Electronic supplementary information

Lattice-Tailored Low-Temperature Processed Electron Transporting Materials Boost the Open-Circuit Voltage of Planar $CsPbBr_3$ Solar Cells up to 1.654 V

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Table S1 Summary of the photovoltaic parameters of single-junction solar cells with high V_{oc}.

Device structure	J_{sc} (mA cm^{-2})	V_{oc} (V)	FF (%)	PCE (%)	Ref.
FTO/c-TiO_2/$CsPbBr_3$/spiro-MeOTAD/Au	5.65	1.536	62.4	5.42	S1
ITO/NiO/$CH_3NH_3PbBr_3$/ICBA/PrC60MA/Ag	5.13	1.50	69.5	5.35	S2
ITO/PEDOT:PSS/$CH_3NH_3PbBr_3$/ICBA/Ca/Al	6.04	1.61	77	7.50	S3
FTO/c-TiO_2/m-TiO_2/GQD/$CsPbBr_3$/Carbon	8.12	1.458	82.1	9.72	S4
FTO/c-TiO_2/m-TiO_2/$CsPb_{0.97}Sm_{0.03}Br_3$/Carbon	7.48	1.594	85.1	10.14	S5
FTO/c-TiO_2/m-TiO_2/$CsPb_{0.97}Sm_{0.03}Br_3$/Cu(Cr,Ba)O_2/Carbon	7.81	1.615	85.5	10.79	S6
FTO/c-TiO_2/m-TiO_2/$CsPbBr_3$/MnS/Carbon	8.28	1.52	83	10.45	S7
FTO/c-TiO_2/$CsPbBr_3$/spiro-MeOTAD/Ag	9.78	1.498	74.47	10.91	S8
FTO/c-TiO_2/$CsPbBr_3$/Carbon	7.37	1.545	82.2	9.35	S9
FTO/c-TiO_2/m-TiO_2/$CsPbBr_3$/$CuInS_2$/ZnS QDs/LPP-Carbon	7.73	1.626	86.3	10.85	S10
FTO/SnO_2/TiO_xCl_{4-2x}/$Cs_{0.91}Rb_{0.09}PbBr_3$/Carbon	7.96	1.629	80.5	10.44	S11

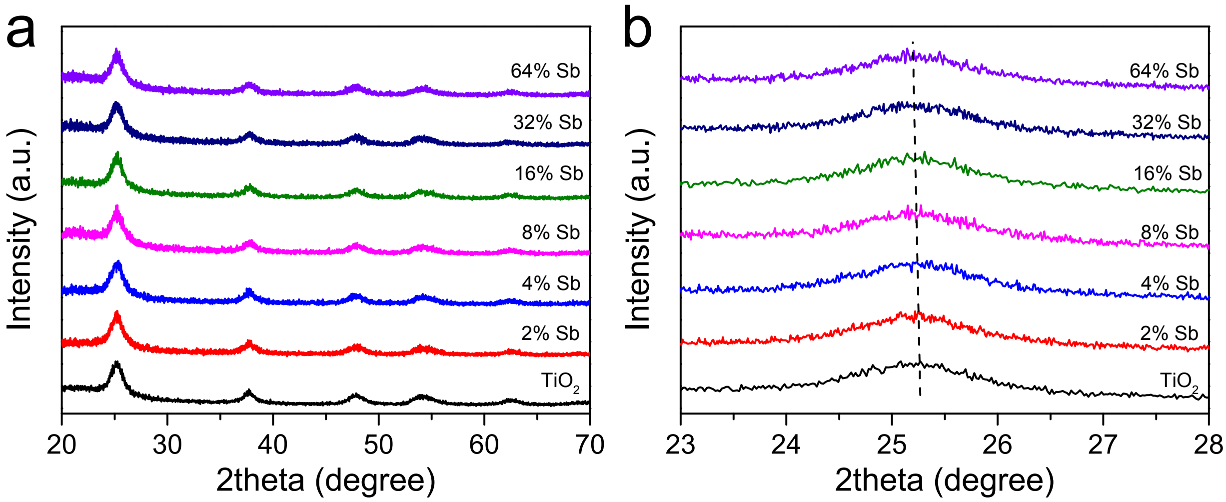

Fig. S1 XRD patterns of Sb-doped TiO_2 nanocrystals with different Sb dosages in precursor solution.

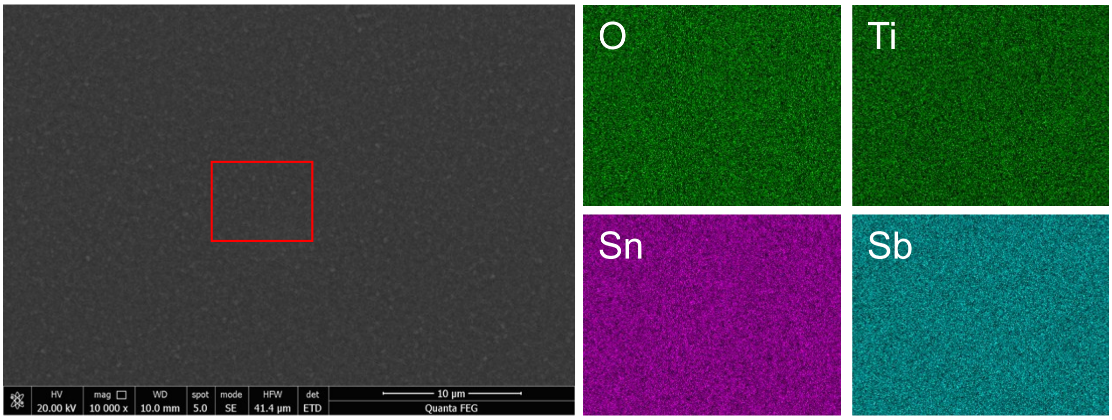

Fig. S2 Top-view image of the Sb-TiO_2 film deposited on FTO substrate and the corresponding elemental mapping images.

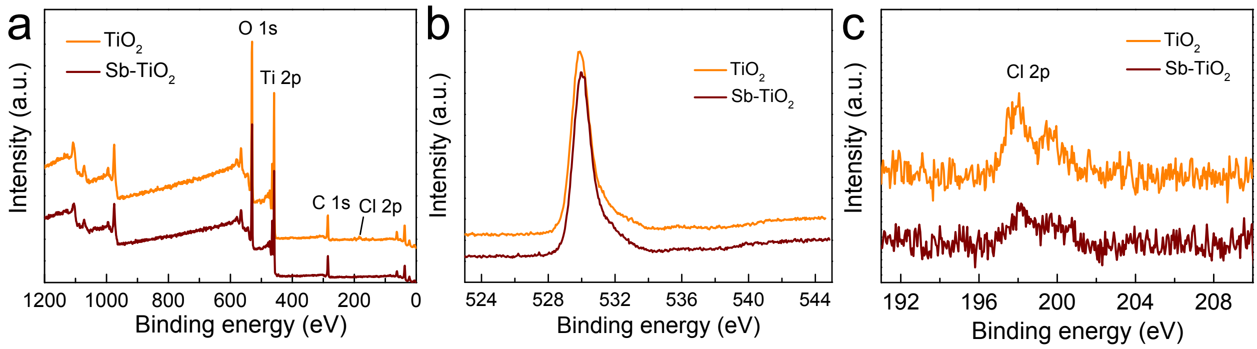

Fig. S3 (a) XPS spectra of TiO_2 films with and without Sb dopant. High-resolution XPS spectra of (b) Sb 3d and (c) Cl 2p core level.

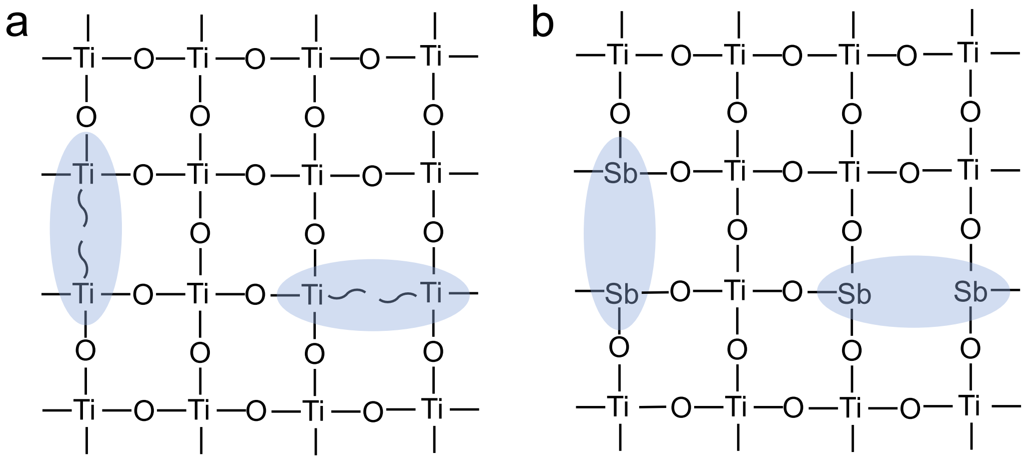

Fig. S4 (a) Defects in the pristine TiO_2, (b) Sb^{3+} substitution at the Ti^{3+} sites passivates oxygen vacancy defects.

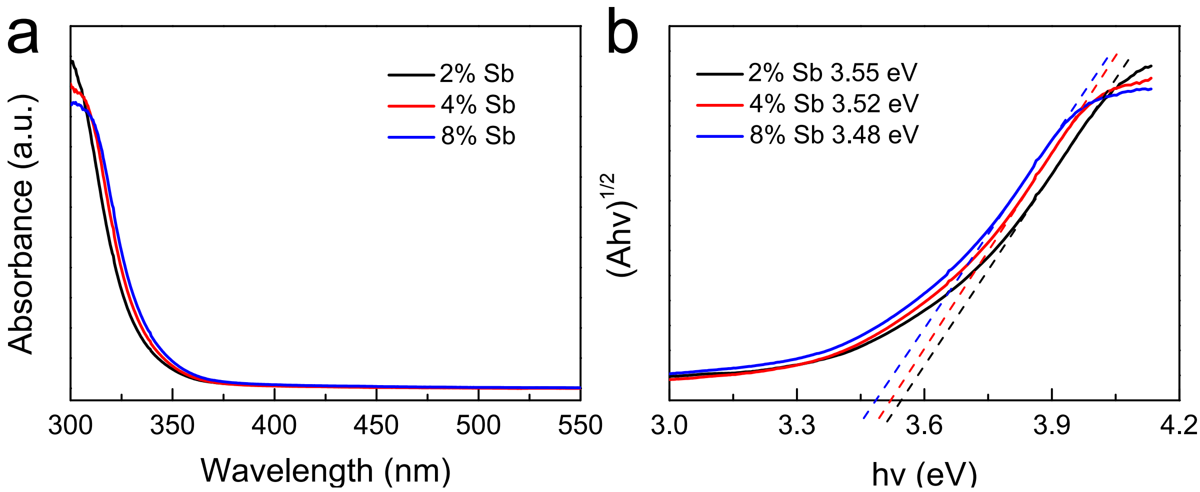

Fig. S5 UV-vis absorption spectra and corresponding bandgaps of Sb-doped TiO_2 films with increased Sb dosages in precursor solution.

Table S2 Electrical parameters of the TiO_2 and Sb-TiO_2 films

ETL	σ (mS cm^{-1})	μ (cm^2 V^{-1} s^{-1})	n_t (cm^{-3})
TiO_2	1.19×10^{-3}	5.44×10^{-4}	3.15×10^{17}
Sb-TiO_2	1.99×10^{-3}	6.58×10^{-4}	2.67×10^{17}

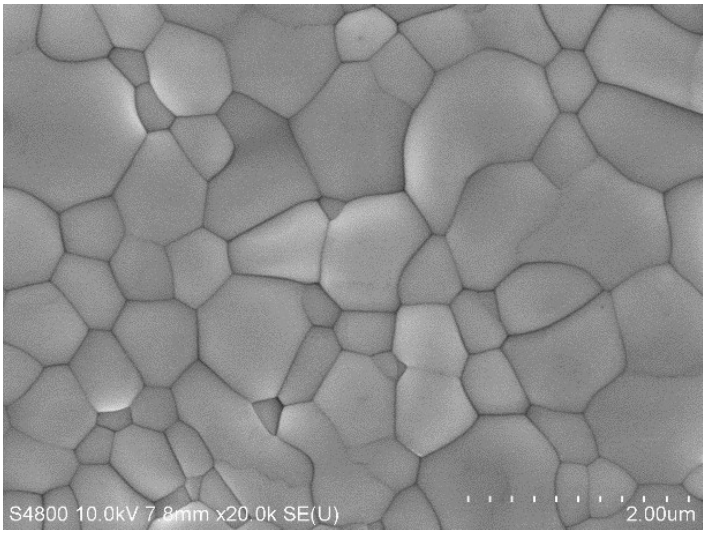

Fig. S6 Top-view SEM image of $CsPbBr_3$ film deposited on traditional mesoporous TiO_2 substrate.

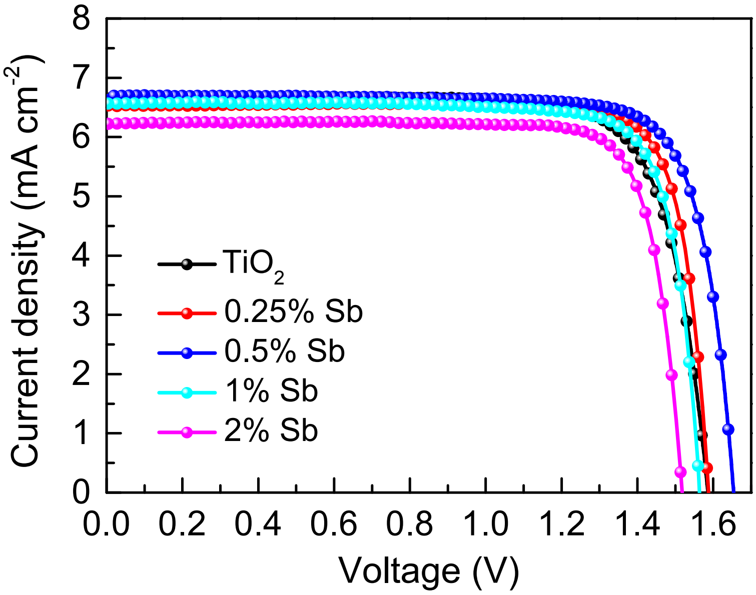

Fig. S7 *J*-*V* curves of $CsPbBr_3$ solar cells based on TiO_2 and Sb-doped TiO_2 with different doping amounts.

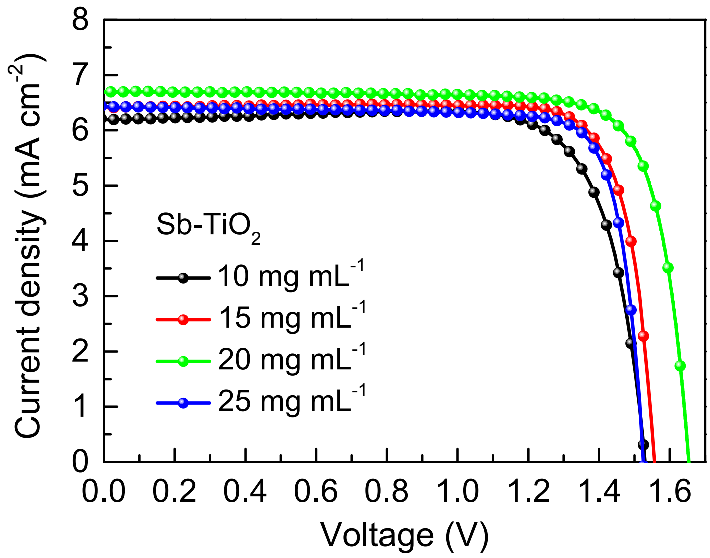

Fig. S8 *J-V* curves of $CsPbBr_3$ solar cells based on Sb-TiO_2 with different Sb-TiO_2 dispersion concentrations.

Table S3 Photovoltaic parameters of inorganic $CsPbBr_3$ PSCs based on TiO_2 and Sb-doped TiO_2 ETLs.

device	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
TiO_2	1.586	6.64	78.4	8.26
0.25% Sb	1.584	6.52	83.4	8.62
0.5% Sb	1.654	6.70	80.4	8.91
1% Sb	1.560	6.56	81.5	8.35
2% Sb	1.515	6.23	82.3	7.77

Table S4 Photovoltaic parameters of inorganic $CsPbBr_3$ PSCs based on Sb-TiO_2 ETLs at different Sb-TiO_2 dispersion concentrations.

Concentration	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
10 mg mL^{-1}	1.526	6.18	79.1	7.48
15 mg mL^{-1}	1.554	6.41	82.4	8.22
20 mg mL^{-1}	1.654	6.70	80.4	8.91
25 mg mL^{-1}	1.522	6.42	82.1	8.03

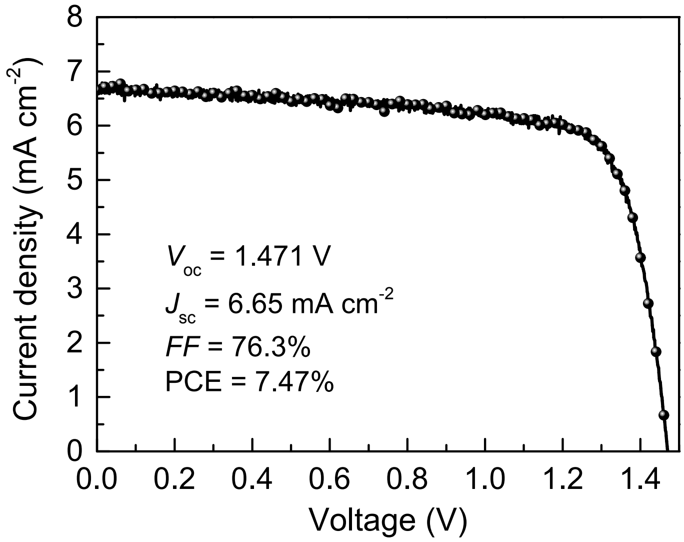

Fig. S9 *J-V* curve of inorganic $CsPbBr_3$ PSC based on the high-temperature processed TiO_2 ETL.

Table S5 Photovoltaic parameters of inorganic $CsPbBr_3$ PSCs based on TiO_2 and Sb doped TiO_2 with V_{oc} over 1.6 V.

device	V_{oc} (V)	J_{sc} (mA cm^{-2})	*FF* (%)	PCE (%)
TiO_2-1	1.606	6.37	76.5	7.83
TiO_2-2	1.607	6.49	77.0	8.02
TiO_2-3	1.608	5.90	83.5	7.92
0.25% Sb-1	1.619	6.52	80.3	8.47
0.25% Sb-2	1.609	6.22	80.1	8.02
0.25% Sb-3	1.630	6.22	75.8	7.69
0.5% Sb-1	1.652	6.44	78.4	8.34
0.5% Sb-2	1.629	6.47	75.3	7.94
0.5% Sb-3	1.654	6.70	80.4	8.91
0.5% Sb-4	1.616	6.46	79.5	8.29
0.5% Sb-5	1.604	6.25	81.4	8.17
0.5% Sb-6	1.606	6.20	85.4	8.50
0.5% Sb-7	1.608	5.92	80.7	7.69
0.5% Sb-8	1.617	6.55	83.6	8.85
0.5% Sb-9	1.624	6.56	80.0	8.52
0.5% Sb-10	1.603	6.34	79.5	8.07
0.5% Sb-11	1.611	5.64	84.4	7.67
0.5% Sb-12	1.603	5.66	85.0	7.71

0.5% Sb-13	1.604	6.34	82.5	8.39

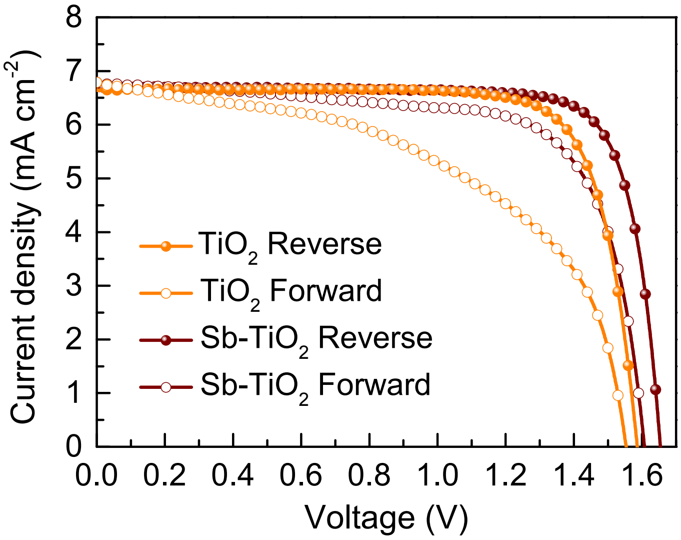

Fig. S10 *J-V* curves of the optimized inorganic $CsPbBr_3$ PSCs based on TiO_2 and Sb-TiO_2 under forward and reverse scan directions.

Table S6 Photovoltaic parameters of optimized inorganic $CsPbBr_3$ PSCs based on TiO_2 and Sb-TiO_2 under forward and reverse scan directions.

device	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
TiO_2 Reverse	1.586	6.64	78.4	8.26
TiO_2 Forward	1.553	6.79	51.8	5.46
Sb-TiO_2 Reverse	1.654	6.70	80.4	8.91
Sb-TiO_2 Forward	1.607	6.79	70.4	7.68

Table S7 TRPL decay lifetimes of $CsPbBr_3$ films on TiO_2 and Sb-TiO_2 coated FTO substrates. τ_1 and τ_2 correspond to the slow and fast decay components, respectively.

Samples	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)	τ_{ave} (ns)
$TiO_2/CsPbBr_3$	0.88	58.5	10.53	41.5	1.42
Sb-$TiO_2/CsPbBr_3$	0.49	67.6	8.66	32.4	0.71

References

S1. Q. A. Akkerman, M. Gandini, F. Di Stasio, P. Rastogi, F. Palazon, G. Bertoni, J. M. Ball, M. Prato, A. Petrozza and L. Manna, *Nat. Energy*, 2016, **2**, 16194.

S2. S. Chen, Y. Hou, H. Chen, M. Richter, F. Guo, S. Kahmann, X. Tang, T. Stubhan, H. Zhang, N. Li, N. Gasparini, C. O. R. Quiroz, L. S. Khanzada, G. J. Matt, A. Osvet and C. J. Brabec, *Adv. Energy Mater.*, 2016, **6**, 1600132.

S3. C. G. Wu, C. H. Chiang and S. H. Chang, *Nanoscale*, 2016, **8**, 4077.

S4. J. Duan, Y. Zhao, B. He and Q. Tang, *Angew. Chem. Int. Ed.*, 2018, **57**, 3787.

S5. J. Duan, Y. Zhao, X. Yang, Y. Wang, B. He and Q. Tang, *Adv. Energy Mater.*, 2018, **8**, 1802346.

S6. J. Duan, Y. Zhao, Y. Wang, X. Yang and Q. Tang, *Angew. Chem. Int. Ed.*, 2019, **58**, 16147.

S7. X. Li, Y. Tan, H. Lai, S. Li, Y. Chen, S. Li, P. Xu and J. Yang, *ACS Appl. Mater. Interfaces*, 2019, **11**, 29746.

S8. G. Tong, T. Chen, H. Li, L. Qiu, Z. Liu, Y. Dang, W. Song, L. K. Ono, Y. Jiang and Y. Qi, *Nano Energy*, 2019, **65**, 104015.

S9. T. Xiang, Y. Zhang, H. Wu, J. Li, L. Yang, K. Wang, J. Xia, Z. Deng, J. Xiao, W. Li, Z. Ku, F. Huang, J. Zhong, Y. Peng and Y.-B. Cheng, *Sol. Energy Mater. Sol. Cells*, 2019, **206**, 110317.

S10. J. Duan, Y. Wang, X. Yang and Q. Tang, *Angew. Chem. Int. Ed.*, 2020, **59**, 4391.

S11. Q. Zhou, J. Duan, Y. Wang, X. Yang and Q. Tang, *J. Energy Chemistry*, 2020, **50**, 1.