Supplementary information for

Electric dipole moment-assisted charge extraction and effective defect passivation in PH1000-based perovskite solar cells by incorporating PCBM and TIPD into $CH_3NH_3PbI_3$ layer

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1. First-principles calculations

Electronic structure calculations are performed with the density functional theory as implemented in the Vienna ab initio simulation package,[1,2] employing projected augmented wave potentials to describe the atomic core electrons and a plane wave basis set with a kinetic energy cutoff of 480 eV to expand the Kohn−Sham electronic states. For the exchange and correlation functional, we used the generalized gradient approximation (GGA) in the Perdew–Burke–Ernzerhof (PBE) form.[3] In order to account for interactions between $CH_3NH_3PbI_3$ and TIPD, a periodic slab model was constructed. A 2 × 2 (17.6 Å × 17.6 Å) tetragonal $CH_3NH_3PbI_3$ (001) surface with 5 atomic layers was cut from the optimized bulk geometry in a previous study.[4] The TIPD molecule was adsorbed on one of the surfaces, and a 20 Å vacuum layer was added along the *z* direction. The Brillouin zone sampling was restricted to the Γ point due to the large supercell size, which consists of 259 atoms in total. The DFT-D3 method with Becke-Jonson damping[5] was adopted to include van der Waals interactions. During structural optimization, the three atomic layers of $CH_3NH_3PbI_3$ far away from the TIPD were fixed, and the other two atomic layers of $CH_3NH_3PbI_3$ and the TIPD were relaxed until the residual forces were less than 0.05 eV $Å^{-1}$. Static calculations based on the optimized geometry were performed for the charge density analysis.

In addition, an isolated MA molecule was optimized using the B3LYP[6,7] exchange and correlation

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functional combining with the aug-cc-pVDZ basis set to obtain its electric dipole moment, electronic density and electrostatic potential (ESP). The calculations were performed by Gaussian 09 program.[8]

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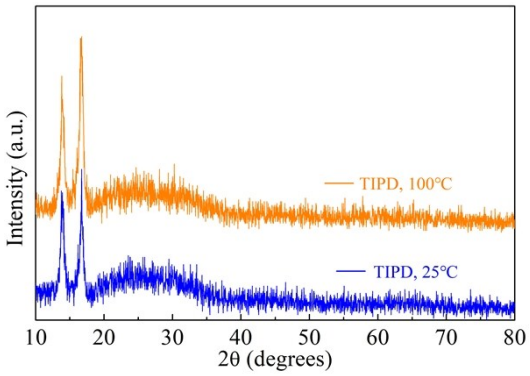

Fig. S1 XRD patterns of the prepared TIPD films dried at 25℃ and 100℃, respectively.

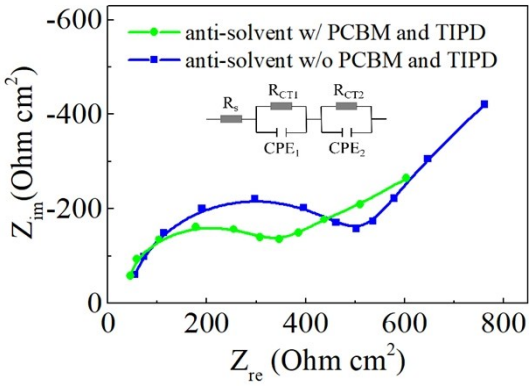

Fig. S2 Electrochemical impedance spectra (EIS) of the PSCs under illumination100 mW/cm^2.

Table S1 The fitting parameters for measured EIS results with different device.

Sample	R_s (Ω·cm^2)	R_{CT1} (mA cm^{-2})	R_{CT2}(mA cm^{-2})	CPE_1 (μF cm^{-2})	CPE_2 (nF cm^{-2})
PSCs with PCBM:TIPD-blended anti-solvent treatment	22.81	774.9	274.4	2.28	6.27
PSCs with CBZ anti-solvent treatment	38.64	1875	452.6	1.56	11.9