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Highly vertically oriented 2D/3D tin perovskite films for efficient and stable solar cells

Ziyong Kang[1,†,*], Mingyu Ma[2,†], Yibo Han[1], Fen Zhang[3], Lu Zhang[4,*], Jingyuan Qiao[5], Zhixin Sheng[1], Peng Feng[5,*] and Hongqiang Wang[1,*]

[1] State Key Laboratory of Solidification Processing, Center for Nano Energy Materials, School of Materials Science and Engineering, Northwestern Polytechnical University, Xi'an, China
[2] School of Physical Science and Technology, ShanghaiTech University, Shanghai, China
[3] Shaanxi Materials Analysis and Research Centre, Northwestern Polytechnical University, Xi'an, China.
[4] Key Laboratory of Applied Surface and Colloid Chemistry Ministry of Education, Shaanxi Key Laboratory for Advanced Energy Devices, Shaanxi, Engineering Lab for Advanced Energy Technology, School of Materials Science Engineering Shaanxi Normal University, Xi'an, China
[5] Frontier Science Center for Flexible Electronics, Xi'an Institute of Flexible Electronics, Northwestern Polytechnical University, Xi'an, China

† These authors contributed equally to this work.

* Correspondence authors; E-mails: 2022100567@mail.nwpu.edu.cn (Z.K.); luzhang@snnu.edu.cn (L.Z.); feng@nwpu.edu.cn (P.F.); hongqiang.wang@nwpu.edu.cn (H.W.).

1. Computational methods

1.1. Electrical conductivity calculation

The conductivity of perovskite film from the current-voltage (*I-V*) characteristic curves was calculated by Equation (1)

$$\sigma = \frac{I}{V} \times \frac{d}{S} \tag{1}$$

where $\frac{I}{V}$ is the slope of lines in conductivity, d and S are the thickness and area of perovskite film.

1.2. Hermans' orientation factor (HF) calculation

The Hermans orientation factor *H*(100) of the (100) can be calculated using Equation (2) and Equation (3).

$$< \cos^2\varphi >= \frac{\int_0^{\frac{\pi}{2}} I(\varphi)\cos^2\varphi \ d\varphi}{\int_0^{\frac{\pi}{2}} I(\varphi) \sin\varphi \ d\varphi} \tag{2}$$

$$H(100) = \frac{3\cos^2\varphi - 1}{2} \quad (3)$$

where $I(\varphi)$ is the scattering intensity along the (100) ring at the angle ϕ with respect to the reference axis. HF equals 1 indicating that the normal of (100) planes is perfectly along the reference axis and equals 0 suggests that (100) planes are randomly oriented, which will display discrete Bragg spots and uniform rings on GIWAXS patterns, respectively.

1.3. Steady-state decay lifetime calculation

The exciton lifetime of the perovskite films can be derived from the femtosecond transient absorption spectroscopy (TAS) by a bi-exponential fitting as shown in Equation (4).

$$y = A_1 e^{\left(-\frac{\tau}{\tau_1}\right)} + A_2 e^{\left(-\frac{\tau}{\tau_2}\right)} \quad (4)$$

where τ is the exciton lifetime, τ_1 and τ_2 are the decay components of the trap-assisted and radiative recombination process respectively. The carrier recombination lifetime can be calculated by Equation (5).

$$\tau = \frac{A_1\tau_1^2 + A_2\tau_2^2}{A_1\tau_1 + A_2\tau_2} \quad (5)$$

2. Analyzation of space-charge-limited current (SCLC) model

The trap density (N_t) of perovskite films from the dark *J-V* curves of the electron-only and hole-only devices can be calculated by Equation (6).

$$N_t = \frac{2\varepsilon\varepsilon_0 V_{TFL}}{qd^2} \quad (6)$$

where q is the electron charge, d is the thickness of perovskite film, ε_0 and ε are the vacuum permittivity and relative dielectric constant of perovskite, and V_{TFL} is ordinate of the intersection of ohmic region and trap-filling limited region.

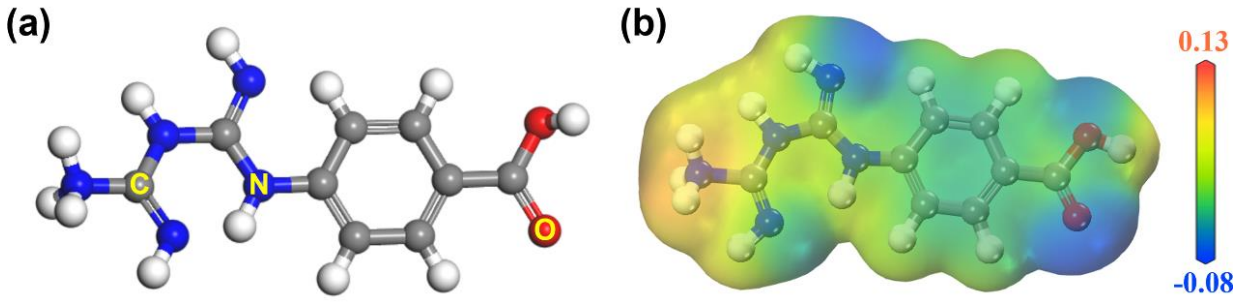

Figure S1. Molecular structure and electrostatic potential diagrams of p-BGBA.

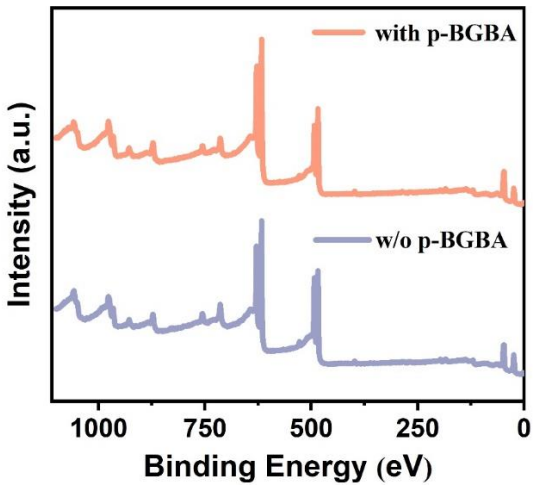

Figure S2. Full XPS spectra of perovskite films without and with p-BGBA.

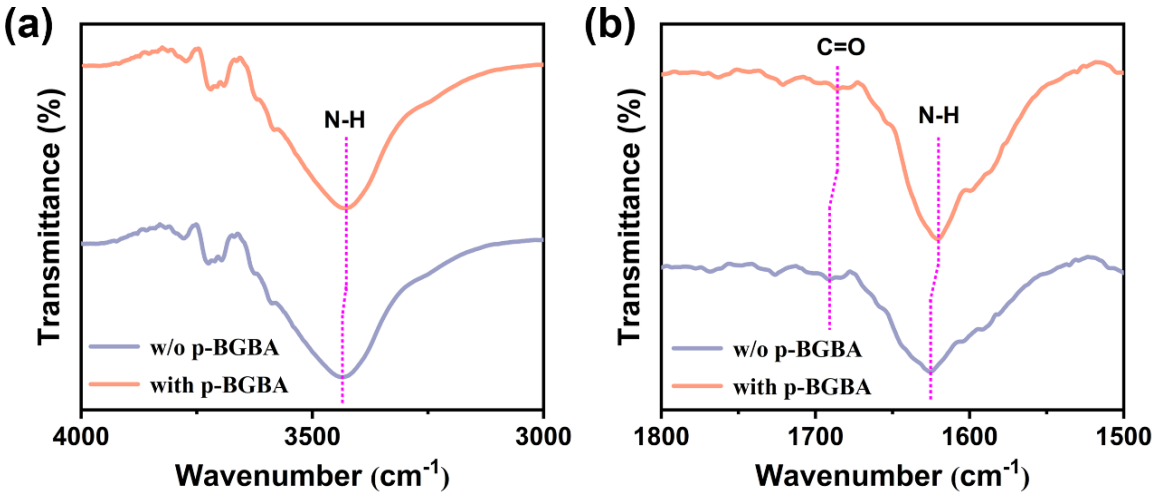

Figure S3. FTIR spectra of the perovskite films without and with p-BGBA. **(a)** N-H stretching region and **(b)** C = O stretching and N-H bending vibration regions.

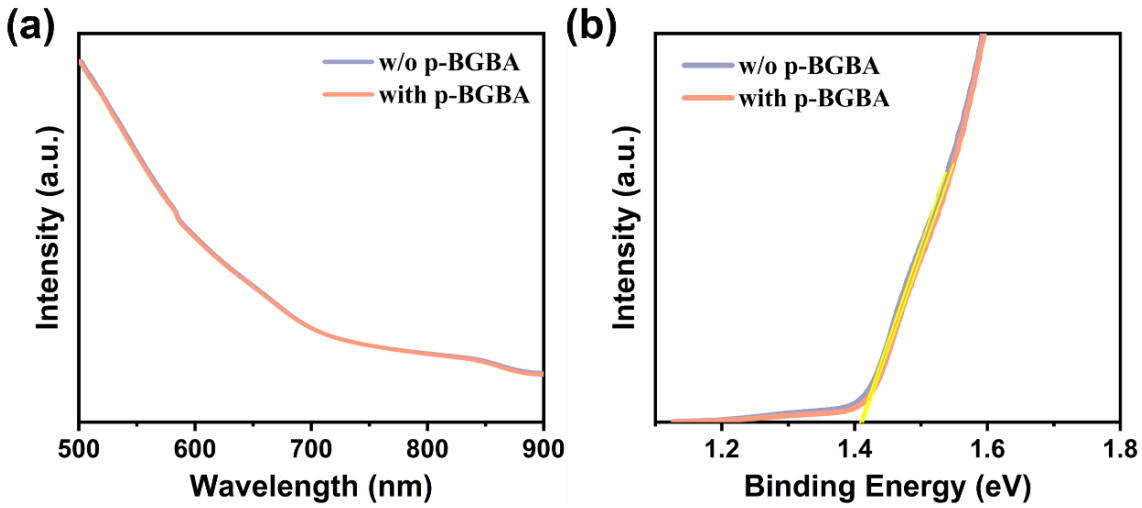

Figure S4. Optical properties of p-BGBA modified perovskite films. **(a)** UV-vis absorption spectra, and **(b)** Tauc-plot of perovskite films without and with p-BGBA.

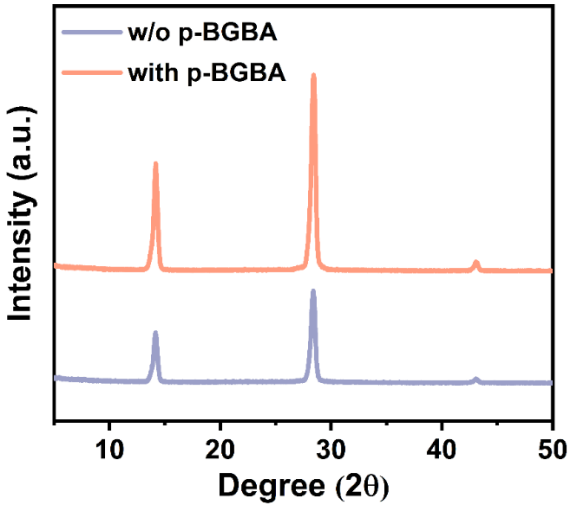

Figure S5. XRD patterns for tin perovskite films without and with p-BGBA.

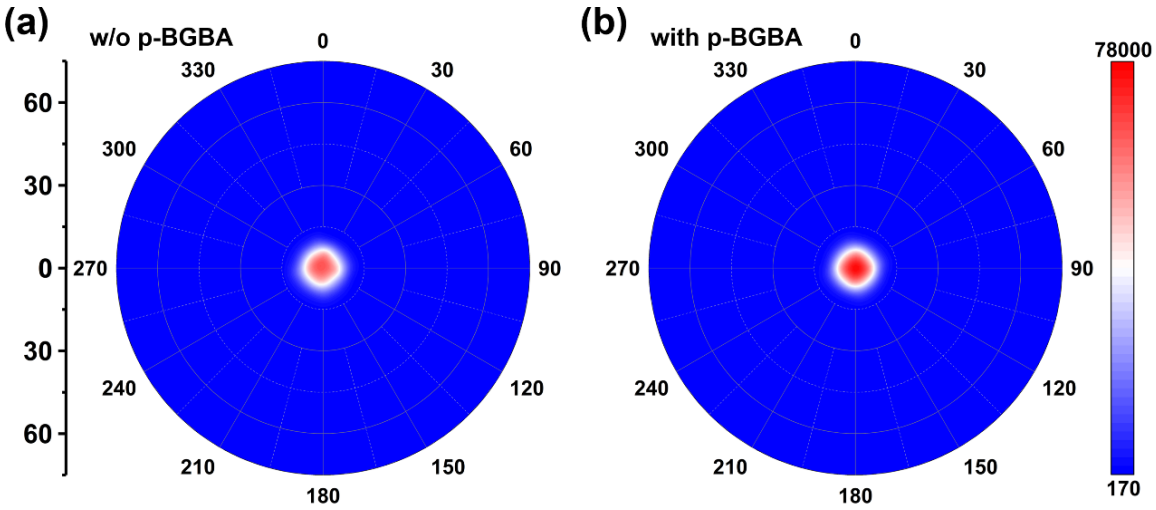

Figure S6. Polar diagrams of the (100) crystal facet of perovskite films without **(a)** and with **(b)** p-BGBA.

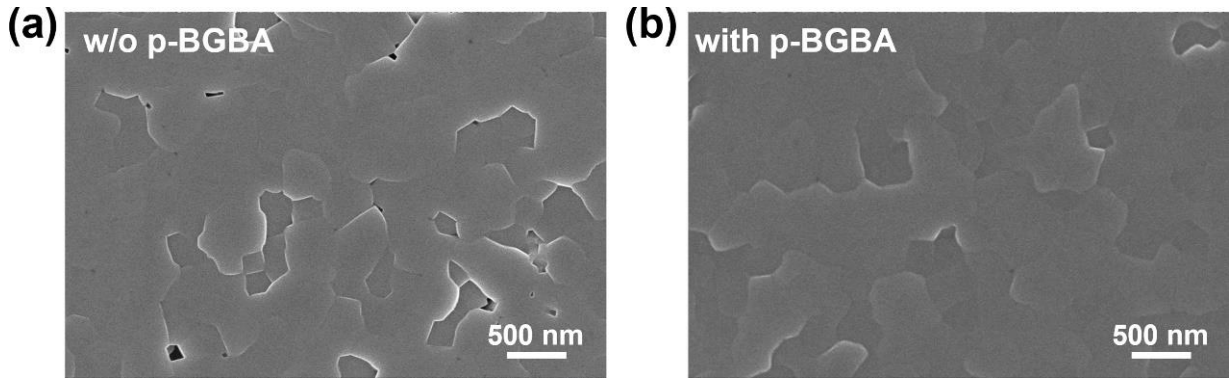

Figure S7. SEM images of perovskite films. **(a)** Without and **(b)** with p-BGBA.

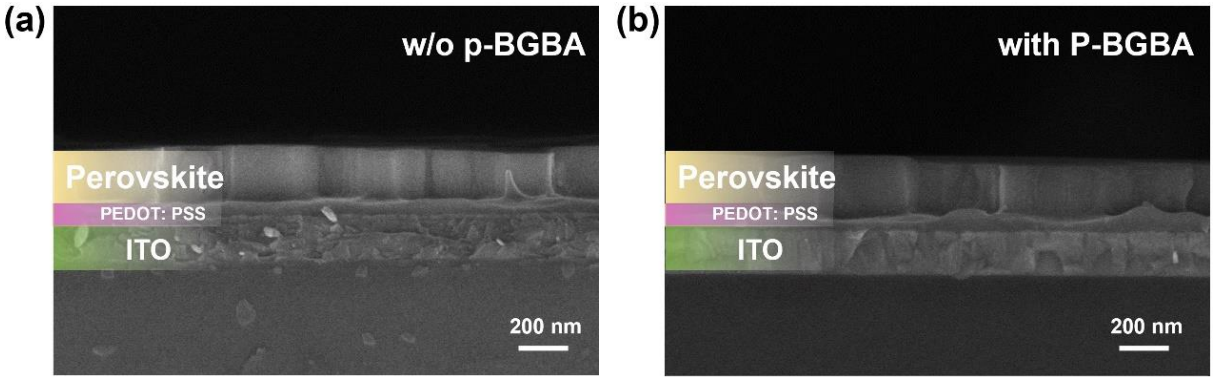

Figure S8. Cross-sectional SEM images of perovskite films. **(a)** Without and **(b)** with p-BGBA.

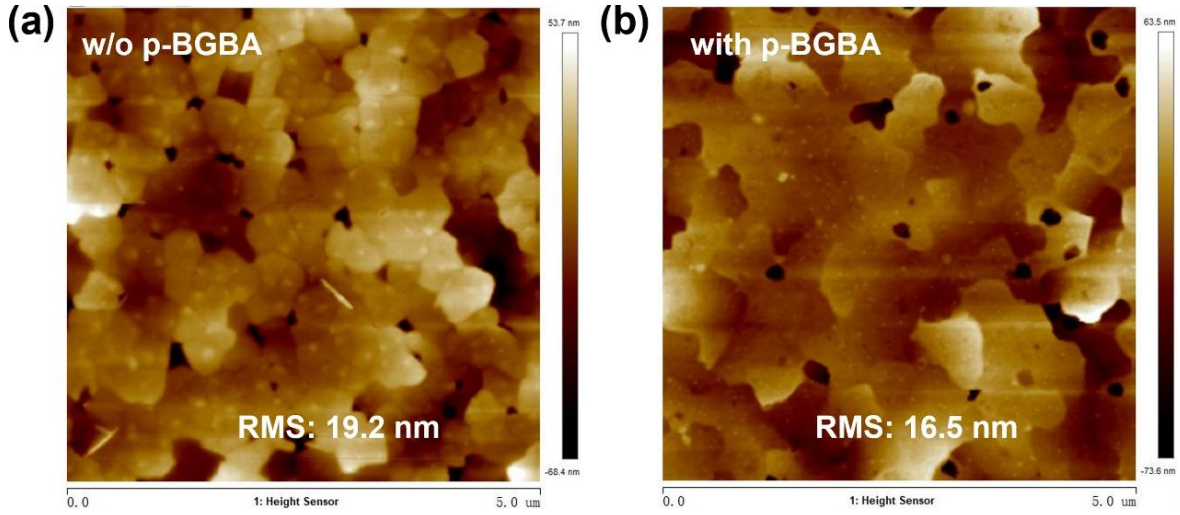

Figure S9. AFM images of perovskite films. **(a)** Without and **(b)** with p-BGBA.

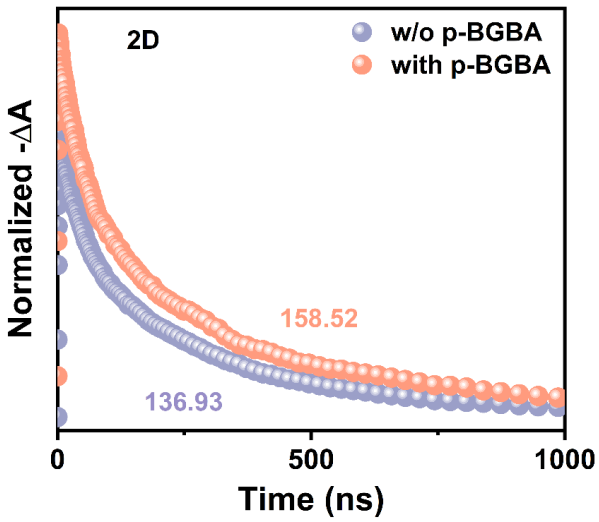

Figure S10. Dynamic transient absorption (TA) decay of the 2D phase.

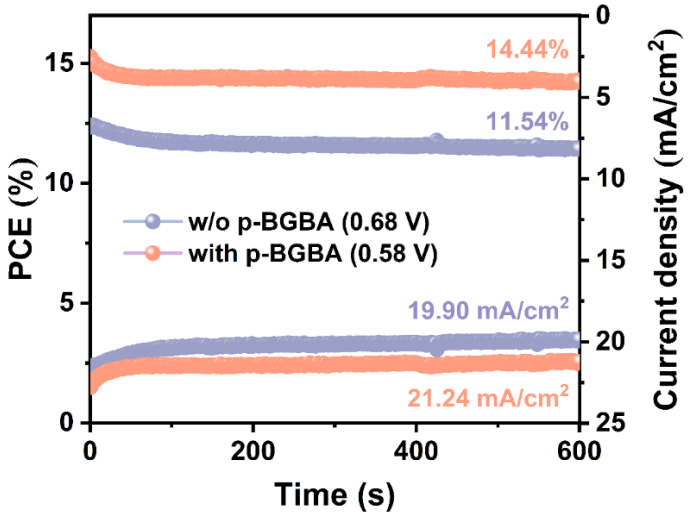

Figure S11. Steady-state power output at the maximum power point.

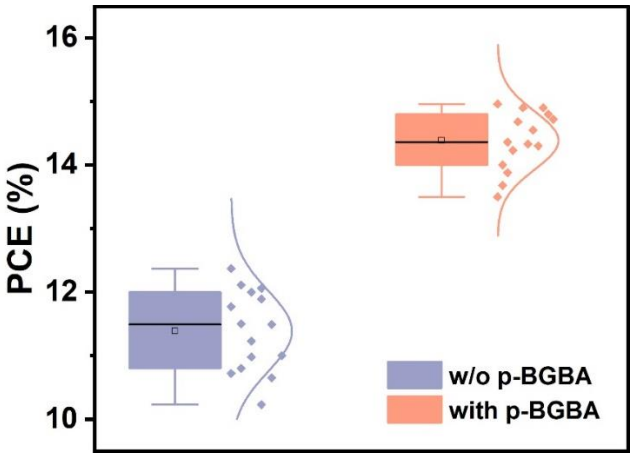

Figure S12. Statistical distribution of the PCEs for 15 tin PSCs without and with p-BGBA.

Table S1. TRPL fitting parameters for perovskite films without and with p-BGBA.

Sample	A_1	τ_1 (ns)	A_2	τ_2 (ns)	τ_{ave} (ns)
w/o p-BGBA	0.62	13.94	0.38	25.26	18.24±1.02
with p-BGBA	0.64	20.63	0.36	49.82	31.14±0.91

Table S2. TA fitting parameters of the perovskite films without and with p-BGBA.

Sample	Phase	A_1	τ_1 (ps)	A_2	τ_2 (ps)	τ_{ave} (ps)
w/o p-BGBA	2D	0.47	33.09	0.53	229.02	136.93
	3D	0.17	9.11	0.83	120.21	101.32
with p-BGBA	2D	0.50	37.72	0.50	279.32	158.52
	3D	0.20	11.51	0.80	183.15	148.82