Efficient perovskite/fullerene planar heterojunction solar cells with enhanced charge extraction and suppressed charge recombination†

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Alcohol soluble titanium chelate TIPD (titanium (diisopropoxide) bis(2,4-pentanedionate)) was used as an electron transporting layer to form an ohmic contact with the negative electrode, aiming to enhance the charge extraction and suppress the charge recombination for high performance CH$_3$NH$_3$PbI$_3$/PCBM-based PHJ perovskite solar cells. The TIPD layer shows excellent suitability to CH$_3$NH$_3$PbI$_3$ perovskite synthesized by different methods. For one-step synthesized CH$_3$NH$_3$PbI$_3$, the power conversion efficiency (PCE) of the device with the TIPD buffer reaches 8.75%, with a nearly 33% increase in comparison with the device without the buffer layer (6.58%). For two-step synthesized CH$_3$NH$_3$PbI$_3$, an open-circuit voltage ($V_{oc}$) of 0.89 V, a short-circuit current density ($J_{sc}$) of 22.57 mA cm$^{-2}$, and a fill factor (FF) of 64.5%, corresponding to a PCE of 12.95% for the device with a TIPD buffer layer were achieved, which is among the best performances reported in the literature for CH$_3$NH$_3$PbI$_3$/PCBM-based PHJ perovskite solar cells.

Introduction

Three-dimensional organometal halide perovskite CH$_3$NH$_3$PbI$_3$ is an ideal light harvesting absorber for solar cells due to its unique features such as low exciton binding energy and a large Bohr exciton radius with large dielectric constants,1 balanced charge mobility with a long diffusion length,2,3 and a direct optical bandgap of around 1.5 eV with a high extinction coefficient.4 Since the milestone breakthrough of introducing organic hole transporting 2,2′,7,7′-tetakis(N,N-di-p-methoxyphenylamino)-9,9′-spirobifluorene (Spiro-OMeTAD) to replace the liquid electrolyte,4,6 all solid state perovskite solar cells have achieved significant progress.2,3,7–9 The great achievement of perovskite solar cells should be attributed to the two communities from dye-sensitized solar cells (DSSCs) and organic photo-voltaics (OPVs) by fine tuning the functional materials and device architectures for the mesoscopic and planar heterojunction (PHJ) structured perovskite solar cells, respectively, over the past few years.

Planar architecture, comprised of thin-film intrinsic perovskite between the hole and electron transporting buffer layers, provides enhanced flexibility for device optimization, multijunction construction, and potentially high throughput manufacturing processes.10 The commonly used hole and electron transporting layers are PEDOT:PSS (poly[3,4-ethylenedioxythiophene]:poly(styrene sulfonate) or spiro-MeOTAD, and PCBM ([6,6]-phenyl-C$_{61}$-butyric acid methyl ester) or compact TiO$_2$, respectively. Therefore, a p–i–n typed PHJ device with the structure of PEDOT:PSS/perovskite/PCBM and an n–i–p typed PHJ device with the structure of a compact TiO$_2$/perovskite/hole transportation layer can be fabricated. Obviously, a pin-hole-free perovskite layer with optimal crystalline morphology and charge transportation layers with an ohmic contact with electrodes are essential to high performance PHJ perovskite solar cells. The quality of perovskite morphology has a great impact on charge dissociation, transportation, and recombination, while the contact properties of the electrodes could affect the charge recombination and collection. Several approaches have been proposed to improve the quality of perovskite morphology such as sequential deposition,7 vapor assisted solution processes,15 additive enhanced crystallization,19 and interdiffusion of solution-processed precursors.20 However, work concerning changing the contact properties of the electrodes is still very limited.18
Titanium chelate shows high electron mobility and is highly transparent in the visible region. It has been successfully used as an electron transporting layer in conventional- and inverted-structured polymer solar cells.\cite{21-25} Herein, we used the alcohol soluble titanium chelate TIPD (titantium (di-isopropoxide) bis(2,4-pentanedionate)) as the electron transporting layer to form an ohmic contact with the negative electrode, aiming to enhance the charge extraction and suppress the charge recombination for high performance CH$_3$NH$_3$PbI$_3$/PCBM-based PHJ perovskite solar cells. To investigate the suitability of the TIPD layer, the one-step and two-step synthesized CH$_3$NH$_3$PbI$_3$ perovskite was used as a light absorber, and PCBM was selected as an electron acceptor. For one-step synthesized CH$_3$NH$_3$PbI$_3$, the power conversion efficiency (PCE) of the device with the TIPD buffer reaches 8.75%, with a nearly 33% increase in comparison with the device without the buffer layer (6.58%). For two-step synthesized CH$_3$NH$_3$PbI$_3$, an open-circuit voltage ($V_{oc}$) of 0.89 V, a short-circuit current density ($J_{sc}$) of 22.57 mA cm$^{-2}$, and a fill factor (FF) of 64.5%, corresponding to a PCE of 12.95% for the device with a TIPD buffer layer were achieved, which is among the best performances reported in the literature for CH$_3$NH$_3$PbI$_3$/PCBM-based PHJ perovskite solar cells.

### Experimental

#### Materials

Patterned FTO glass (sheet resistance: $\sim$15 $\Omega$ sq$^{-1}$) was purchased from Wuhan Geao Instruments Science & Technology Co., Ltd (China). PEDOT:PSS (Clevios P VP Al 4083) was purchased from H. C. Stark. Methylamine solution (40 wt% in methanol), hydriodic acid (57 wt% in H$_2$O), and lead(II) iodide (99.999%) were purchased from Sigma-Aldrich. 75% TIPD isopropanol solution was purchased from Alfa Aesar. All these commercially available materials were used as received without further purification.

#### Synthesis: one-step

CH$_3$NH$_3$PbI$_3$ perovskite was synthesized from an equimolar mixture of CH$_3$NH$_3$I and PbI$_2$ in N,N-dimethylmethanamide (DMF) at 60 °C and stirred overnight according to the references with minor modifications.\cite{4,26} CH$_3$NH$_3$I was synthesized by reacting CH$_3$NH$_2$ with HI. Typically, an 11.2 mL HI aqueous solution (57% mol/mol) was added into an 11.6 mL CH$_3$NH$_2$ solution (40%, 0.15 mol) in a 50 mL round-bottomed flask at 0 °C for 2 h with constant stirring under a nitrogen atmosphere. The obtained mixture solution was evaporated at 50 °C with a rotary evaporator, and the resulting powder was washed with ethyl ether and vacuum dried at 60 °C overnight.

#### Synthesis: two-step

CH$_3$NH$_3$PbI$_3$ perovskite was prepared by following the vapor-assisted solution process with some modification.\cite{15} Typically, a PbI$_2$ DMF solution (400 mg mL$^{-1}$) was spin-coated (2000 rpm) on the FTO/PEDOT:PSS substrate, followed by transferring the substrate into a Petri dish with CH$_3$NH$_3$I powder spread around, and then heated at 150 °C for 2 h in a vacuum oven.

### Device fabrication

The FTO glass was sequentially cleaned by ultrasonic treatment in detergent, deionized water, acetone, and isopropanol, followed by ultraviolet-ozone (UVO) treatment for 15 min. PEDOT:PSS aqueous solution filtered through a 0.45 μm filter was spin-coated (2000 rpm, 60 s) on the FTO electrode, and then baked at 150 °C in air for 10 min. Then the FTO/PEDOT: PSS substrate was transferred to a nitrogen-filled glove-box for the film deposition. For one-step synthesized CH$_3$NH$_3$PbI$_3$, the perovskite absorber was deposited by spin coating (5000 rpm) its DMF precursor solution (15 wt%) on the FTO/PEDOT:PSS substrate for 30 s, followed by thermal annealing at 150 °C for 10 min. For two-step synthesized perovskite, the absorber layer was obtained by in situ formation of the CH$_3$NH$_3$PbI$_3$ layer on the FTO/PEDOT:PSS substrate as mentioned in the two-step synthesis. After that, the PCBM solution (20 mg mL$^{-1}$ in chlorobenzene) was spin-coated (1000 rpm, 30 s) on the perovskite absorber layer followed by thermal annealing at 100 °C for 10 min. The TIPD electron collection layer was prepared by spin coating (2000 rpm) a 3.75 wt% TIPD isopropanol solution on the PCBM layer and then baking it at 150 °C for 10 min. Finally, a 100 nm Al electrode was thermally deposited under a base pressure of 5 × 10$^{-5}$ Pa. The active area of the device is ca. 4 mm$^2$.

### Device characterization

The current density–voltage ($J$–$V$) measurements of the devices were conducted with a computer-controlled Keithley 2400 Source Measure Unit (SMU) in a nitrogen-filled glove-box under simulated AM1.5G irradiation (100 mW cm$^{-2}$) using a xenon-lamp-based solar simulator (SAN-EL, AAA grade). The incident photon to current efficiency (IPCE) was measured by QE-R systems (Enli Tech.) The light intensity at each wavelength was calibrated with a standard single-crystal Si photovoltaic cell. The IPCE measurements were performed under ambient conditions at room temperature.

### Instrumentation

A Lambda 950 UV/Vis/NIR spectrophotometer was used to measure the absorption of the thin film and the reflection of the devices. An AC Mode III (Agilent) atomic force microscope (AFM) was used to measure the surface morphologies of the thin films operated in the tapping mode. The surface and the cross-section morphology of the samples were observed by scanning electron microscopy (SEM) of FEI Quanta 200F at an accelerating voltage of 30 kV. X-ray diffraction (XRD) patterns were recorded with a diffractometer (Shimadzu XRD 6000) using Cu Kα ($\lambda = 0.15406$ nm) radiation with a nickel filter operating at 40 kV and 10 mA in the 2θ range of 5–60° at a scanning rate of 3° min$^{-1}$. A Dektak XT (Bruker) surface profilometer was used to measure the thickness of the films.
involved in the devices. All the measurements were carried out under ambient conditions at room temperature.

For a steady photoluminescence (PL) test, a second harmonic field (400 nm) generated from a femtosecond oscillator (Vitara, Coherent) was employed as the excitation source, and an ultra-steep long-pass filter (BLP01-405R-25, Semrock) was used to eliminate the residual excitation light. The emission spectra were collected and analyzed by a spectrograph (Sp 2500i, Princeton Instruments) equipped with a charge-coupled device cooled by liquid nitrogen. For time-resolved photoluminescence (TRPL) measurement, the spectrograph was equipped with a fast single-photon avalanche photodiode (PDM, Picoquant), and the temporal resolution for the detection is 50 ps.

**Results and discussion**

In this study, a sandwiched planar heterojunction device structure was designed for perovskite solar cells, as shown in Fig. 1a, where PEDOT:PSS works as the hole transporting layer, CH$_3$NH$_3$PbI$_3$ is the photo harvesting layer and electron donor, PCBM works as the electron acceptor, and TIPD works as the electron transporting layer. For comparison, control devices without TIPD are also fabricated. The device structures involved in this study are: (A) FTO/PEDOT:PSS/perovskite (one-step)/PCBM/Al (control device), (B) FTO/PEDOT:PSS/perovskite (one-step)/PCBM/TIPD/Al, (C) FTO/PEDOT:PSS/perovskite (two-step)/PCBM/Al (control device), and (D) FTO/PEDOT:PSS/perovskite (two-step)/PCBM/TIPD/Al. The multilayered sandwich structure (D) was further confirmed by cross-section scanning electron microscopy (SEM) as shown in Fig. 1b. The optimized thicknesses for PEDOT:PSS, CH$_3$NH$_3$PbI$_3$, PCBM, and TIPD are 30, 340, 30, and 15 nm, respectively. The energy levels of the materials involved in the perovskite solar cells are given in Fig. 1c. The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energy levels of CH$_3$NH$_3$PbI$_3$, PCBM, and TIPD are taken from the literature. When light irradiates from the FTO side, the CH$_3$NH$_3$PbI$_3$ layer absorbs photons to produce excitons, and the weakly bonded photo-generated excitons diffuse towards and dissociate at the PEDOT:PSS/CH$_3$NH$_3$PbI$_3$ and CH$_3$NH$_3$PbI$_3$/PCBM interfaces into free carriers with ease. Since the LUMO level of TIPD (−3.9 eV) is the same as that of PCBM, electrons can easily be transported to the Al electrode through TIPD. When the HOMO level of TIPD (−6.0 eV) is greatly lower than that of CH$_3$NH$_3$PbI$_3$ (−5.4 eV), the hole cannot be transported through TIPD to the Al electrode. On the other hand, the TIPD works as an electron transporting/hole blocking layer and this should benefit the charge collection.

CH$_3$NH$_3$PbI$_3$ perovskite was synthesized by one-step (mixing CH$_3$NH$_3$I and PbI$_2$ in DMF (N,N-dimethylformamide)) and two-step methods, respectively, (spin-coated PbI$_2$ treated with CH$_3$NH$_3$I vapor) according to literature reports with some modification. The absorption and photoluminescence spectra of the obtained CH$_3$NH$_3$PbI$_3$ perovskite thin film spin-coated on quartz slides are given in Fig. S1 in the ESI.

The crystalline quality of the resulting perovskite films deposited on the FTO/PEDOT:PSS substrate was investigated by X-Ray Diffraction (XRD) spectroscopy. For comparison, the XRD spectra of the FTO/PEDOT:PSS substrate (Fig. 2a), CH$_3$NH$_3$I powder (Fig. 2b), as well as the PbI$_2$ spin-coated on the FTO/PEDOT:PSS substrate (Fig. 2c) are also given. For the FTO substrate, although coated with an amorphous PEDOT:PSS layer, the diffraction peaks of (110), (101), (200), (211), and (220) are still visible as shown in Fig. 2a. Fig. 2b displays the XRD spectrum of the synthesized CH$_3$NH$_3$I powder, which shows four strong diffraction peaks at 10.16°, 20.02°, 30.02°, and 50.87°, in good agreement with previous reports. Fig. 2c shows the XRD pattern of PbI$_2$ spin-coated on the FTO/PEDOT:PSS substrate, where four diffraction peaks of (001), (002), (003) and (004) lattice planes can be seen, indicating the preferential crystal growth orientation along the c axis. Unlike the PbI$_2$ deposited on the mesoporous TiO$_2$ layer, there are no additional diffraction peaks observed from the hexagonal 2H polytype of PbI$_2$ orientation. Fig. 2d and 2e illustrate the XRD pattern of CH$_3$NH$_3$PbI$_3$ perovskite synthesized by one-
step and two-step methods, respectively. In comparison with PbI₂ and CH₃NH₃I, a series of new diffraction peaks can be observed, which are in good agreement with the tetragonal phase data of the CH₃NH₃PbI₃ perovskite as reported in the literature. Notably, the intensities of the diffraction peaks of two-step synthesized CH₃NH₃PbI₃ are much stronger than those of one-step synthesized, indicating a high level of phase purity.

To evaluate the charge transfer process, steady-state and time-resolved photoluminescence (PL) characterization methods were performed. As shown in Fig. 3a, the steady-state PL intensity of CH₃NH₃PbI₃ is greatly (>90%) quenched after PCBM deposition, indicating that there is an effective charge transfer from CH₃NH₃PbI₃ to PCBM. Interestingly, while further depositing TIPD on the PCBM, the PL intensity decreased further (inducing more effective PL quench (>95%)), indicating a more efficient charge transfer process between CH₃NH₃PbI₃ and PCBM with the adjacent TIPD layer. We characterized the PL dynamics for a better understanding of the charge transfer in the samples of CH₃NH₃PbI₃, CH₃NH₃PbI₃/PCBM, and CH₃NH₃PbI₃/PCBM/TIPD layers deposited on FTO/PEDOT:PSS substrates. The emission dynamics in such multilayers can be accurately understood with a diffusion limited quenching model. Here, we adopted a phenomenological bi-exponential function \( I(t) = A_1e^{-t/\tau_1} + A_2e^{-t/\tau_2} \) to fit the PL decay in the temporal window of the first 15 ns for a qualitative comparison of charge transfer in these different samples as shown in the Fig. 3b insets.

The time-resolved PL behavior was characterized to probe the influence of the TIPD layer on charge dissociation, as shown in Fig. 3b. The PL lifetimes of CH₃NH₃PbI₃, CH₃NH₃PbI₃/PCBM, and CH₃NH₃PbI₃/PCBM/TIPD layers deposited on FTO/PEDOT:PSS substrates were obtained by fitting the measured PL spectra with a bi-exponential decay function according to a previous report (Fig. 3b insets). The fitted parameters are \( \tau_1 \sim 2.02 \text{ ns (} A_1 \sim 52\%), \quad \tau_2 \sim 6.01 \text{ ns (} A_2 \sim 48\% \) for the samples of CH₃NH₃PbI₃ on FTO/PEDOT: PSS. The average lifetime is slightly slower than that of the pure CH₃NH₃PbI₃ due to the charge transfer between CH₃NH₃PbI₃ and PEDOT:PSS.

When further depositing the PCBM layer on the CH₃NH₃PbI₃ perovskite layer, the PL decay much faster (\( \tau_1 \sim 0.97 \text{ ns (} A_1 \sim 49\%), \quad \tau_2 \sim 4.71 \text{ ns (} A_2 \sim 51\% \)), indicating that there is efficient charge transfer from perovskite to PCBM. Interestingly, when further depositing the TIPD layer on the PCBM layer, a further decrease of the PL lifetime (\( \tau_1 \sim 0.67 \text{ ns (} A_1 \sim 82\%), \quad \tau_2 \sim 3.88 \text{ ns (} A_2 \sim 12\% \)) has been observed, indicating that the additional TIPD layer could induce extremely
fast charge-carrier transfer at the interface, which could suppress the charge recombination and enhance the charge separation and collection in these devices.\textsuperscript{5,18,31} The surface morphology evolution of the multilayered films was investigated by scanning electron microscopy (SEM) and atomic force microscopy (AFM) as shown in Fig. 4. The SEM and AFM images of spin-coated PbI\textsubscript{2} on the PEDOT:PSS/FTO substrate are displayed in Fig. 4a and 4e, respectively. The obtained PbI\textsubscript{2} film shows a root-mean-square (rms) roughness of 8.9 nm with some pinholes (150 nm in diameter) in a scan size of 30 \( \mu \)m by 30 \( \mu \)m. When it reacted with CH\textsubscript{3}NH\textsubscript{3}I vapor, plate-like and pinhole-free CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} perovskite films were formed, as shown in Fig. 4b (SEM) and 4f (AFM). The rms roughness of the two-step synthesized film is much smaller (13.2 nm) than that (38.6 nm) of the one-step solution processed films (Fig. S2†). When PCBM is spin-coated on the perovskite film, the grain boundaries are filled (Fig. 4c) and the surface becomes much smoother (rms = 4.9 nm, Fig. 4g). With TIPD spin-coating, the grain boundaries are further filled (Fig. 4d) and the rms roughness of the film is further reduced to 2.4 nm (Fig. 4h). A similar trend but a more obvious effect can be seen for the one-step synthesized perovskite film as shown in Fig. S2.† As shown in Fig. 4c and 4d, the deposition of TIPD can cause the grain boundaries of the perovskite film to become more shallow, which helps to reduce the pinhole and leakage current. The filling effect of the PCBM/TIPD double layer on perovskite grain boundaries is more obvious for one-step synthesized perovskite, as shown in Fig. S2.† The better filling effect on perovskite pinholes and grain boundaries by the PCBM/TIPD double layer can improve the contact area of perovskite/electron extraction materials and reduce the leakage current, which induces a much higher \( J_{sc} \) and an enhanced \( V_{oc} \).\textsuperscript{30}

The effects of the TIPD electron transporting layer on the photovoltaic performance of perovskite solar cells were examined by constructing devices with and without this layer. To further investigate the suitability of the TIPD layer, one-step and two-step synthesized CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} perovskites were prepared as a photoactive layer. The current density–voltage (\( J-V \)) curves of the devices in the dark and under the illumination of standard one sun are displayed in Fig. 5, and the characteristic parameters (average of 20 individual devices) of open-circuit voltage (\( V_{oc} \)), short-circuit current density (\( J_{sc} \)), filling factor (FF) and PCE are summarized in Table 1. To further clarify the photovoltaic characteristics, a model which is derived from the single heterojunction solar cell was used to analyse the \( J-V \) characteristics of the device. The reverse saturated current density (\( J_0 \)) of the device with TIPD is 1.04 \( \times 10^{-8} \) and 8.66 \( \times 10^{-9} \) A cm\textsuperscript{-2} for one-step and two-step synthesized perovskite, respectively, two orders higher than that (8.80 \( \times 10^{-8} \) and 6.39 \( \times 10^{-7} \) A cm\textsuperscript{-2}) of devices without the TIPD layer (detailed information is given in the ESI†). Fig. 5a compares the \( J-V \) curves of the devices based on one-step synthesized CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} perovskite with and without the TIPD layer. The control device without the TIPD layer shows a PCE of 6.58% with a \( J_{sc} \) of 13.26 mA cm\textsuperscript{-2}, a \( V_{oc} \) of 0.82 V, and an FF of 60.5%. These values of the control device are consistent with previous reports.\textsuperscript{12} In contrast, the device with the TIPD layer shows enhanced \( V_{oc} \), \( J_{sc} \), FF, and the overall PCE, which is 0.85 V, 15.10 mA cm\textsuperscript{-2}, 68.2%, and 8.75%, respectively. The enhanced \( J_{sc} \) was further confirmed by the incident photon-to-electron conversion efficiency (IPCE) measurement as shown in Fig. 5b. A similar trend can also be observed in devices based on two-step synthesized CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} perovskite, indicating that TIPD has good suitability as an electron collection layer to CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3} perovskite with different fabrication processes. The \( J-V \) curves of devices based on two-step synthesized

![Fig. 4](image_url) SEM (a–d) and AFM (e–h) images of PbI\textsubscript{2}, perovskite, perovskite/PCBM, and perovskite/PCBM/TIPD spin-coated on the FTO/PEDOT:PSS substrate, respectively.
CH$_3$NH$_3$PbI$_3$ with different CH$_3$NH$_3$PbI$_3$ and TIPD thicknesses are shown in Fig. S3 and S4,† respectively, and the parameters are summarized in Table S1.†

The best performance was achieved at a thickness of 340 nm for CH$_3$NH$_3$PbI$_3$ and 15 nm for TIPD. As shown in Fig. 5c, the control device without the TIPD layer shows a PCE of 8.66%, with a $V_{oc}$ of 0.838 V, a $J_{sc}$ of 15.42 mA cm$^{-2}$, and an FF of 67.7%. Surprisingly, the $V_{oc}$, $J_{sc}$ and PCE of the device with the TIPD layer are all greatly enhanced to 0.89 V, 22.57 mA cm$^{-2}$ and 12.95%, respectively, among the highest values reported in the literature so far for perovskite/fullerene based solar cells.

The integrated photocurrent density of the devices is calculated by integrating the IPCE spectra with a standard AM 1.5G solar spectrum, using the following equation:

$$J_p = \int_{300}^{800} IPCE(\lambda)E(\lambda)d\lambda$$

where $J_p(\lambda)$ is the current density corresponding to the wavelength ($\lambda$), $E(\lambda)$ is the solar spectral irradiance at a specific wavelength ($\lambda$) and IPCE(\lambda) is the obtained IPCE profile as a function of wavelengths ($\lambda$). For devices based on one-step synthesized CH$_3$NH$_3$PbI$_3$ perovskite, $J_p$ calculated by integrating the IPCE spectra are 12.95 and 14.55 mA cm$^{-2}$ for the devices without and with the TIPD layer, respectively. $J_p$ calculated by
integrating the IPCE curves are 14.63 and 20.94 mA cm\(^{-2}\) for the two-step based devices without and with the TIPD layer, respectively. The slightly lower value of \(J_P\) compared to the measured \(J_{sc}\) may be attributed to the measurement of \(J-V\) that was carried out in a nitrogen-filled glove-box, whereas the IPCE measurement was carried out under ambient conditions, which deteriorate the photovoltaic performance of the devices. Considering these conditions, the \(J_P\) are in good agreement with the measured \(J_{sc}\) from \(J-V\) curves (Fig. 5d).

In order to study the influence of an isopropanol solvent on the perovskite film beneath the PCBM layer, a device with the structure of FTO/PEDOT:PSS/perovskite/PCBM was prepared according to the same procedure as that for the control device C, but the PCBM surface was flushed with isopropanol by spin coating it at 2000 rpm and then baking at 150 °C for 10 min before the deposition of the Al electrode. It was found that the device shows similar photovoltaic performance to the control device C (Fig. S5†), which indicates that the PCBM film is dense enough to prevent the isopropanol solvent from deteriorating the perovskite film.

By comparing the devices with and without the TIPD layer, it is found that no matter one-step or two-step synthesized CH\(_3\)NH\(_3\)PbI\(_3\) perovskite, the increased \(V_{oc}\) should be ascribed to the lower work function of TIPD compared with that of Al, which lowered the contact energy barrier.\(^{18}\) While the great enhancement in PCE for devices with the TIPD layer should be mainly attributed to the enhanced \(J_{sc}\). For excitonic solar cells, the photo induced current is closely related to the light harvesting of the photoactive layer, the exciton dissociation into free electrons and holes (charge transfer process), and the charge collection at both contact electrodes.\(^{12,32,33}\) To check the light absorption of the active layer in our experiment, the reflectance spectra (Fig. S6†) of the devices with and without the TIPD layer were measured according to a previous study.\(^{34}\) The integration reflection intensity of the devices with and without the TIPD layer is 6014 and 6005, respectively, indicating that the additional TIPD layer just slightly enhances the light distribution within the photoactive layer, while this should not be the major reason of the enhanced \(J_{sc}\). The most important reason should be ascribed to the significantly accelerated charge transfer which induced greatly enhanced charge extraction and suppressed charge recombination as shown in Fig. 3.

Conclusions

In conclusion, we successfully demonstrated high performance perovskite solar cells by employing TIPD as an electron transporting layer. The devices with the TIPD layer show an enhanced photocurrent due to the remarkably improved charge extraction and suppressed charge recombination. The TIPD layer shows excellent suitability to CH\(_3\)NH\(_3\)PbI\(_3\) perovskite synthesized by different methods. Under the illumination of AM1.5G 100 mW cm\(^{-2}\), the PCE of the device based on one-step and two-step synthesized CH\(_3\)NH\(_3\)PbI\(_3\) with the TIPD layer reaches 8.37% and 12.95%, respectively, which is a great improvement in comparison with that of control devices without a TIPD layer. Our findings indicate that TIPD is a promising electron transporting layer for the fabrication of CH\(_3\)NH\(_3\)PbI\(_3\) solar cells with high performance.

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Notes and references

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