necessarily require doping.[25–27] Despite these advantages, there are some associated challenges with P3HT. It is a lower bandgap semiconductor compared to spiro-OMeTAD, i.e., ≈1.9 eV, and its conduction band minimum (CBM) is relatively close to perovskite's CBM, which results in nonradiative recombination at the interface and negatively affects the open-circuit voltage (V_{OC}).[28,29]

As a strategy to reduce recombination and resolve energetic misalignment, the interface between perovskite and P3HT is modulated by a dipole-forming molecule *n*-hexyl trimethyl ammonium bromide (HTAB) to mitigate the interfacial charge recombination. These 2D structures forming long-alkyl chain molecules improve phase stability and reduce surface defects.[12,30,31] We revealed that HTAB acts as a mediator between $CsPbI_3$ and P3HT. We have achieved over 15% power conversion efficiency (PCE) for $CsPbI_3$-based solar cells with P3HT as an HTM. This work does not provide a detailed study of the nonradiative recombination and chemical interaction. Nevertheless, it can be a good starting point for a study on how the interface can be engineered for P3HT as HTL, which can be improved in future work on $CsPbI_3$.

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2. Results and Discussion

We have prepared the β-$CsPbI_3$ film as reported in our recent work on $CsPbI_3$.[32,33] Conventionally, a long-alkyl chain molecule, i.e., *n*-octyl ammonium iodide (*n*-OAI), is used to improve stability. Here, we have introduced *n*-HTAB instead of *n*-OAI to passivate the interface. The chemical structure of *n*-HTAB and P3HT is shown in Figure S1, Supporting Information.

We performed steady-state photoluminescence (stPL) emission spectroscopy. In the stPL spectra, we observed a peak emission at 732 nm for the bare $CsPbI_3$ film, whereas, for the HTAB-treated $CsPbI_3$ film, the emission peak shifted slightly to 729 nm, corresponding to a bandgap E_g of $CsPbI_3$, with the reported literature values of ≈1.69–1.70 eV for $CsPbI_3$[34,35] (see **Figure 1**a). The higher PL intensity and blueshift in the emission peak indicate surface defect passivation in the HTAB-treated film.[11,36–38] Further, we have conducted stPL measurements for $CsPbI_3$ and HTAB-treated $CsPbI_3$ film capped with P3HT, deposited on quartz. We also observe a blueshift (at ≈729 nm) with HTL that indicates that HTAB largely passivates the interface and surface defects (see Figure 1b). We observe a PL quenching (a decrease in PL intensity) for HTL-capped film. Both charge extraction at the interface and defects states can contribute to the PL quenching.[39,40] The technique transient surface photovoltage (trSPV) helps us to differentiate the quenching due to charge extraction and defects states.[41] To validate and study the charge extraction at the interface (hole selectivity), we have measured trSPV.

We have used trSPV measurements to characterize the charge selectivity at the interface. The trSPV signals are directly proportional to the separated charges ($SPV(t) \approx (n(t)\text{-}p(t)) \times 0.5L$) at the buried interface of a device (L is the thickness of the light absorber layer, i.e., perovskite film in our study). In our recent work, we have employed this technique to study the charge extraction induced by the surface dipoles.[32] It is a state-of-the-art technique to study the interfacial charge dynamics and nonradiative recombination at the interface.[41–44] We have measured transient SPV for different samples, i.e., $CsPbI_3$, $CsPbI_3$/P3HT, $CsPbI_3$/HTAB, and $CsPbI_3$/HTAB/P3HT (all deposited on glass/FTO/TiO_2). In trSPV, an exponential rise in signals with a large amplitude shows an efficient charge extraction at the interface.[41–43] Nonradiative recombination or misalignment of the bands leads to deaccelerated rise and damping in amplitude.[41,44] These two processes (extraction and recombinations) have been highlighted in **Figure 2**b. TrSPV measurements suggest a significant boost of hole extraction in the HTAB-treated sample, in the presence of adjacent HTL, ≈3 times higher in the SPV amplitude and much faster signal rise than that of the bare $CsPbI_3$ sample in contact with the HTL. We thus attribute the higher SPV amplitude to a greater concentration of extracted free holes. It is worth mentioning that $CsPbI_3$ with P3HT and $CsPbI_3$ treated with HTAB (without P3HT) show almost similar signals, meaning favorable band banding for free hole extraction in the HTAB-treated samples. Overall, the SPV measurements show that HTAB treatment positively affects charge extraction.

Further, Figure S2b,c, Supporting Information shows the SPV spectral response with time as contour plots in the 0.8–3 eV photon energy range. We observe a clear signal below the bandgap (1.55 eV) related to the absorption of shallow and deep defects.[45] HTAB-treated samples have less pronounced signals from deep defects with activation energy around 1.0 eV. These results are in agreement with the increase of PL after HTAB treatment. It can be inferred that *n*-HTAB may interact chemically with mobile iodine ions to reduce the iodine defects. The HTAB molecule has a long-chain aliphatic chain ($C_6H_{13}^-$) connected to the functional group ($N^+(CH_3)_3^-$) as shown in Figure S1, Supporting Information. The functional group interacts with the mobile iodine ions through the electrostatic interactions;[28,46] however, the aliphatic chain templates the self-assembly of P3HT on the surface of perovskites.[28,47] These long-chain molecules make a 2D layer, improving the stability of the PSCs.[12,30,31]

The trSPV measurements indicate improved charge extraction at the interface. However, understanding the mechanism behind this enhanced extraction necessitates a closer examination of the energetics at the interfaces. We have employed the Kelvin probe (KP) method to measure the work function (WF), denoted as (Φ), which represents the energy difference between the vacuum level E_{vac} and the Fermi level (E_f) of the material, as described in Equation (1)

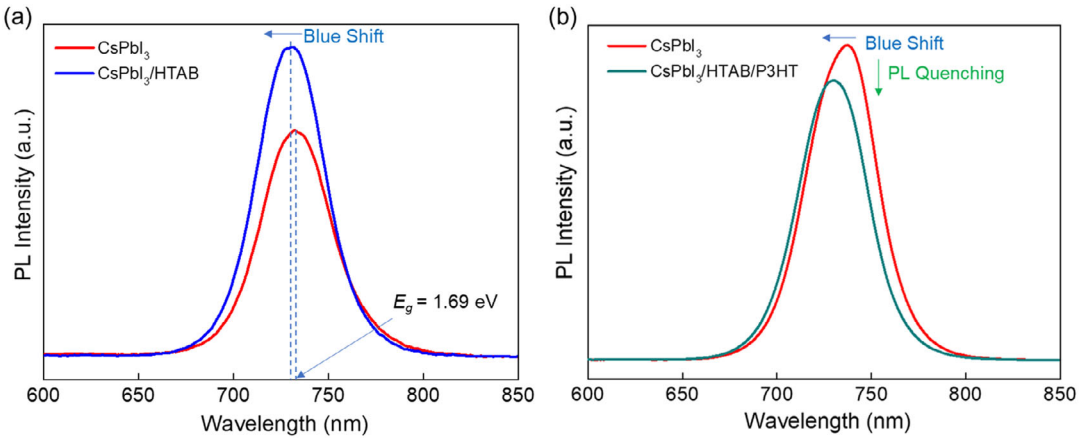

Figure 1. a) Steady-state photoluminescence emission spectra of bare $CsPbI_3$ and HTAB-treated $CsPbI_3$ film. b) Steady-state photoluminescence spectra of HTAB-treated perovskite films capped with P3HT (HTM).

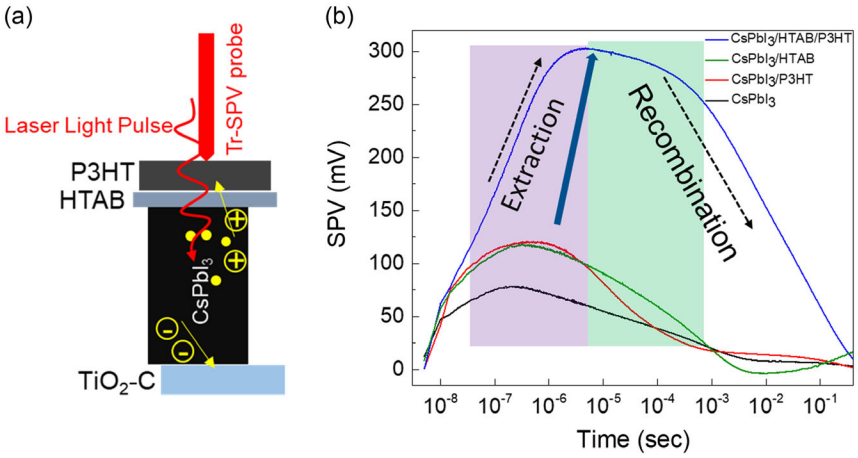

Figure 2. Transient SPV study: a) charge extraction and recombination model describing carrier transport to HTLs. b) trSPV measurements at an excitation source of 688 nm (1.8 eV) with the light intensity of 1 sun illumination of bare $CsPbI_3$ film, $CsPbI_3$ film covered with P3HT on the top, and $CsPbI_3$ passivated with HTAB with and without HTL. All the samples were deposited on FTO/TiO_2 substrates.

$$\Phi = E_{vac} - E_f \quad (1)$$

The determination of the WF is crucial to explain free hole extraction enhancement from the point of view of energy-level alignments. It is known that the CBM of P3HT is very close to the perovskite CBM, which gives rise to nonradiative recombination due to the recombination of photogenerated electrons.[28,29,48] The energy levels of a material can be tuned by dipoles that cause a change in vacuum level (E_{vac}), CBM, and VBM as well.[49] To investigate the impact of the HTAB molecule on energy levels, we conducted KP measurements on $CsPbI_3$, $CsPbI_3$/P3HT, and $CsPbI_3$/HTAB/P3HT (all deposited on glass/FTO) as shown in Figure S3, Supporting Information. **Figure 3** illustrates that WF shifts downward in energy. The downward shift in the WF indicates the dipole formation at the HTL.[49,50] This implies that HTAB acts as a Lewis base and its treatment induces a change in the interface that improves energy-level alignment. These results agree with enhanced charge extraction and improved band bending by HTAB at the interface.

Further, we have used bare $CsPbI_3$ and HTAB-capped $CsPbI_3$ absorbers to fabricate n–*i*–p devices in the following architecture: glass/FTO/compact-TiO_2/$CsPbI_3$/P3HT/Au and glass/FTO/compact-TiO_2/$CsPbI_3$:HTAB/P3HT/Au as shown in **Figure 4**a. Figure 4b shows *J*–*V* curves for the champion devices with and without HTAB-treated $CsPbI_3$ films. The HTAB-treated absorber-based device exhibits a PCE of 15.8%, a V_{OC} of 1.14 V, a current density (J_{SC}) of 19.4 mA cm^{-2}, and a fill factor (FF) of 71.4%, clearly outperforming the champion device without HTAB, reaching a PCE of 12.9%, a V_{OC} of 1.07 V, a J_{SC} of 19.2 mA cm^{-2}, and an FF of 62.3% (Table S1, Supporting Information). It also shows that HTAB-treated devices show less hysteresis and better stability as compared to other devices and stabilize PCE for an initial 500 s at maximum power point, as

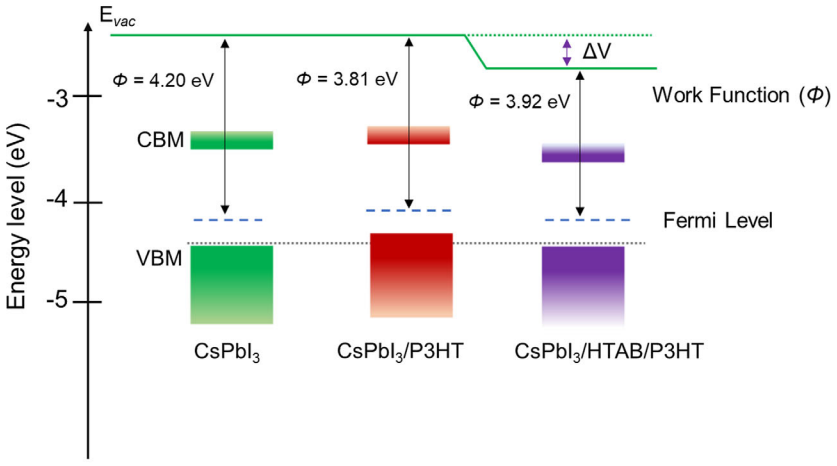

Figure 3. WF (Φ) measurements with the KP method for bare $CsPbI_3$ film, $CsPbI_3$ film covered with P3HT on the top, and $CsPbI_3$ passivated with HTAB with HTL. All the samples were deposited on glass/FTO substrates and measured in a nitrogen environment. The values of CBM and VBM are not absolute.

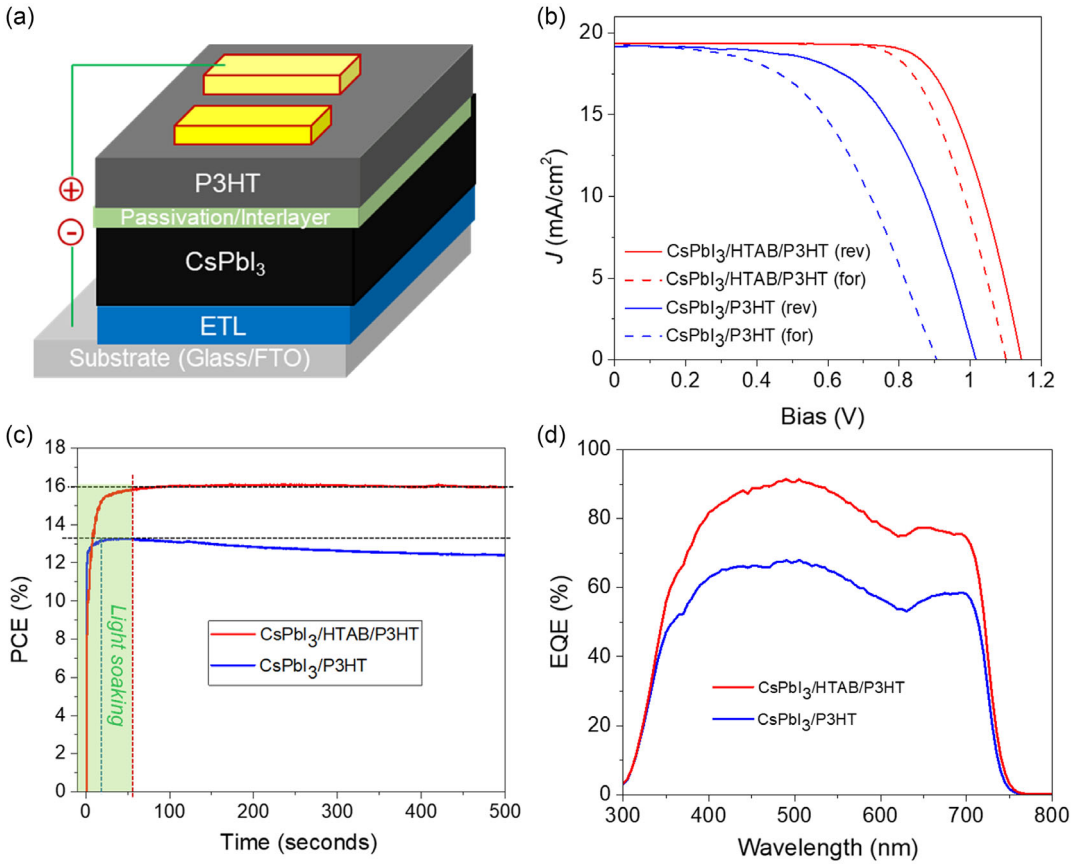

Figure 4. a) Device architecture of n–*i*–p planar $CsPbI_3$ PSCs. b) *J*–*V* curve of the champion devices for $CsPbI_3$ with and without HTAB treatment, measured at 1 sun AM1.5G at room temperature inside the nitrogen-filled glove box at the scan rate of 200 mV s^{-1} for both forward (dash line) and reverse (solid line). c) Stabilized PCE output at the maximum power point for an initial 500 s. d) EQE curves for the champion devices.

demonstrated in Figure 4c. Generally, it needs a light soaking for inorganic PSCs to achieve high efficiency.[51] There has been a lot of discussion about the "light soaking effect" and its mechanism in PSCs.[52–57] Considering the higher PCE of target HTAB devices, it will need a longer soaking time to reach equilibrium. Additionally, formed dipoles can play a role in transient behaviour. We attribute enhancement in device performance to dipole formation at the interface and defect passivation that leads to an improved charge extraction.[58,59] The external quantum efficiency (EQE) spectrum tells the conversion efficiency of photons illuminated on a solar cell to collected electrons at individual wavelengths. Figure 4d shows the EQE spectra of the champion devices with their *J*–*V* curves given in Figure 4b. It indicates that HTAB-treated devices exhibit an EQE of around 90% in broad wavelengths ranging from 400 to 590 nm, indicating a reduction in nonradiative recombinations.[60] It is reported that, for all compositions, open-circuit voltage loss (defined as $E_g - eV_{oc}$, whereas E_g is the optical bandgap of the absorber, e is the elementary charge, and V_{oc} is the measured open-circuit voltage of the device) is higher in inorganic PSCs than in organic–inorganic solar cells due to energy-level misalignment at the interfaces.[48,61] Here, we see a clear improvement in V_{OC} from 1.01 V to 1.14 V for our champion devices, which infers a better energy alignment at the interface due to this dipole molecule treatment. The significant increase in V_{OC} and EQE indicates the reduction in nonradiative recombination at the interfaces due to less defects as revealed by PL and trSPV measurements.[62,63]

3. Conclusion

Due to its low cost and temperature robustness, the organic dopant-free polymer P3HT can be an alternative to relatively temperature-sensitive spiro-OMeTAD as HTL in n–*i*–p devices. However, it requires improvement of nonradiative recombination losses and energetic alignment at the interface. We have employed a dipole molecule to mitigate these challenges. This dipole-forming molecule helps to improve the film quality by decreasing the surface defects. The molecule HTAB forms a layer on the $CsPbI_3$ surface and induces a surface dipole that leads to better energy levels at the interface. The trSPV measurements reveal that HTAB acts as a charge extraction mediator and also passivates the sub-bandgap defects in $CsPbI_3$. We also observed PL quenching in HTL-capped samples, indicating a charge extraction. It further suggests a boost in charge extraction in HTAB-treated samples. With HTAB-treated $CsPbI_3$ film, over 15% efficiency was achieved for dopant-free P3HT-based devices. This work further suggests that dopant-free

devices can endure the higher temperature under operational conditions.

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 support the findings of this study are available in the supplementary material of this article.

Keywords

$CsPbI_3$, hole-transport layers, inorganic perovskite solar cells, interface design, poly(3-hexylthiophene), surface dipoles, time-resolved surface photovoltage

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