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## Supporting Information

This file includes Figure S1-S11 and table S1-S2.



Figure S1. <sup>1</sup>H NMR spectra of MAPbI<sub>3</sub>, GABAI, (GABA)<sub>0.1</sub>MA<sub>0.9</sub>PbI<sub>3</sub>, and GABAI-CsPbI<sub>3</sub> in deuterated Dimethyl sulphoxide–d6 (DMSO-d6).



**Figure S2.** Intensity and FWHM variations of (110) peak of the controlled MAPbI<sub>3</sub> films (0 mol%) and (GABA)<sub>x</sub>MA<sub>1-x</sub>PbI<sub>3</sub> films with different GABA-incorporation concentrations: 2.5 mol% (x=0.025), 5 mol% (x=0.05), 10 mol% (x=0.1), 20 mol% (x=0.2), and 50 mol% (x=0.5).



Figure S3. PL spectra of controlled MAPbI<sub>3</sub>, (GABA)<sub>0.025</sub>MA<sub>0.975</sub>PbI<sub>3</sub>, (GABA)<sub>0.05</sub>MA<sub>0.95</sub>PbI<sub>3</sub>, (GABA)<sub>0.1</sub>MA<sub>0.9</sub>PbI<sub>3</sub>, (GABA)<sub>0.2</sub>MA<sub>0.8</sub>PbI<sub>3</sub>, and (GABA)<sub>0.5</sub>MA<sub>0.5</sub>PbI<sub>3</sub> perovskite films.



**Figure S4.** Time-resolved photoluminescence (TRPL) spectra of MAPbl<sub>3</sub> and (GABA)<sub>0.1</sub>MA<sub>0.9</sub>Pbl<sub>3</sub> perovskite films on (a) bare glass substrates. (b) glass/TiO<sub>2</sub> substrates.



**Figure S5**. Representative *J-V* curves of solar cells based on the MAPbI<sub>3</sub>, (GABA)<sub>0.025</sub>MA<sub>0.975</sub>PbI<sub>3</sub>, (GABA)<sub>0.05</sub>MA<sub>0.95</sub>PbI<sub>3</sub>, (GABA)<sub>0.1</sub>MA<sub>0.9</sub>PbI<sub>3</sub>, (GABA)<sub>0.2</sub>MA<sub>0.8</sub>PbI<sub>3</sub>, and (GABA)<sub>0.5</sub>MA<sub>0.5</sub>PbI<sub>3</sub> perovskite films.



**Figure S6**. The UPS spectrum of GABAI solution soaked  $TiO_2$  film. The work function of bare  $TiO_2$  was ~4.03 eV. With GABA<sup>+</sup>, the work function of the  $TiO_2$  shifted to ~4.13 eV.



**Figure S7**. (a) Tauc plot of MAPbI<sub>3</sub> and  $(GABA)_{0.1}MA_{0.9}PbI_3$  films from UV-vis spectra.  $(\alpha hv)^{1/2}$  as a function of photon energy. Eg is obtained by linear fit of the data at absorption onset region. (b)The UPS spectrum of MAPbI<sub>3</sub> and  $(GABA)_{0.1}MA_{0.9}PbI_3$  films. Schematic energy level diagrams of (c) TiO<sub>2</sub>, MAPbI<sub>3</sub>, and Spiro-OMeTAD and (d) TiO<sub>2</sub> with GABA<sup>+</sup>,  $(GABA)_{0.1}MA_{0.9}PbI_3$ , and Spiro-OMeTAD in PSCs.



**Figure S8.** *J-V* characteristics of the best-performing device achieved with MAPbl<sub>3</sub> and (GABA)<sub>0.1</sub>MA<sub>0.9</sub>Pbl<sub>3</sub> films obtained by both forward (F) and reverse (R) scanning direction with scanning rate of (a)0.011 V/S (a) 0.044 V/S(a) 0.15 V/S (a) 0.3 V/S.



**Figure S9.** Photovoltaic parameter statistics of the solar cells based on controlled MAPbI<sub>3</sub> films (0 mol%) and (GABA)<sub>x</sub>MA<sub>1-x</sub>PbI<sub>3</sub> films with different GABA-incorporation concentrations: 2.5 mol% (x=0.025), 5 mol% (x=0.05), 10 mol% (x=0.1), 20 mol% (x=0.2), and 50 mol% (x=0.5).



**Figure S10.** XRD patterns of (a) controlled MAPbI<sub>3</sub> films and (b) (GABA)<sub>0.1</sub>MA<sub>0.9</sub>PbI<sub>3</sub> films stored at room temperature in a 60% relative humidity for different days. The pound sign (#) represent the diffraction peak of PbI<sub>2</sub> indexed to the (001) plane.



**Figure S11.** Comparison of ambient air stability of high-performance solar cell devices based on MAPbI<sub>3</sub> and (GABA)<sub>0.1</sub>MA<sub>0.9</sub>PbI<sub>3</sub> perovskite films.

**Table S1.** The extracted equivalent circuit element parameters of series of the devices with the MAPbl<sub>3</sub> and  $(GABA)_xMA_{1-x}Pbl_3$  films with different GABA-incorporation concentrations: 2.5 mol% (x=0.025), 5 mol% (x=0.05), 10 mol% (x=0.1), 20 mol% (x=0.2), and 50 mol% (x=0.5).

	MAPbl <sub>3</sub>	x=0.025	x=0.05	x=0.1	x=0.2	x=0.5
$\overline{\mathbf{R}_{sh}}$ (KQ)	0.202	0.228	0.226	0.225	0.218	0.217
$\mathbf{R}_{ct}$ (KQ)	1.762	1.197	0.892	0.712	3.232	5.013

**Table S2.** Photovoltaic parameters of the best-performance solar cell based on (GABA)<sub>0.1</sub>MA<sub>0.9</sub>PbI<sub>3</sub> and MAPbI<sub>3</sub> film measured with forward and reverse scan at scan rate of 0.011 V/S, 0.044 V/S, 0.15 V/S, 0.3 V/S, respectively.

Scan rate	Sample	Scan	Voc	Jsc	FF	PCE
( <b>v/s</b> )		direction	(V)	(mA/cm²)	(%)	(%)
0.011		Forward	1.08	21.95	75.71	17.94
	(GABA) <sub>0.1</sub> NIA <sub>0.9</sub> PDI <sub>3</sub>	Reverse	1.09	22.03	75.72	18.18
	MAPbl	Forward	1.05	20.21	63.02	13.37
	3	Reverse	1.08	20.86	63.14	14.22
0.044	(GABA), 1MA, Pbl	Forward	1.08	22.06	73.33	17.47
	, ,0.1 0.9	Reverse	1.08	22.07	73.38	17.4
		Forward	1.01	19.85	55.69	11.1
	IVIAPDI <sub>3</sub>	Reverse	1.04	20.03	60.79	12.60
0.15	(GABA), 1MA, Pbl	Forward	1.09	22.00	74.08	17.70
	· /0.1 0.9 3	Reverse	1.10	22.08	75.03	18.22
	MAPbl	Forward	1.04	20.15	62.99	13.20
		Reverse	1.06	20.37	67.10	14.48
0.3	(GABA), 1MA, Pbl	Forward	1.06	22.05	73.59	17.20
	, ,0.1 0.9 3	Reverse	1.07	22.06	74.01	17.4
	MAPbl <sub>a</sub>	Forward	1.02	20.00	64.72	13.20
	3	Reverse	1.05	20.89	65.82	14.4