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$_3$NH$_3$PbI$_3$ 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$_3$NH$_3$PbI$_3$ and TIPD, a periodic slab model was constructed. A 2 × 2 (17.6 Å × 17.6 Å) tetragonal CH$_3$NH$_3$PbI$_3$ (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$_3$NH$_3$PbI$_3$ far away from the TIPD were fixed, and the other two atomic layers of CH$_3$NH$_3$PbI$_3$ 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

References:

8. Frisch, M. J.; Trucks, G. W.; Schlegel, H. B.; et al., Gaussian 09 (Revision D.01), Gaussian Inc.,
   Wallingford CT, 2013.
Fig. S1 XRD patterns of the prepared TIPD films dried at 25°C and 100°C, respectively.
Fig. S2  Electrochemical impedance spectra (EIS) of the PSCs under illumination 100 mW/cm².

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

<table>
<thead>
<tr>
<th>Sample</th>
<th>$R_s$ (Ωˑcm²)</th>
<th>$R_{CT1}$ (mA cm⁻²)</th>
<th>$R_{CT2}$ (mA cm⁻²)</th>
<th>$CPE_1$ (μF cm⁻²)</th>
<th>$CPE_2$ (nF cm⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PSCs with PCBM:TIPD-blended anti-solvent treatment</td>
<td>22.81</td>
<td>774.9</td>
<td>274.4</td>
<td>2.28</td>
<td>6.27</td>
</tr>
<tr>
<td>PSCs with CBZ anti-solvent treatment</td>
<td>38.64</td>
<td>1875</td>
<td>452.6</td>
<td>1.56</td>
<td>11.9</td>
</tr>
</tbody>
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