

Inorganic perovskite/organic tandem solar cells with efficiency over 20%

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SUPPORTING INFORMATION

Experimental section

Materials characterization

The thicknesses of the films were measured by using a KLA Tencor D-120 profilometer. Absorption spectra for the films were recorded on a Shimadzu UV-1800 spectrophotometer. The SEM images were taken with a Zeiss Merlin field emission SEM (FE-SEM) operated at an accelerating voltage of 5 kV.

Fabrication of single-junction perovskite solar cells

Patterned ITO glass with a sheet resistance of $15 \Omega \text{ sq}^{-1}$ was cleaned by ultrasonic treatment in detergent, deionized water, acetone, isopropanol sequentially and then treated with UV-ozone for 10 min. SnO_2 colloid solution was spin-coated onto ITO glass at 3000 rpm for 30 s and then annealed at 150°C in air for 30 min. ZnO precursor solution (20 mg Zinc acetate dihydrate, $5.6 \mu\text{L}$ ethanolamine, 1 mL dimethoxy ethanol) was spin-coated onto SnO_2 layers at 4000 rpm for 30 s and then annealed at 200°C in air for 20 min. Then the substrates were treated with UV-ozone for 10 min and transferred into a N_2 glovebox. The perovskite precursor solution was prepared by dissolving CsI, PbI_2 , PbBr_2 with a molar ratio of 0.8 : 0.5 : 0.3 in the mixed solvent of DMF/DMSO (4 : 1, v/v). The solution was spin-coated onto the substrates for 35 s, and annealed at 250°C for 10 min. PTAA solution (10 mg/mL in chlorobenzene) was then spin-coated

onto the perovskite layer at 3000 rpm for 30 s. Finally, MoO_3 ($\sim 6 \text{ nm}$) and Ag ($\sim 80 \text{ nm}$) were evaporated onto the PTAA through a shadow mask under vacuum (ca. 10^{-4} Pa).

Fabrication of single-junction organic solar cells

ZnO nanoparticles films were prepared on cleaned ITO-coated glasses by spin coating a ZnO nanoparticles suspension at 3000 rpm for 30s, followed by annealing at 80°C for 5 min. A D18:Y6 blend (1:1.6 w/w) in chloroform (CF) with 0.15% 1-Chlorine naphthalene(CN) additive was spin-coated onto ZnO layer. MoO_3 ($\sim 6 \text{ nm}$) and Ag ($\sim 80 \text{ nm}$) were successively evaporated onto the active layer through a shadow mask under vacuum (ca. 10^{-4} Pa).

Fabrication of tandem solar cells

The preparation of SnO_2 , ZnO, CsPbI_2Br , and PTAA were the same with above. Then MoO_3 ($\sim 4 \text{ nm}$) and Au ($\sim 1 \text{ nm}$) were evaporated onto PTAA sequentially. Then the ZnO nanoparticles, D18:Y6, MoO_3 , and Ag were prepared using the same condition with above.

Device measurements

The effective area for the devices is 4 mm^2 . $J-V$ curves were measured by using a computerized Keithley 2400 Source-Meter and a Xenon-lamp-based solar simulator (Enli Tech, AM 1.5G, 100 mW/cm^2). The illumination intensity of solar simulator was determined by using a monocrystalline silicon solar cell (Enli SRC 2020, $2 \times 2 \text{ cm}^2$) calibrated by NIM. The external quantum efficiency (EQE) spectra were measured by using a QE-R3011 measurement system (Enli Tech).

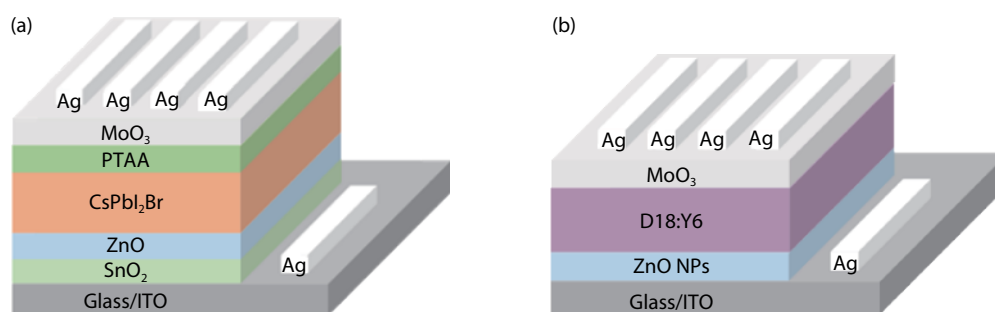


Fig. S1. (Color online) The structures for the (a) single-junction perovskite cells and organic (b) cells.

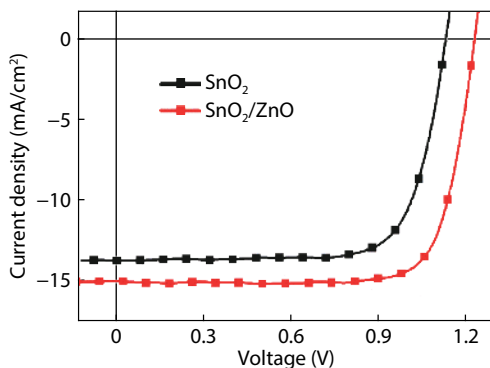


Fig. S2. (Color online) J - V curves for single-junction perovskite solar cells with different electron-transport layer.

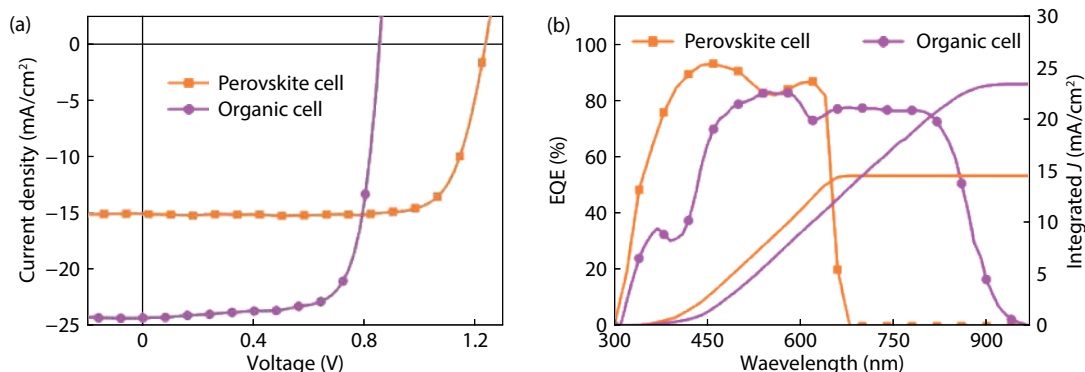


Fig. S3. (Color online) (a) J - V curves for the single-junction perovskite (CsPbI_2Br) cell and organic (D18:Y6) cell; (b) EQE spectra for the cells.

Table 1. Performance data for the single-junction perovskite cell, single-junction organic cell, and tandem cell.

	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE ^a (%)
Perovskite cell	1.23	15.03	78.2	14.48 (14.13)
Organic cell	0.85	24.22	73.7	15.23 (14.95)
Tandem cell	2.05	13.07	75.3	20.18 (19.95)

^aData in parentheses stand for the average PCEs for 10 cells.

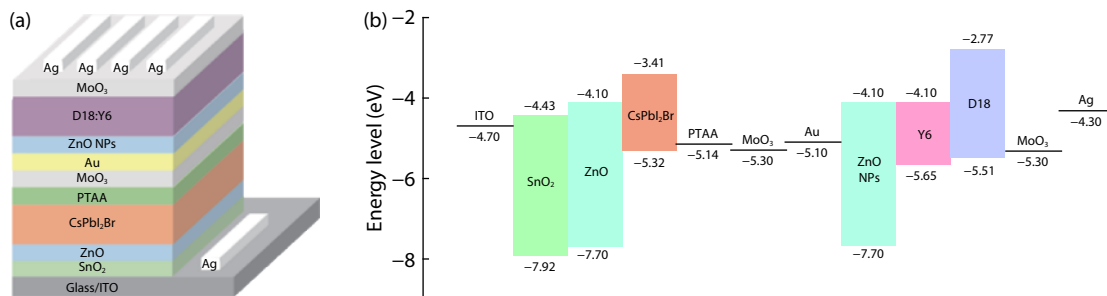
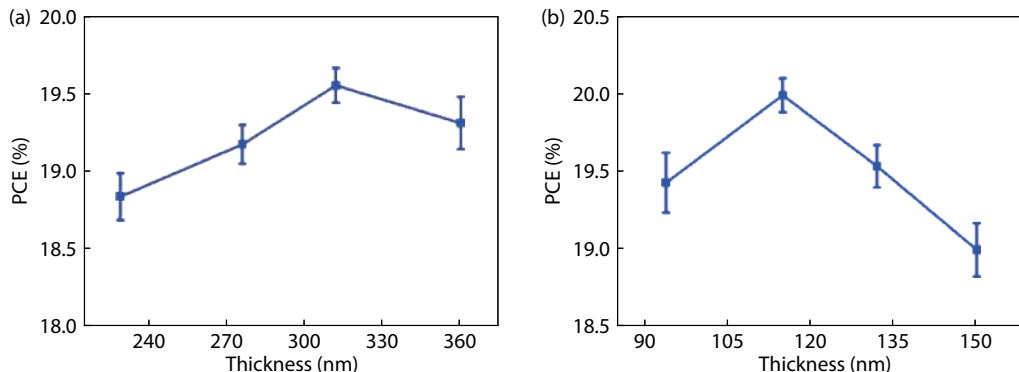
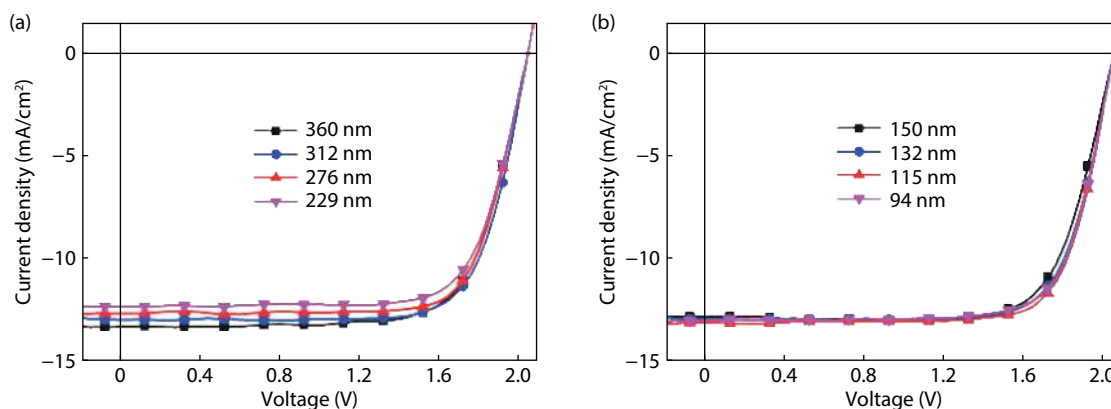


Fig. S4. (Color online) (a) Structure of the tandem cell. (b) Energy level diagram.

Table 2. Performance of tandem cells changing with the thickness of the CsPbI₂Br layer (thickness of D18:Y6 layer: 132 nm).

Thickness (nm)	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE ^a (%)
229	2.05	12.33	75.3	19.01 (18.84)
276	2.05	12.60	75.1	19.36 (19.17)
312	2.05	12.91	74.6	19.72 (19.55)
360	2.04	13.27	72.1	19.57 (19.31)

^aData in parentheses stand for the average PCEs for 10 cells.Fig. S5. (a) PCE for tandem cells changing with the thickness of the CsPbI₂Br layer (the thickness of D18:Y6 layer: 132 nm). (b) PCE for tandem cells changing with the thickness of the D18:Y6 layer (the thickness of CsPbI₂Br layer: 312 nm).Fig. S6. (Color online) (a) J - V curves for the tandem cells changing with the thickness of the CsPbI₂Br layer (thickness of D18:Y6 layer: 132 nm). (b) J - V curves for the tandem cells changing with the thickness of the D18:Y6 layer (thickness of CsPbI₂Br layer: 312 nm).Table 3. Performance of tandem cells changing with the thickness of the D18:Y6 layer (thickness of CsPbI₂Br layer: 312 nm).

Thickness (nm)	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE ^a (%)
94	2.05	12.97	74.4	19.78 (19.43)
115	2.05	13.07	75.3	20.18 (19.99)
132	2.05	12.91	74.6	19.72 (19.55)
150	2.05	12.90	73.7	19.28 (18.99)

^aData in parentheses stand for the average PCEs for 10 cells.

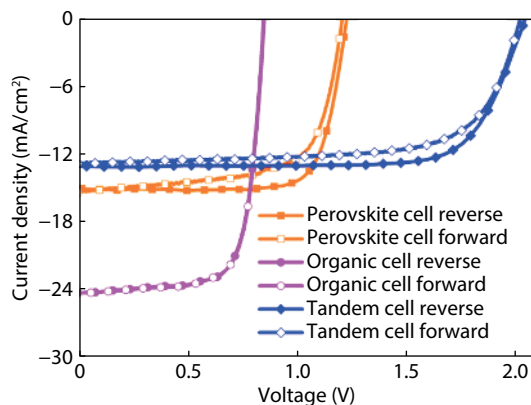


Fig. S7. (Color online) J - V curves for the single-junction perovskite cell, single-junction organic cell, and tandem cell under forward and reverse scans.

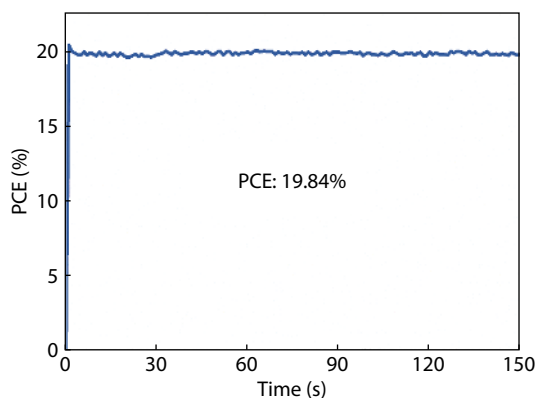


Fig. S8. Steady-state PCE for the best tandem cell.

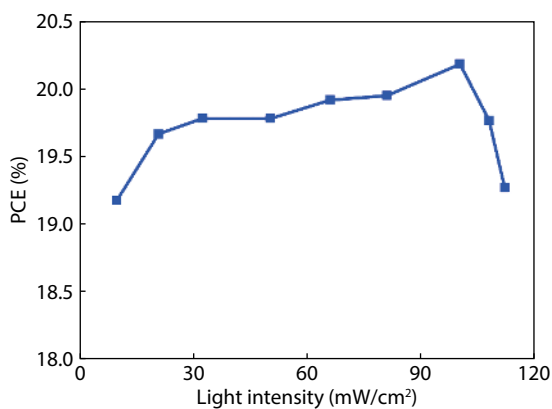


Fig. S9. PCE for the tandem cell under different light intensities.

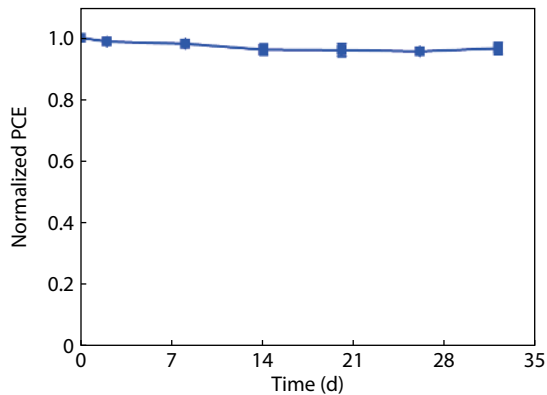


Fig. S10. PCE change for tandem cells stored in N_2 glovebox.