Supporting Information for

Antimony Potassium Tartrate Stabilizes Wide-Bandgap Perovskites

for Inverted 4T All-Perovskite Tandem Solar Cells with Efficiencies

over 26%

Xuzhi Hu¹, Jiashuai Li¹, Chen Wang¹, Hongsen Cui¹, Yongjie Liu¹, Shun Zhou¹, Hongling Guan¹, Weijun Ke^{1, *}, Chen Tao^{1, *} and Guojia Fang^{1, *}

¹ Key Laboratory of Artificial Micro/Nano Structures of Ministry of Education,

School of Physics and Technology, Wuhan University, Wuhan 430072, P. R. China

*Corresponding authors. E-mail: <u>weijun.ke@whu.edu.cn</u> (Weijun Ke); <u>taochen635@whu.edu.cn</u> (Chen Tao); <u>gjfang@whu.edu.cn</u> (Guojia Fang)

Supplementary Figures and Tables



Fig. S1 AFM morphology and corresponding RMS roughness of perovskite **a** without and **b** with APTA additive



Fig. S2 PL peak changes of perovskite film \mathbf{a} without and \mathbf{b} with APTA under continuous laser irradiation

samples	$\tau_1(ns)$	A ₁	$\tau_2(ns)$	A ₂	$\tau_{avg}(ns)$
Control	30.23	33.88	254.62	66.12	241.75
APTA	65.14	23.20	852.44	76.80	834.46

Table S1 Fitted Parameters of TRPL Spectra



Fig. S3 Cross-sectional SEM image of PSC a without and b with APTA additive



Fig. S4 The statistic performance parameters of a V_{OC} , b J_{SC} , c FF and d PCE for inverted WBG PSCs with different concentration of APTA



Fig. S5 J-V curves of the champion PSCs prepared **a** without or **b** with APTA measured in both reverse and forward directions, respectively. The photovoltaic parameters of the devices are both shown in each picture



Fig. S6 The first derivative of the EQE curve



Fig. S7 Statistical data of a $V_{\rm OC}$, b $J_{\rm SC}$ and c FF for WBG devices

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sample	V _{oc} (V)	$J_{ m SC}$ (mA cm ⁻²)	FF (%)	PCE (%)	
control	1.09±0.034	20.7±0.16	82.4±0.5	19.02±0.32	
APTA	1.13±0.023	20.8±0.15	84.3±0.3	20.12±0.33	

Table S2 Standard deviations of the photovoltaic parameters of the WBG devices



Fig. S8 a I 3d and b Br 3d XPS spectra for perovskite film w/o and with APTA



Fig. S9 UV-vis absorbance spectra of perovskite films w/o and with APTA



Fig. S10 UPS spectra of perovskite films w/o or with APTA

samples	Eg (eV)	$E_{\text{onset}}(\text{eV})$	$E_{\rm cutoff}(eV)$	$E_{\rm VBM}({ m eV})$	$E_{\rm CBM}({ m eV})$	$E_{\rm F}$ (eV)
Control	1.67	1.41	17.09	5.53	3.86	4.12
APTA	1.67	1.49	17.01	5.69	4.02	4.20

Table S3 Energy level parameters of different perovskites



Fig. S11 The derivative of capacitance spectra of WBG devices **a** without and **b** with APTA incorporation

samples	$Rs(\Omega)$	R rec (Ω)
Control	70.1	491.2
APTA	49.8	1274.5

Table S4 EIS Parameters for the PSCs based on different perovskite



Fig. S12 Schematic device configuration of Sn-Pb NBG perovskite solar cell



Fig. S13 Transmittance spectrum of semi-transparent perovskite top cell



Fig. S14 J-V curves of the semi-transparent WBG PSC a without and b with APTA

year	subcells	Device structure	PCE of 4-T tandem solar cells (%)	REFs
2016	top	ITO/NiO _x /MAPbI ₃ /PCBM/C60/ITO	19.08	[S1]
	bottom	ITO/PEDOT:PSS/MA _{0.5} FA _{0.5} Sn _{0.25} Pb _{0.75} I ₃ /PCBM/C60/Ag		
2016	top	$ITO/NiO_x/FA_{0.83}Cs_{0.17}Pb_{I0.83}Br_{0.17}/PCBM/SnO_2/ZTO/ITO$	20.1	[S2]
	bottom	$ITO/PEDOT:PSS/FA_{0.75}Cs_{0.25}Sn_{0.5}Pb_{0.5}I_3/PCBM/BCP/Ag$	20.1	
2017	top	FTO/SnO ₂ /C60-SAM/MA _{0.7} FA _{0.3} PbI ₃ /spiro-OMeTAD/MoO _x /Au/MoO _x		[S3]
	bottom	ITO/PEDOT:PSS/(FASnI ₃) _{0.6} (MAPbI ₃) _{0.4} /PCBM/BCP/Ag	21.2	
2017	top	ITO/NiOx/MA _{0.9} Cs _{0.1} Pb(I _{0.6} Br _{0.4}) ₃ /C60/ C60/ITO		[S4]
	bottom	ITO/PEDOT:PSS/MASn_0.5Pb_0.5I_3/IC_60BA/C60/Ag	16.7	
2018	top	FTO/SnO ₂ /C ₆₀ -SAM/FA _{0.8} Cs _{0.2} Pb(I _{0.7} Br _{0.3}) ₃ /spiro-OMeTAD/MoOx/ITO		[85]
	bottom	ITO/PEDOT:PSS/(FASnI3)0.6(MAPbI3)0.4/PCBM/BCP/Ag	23.1	
2020	top	$ITO/SnO_{2}/Cs_{0.1}(MA_{0.17}FA_{0.83})_{0.9}Pb(I_{0.83}Br_{0.17})_{3}/spiro-OMeTAD/MoO_{x}/ITO$	23.0	[S6]
2020 t	bottom	ITO/PEDOT:PSS/FA0.8MA0.2Sn0.5Pb0.5I3/PCBM/C60/BCP/Ag.		[~~]
2022	top	$ITO/MeO\text{-}2PACz/FA_{0.8}Cs_{0.2}PbI_{0.8}Br_{0.2}/C_{60}/SnO_2/ITO$	26.0	[07]
	bottom	ITO/PEDOT:PSS/FA0.7MA0.3Sn0.5Pb0.5I3/C60/BCP/Ag.	26.0	[8/]
2023	top	$ITO/MeO-2PACz/FA_{0.75}Cs_{0.25}PbI_{0.8}Br_{0.2}/C_{60}/SnO_2/ITO$	26.3	This work
	bottom	ITO/PEDOT:PSS/FA0.7MA0.3Sn0.5Pb0.5I3/C60/BCP/Ag.		

 Table S5 Summary of all-perovskite 4-T tandem solar cells

Supplementary References

- [S1]Z. Yang, A. Rajagopal, C. Chueh, S. B. Jo, B. Liu et al., Stable Low-bandgap Pb-Sn binary perovskites for tandem solar cells. Adv. Mater. 28, 8990-8997 (2016). <u>https://doi.org/10.1002/adma.201602696</u>
- [S2] G. E. Eperon, T. Leijtens, K.A. Bush, R. Prasanna, T. Green et al., Perovskiteperovskite tandem photovoltaics with optimized band gaps. Science 354, 861 (2016). <u>https://doi.org/10.1126/science.aaf9717</u>
- [S3] D. Zhao, Y. Yu, C. Wang, W. Liao, N. Shrestha et al., Low-bandgap mixed tinlead iodide perovskite absorbers with long carrier lifetimes for all-perovskite tandem solar cells. Nat. Energy 2, 17018 (2017). <u>https://doi.org/10.1038/nenergy.2017.18</u>
- [S4] A. Rajagopal, Z. Yang, S. B. Jo, I. L. Braly, P. W. Liang et al., Highly efficient perovskite-perovskite tandem solar cells reaching 80% of the theoretical limit in photovoltage. Adv. Mater. 29, 1702140 (2017). https://doi.org/10.1002/adma.201702140
- [S5] D. Zhao, C. Wang, Z. Song, Y. Yu, C. Chen et al., Four-terminal all-perovskite tandem solar cells achieving power conversion efficiencies exceeding 23%. ACS Energy Lett. 3, 305 (2018). <u>https://doi.org/10.1021/acsenergylett.7b01287</u>
- [S6] B. A. Nejand, I. M. Hossain, M. Jakoby, S. Moghadamzadeh, T. Abziehe et al., Vacuum-assisted growth of low-bandgap thin films (FA_{0.8}MA_{0.2}Sn_{0.5}Pb_{0.5}I₃) for all-perovskite tandem solar cells. Adv. Energy Mater. **10**, 1902583 (2020). <u>https://doi.org/10.1002/aenm.201902583</u>
- [S7] W. Zhang, L. Huang, W. Zheng, S. Zhou, X. Hu, J. Zhou, J.Li, J, Liang, W. Ke, G. Fang. Revealing key factors of efficient narrow-bandgap mixed lead-tin perovskite solar cells via numerical simulations and experiments. Nano Energy 96, 107078 (2022). <u>https://doi.org/10.1016/j.nanoen.2022.107078</u>