

Manipulating the crystallization and interfacial charge behavior with a jellyfish-like molecular template for efficient perovskite solar cells

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Supplementary note 1

Materials: Indium Tin Oxides (ITO) glass substrates, lead iodide (PbI_2), Methylammonium iodide ($\text{CH}_3\text{NH}_3\text{I}$, MAI), 4-tert-butylpyridine and lithium bis (trifluoromethanesulfonyl) imide (Li-TFSI), SnO_2 aqueous colloidal dispersion (15wt%, ALFA), 2,2',7,7'-tetrakis (N,N-dip-methoxyphenylamine) 9,9'-Spirobifluorene (Spiro-OMeTAD) and all anhydrous solvents were purchased from YOUXUAN Technology Co. Ltd. (China). MoS_2 was purchased by Nanjing XFNANO Materials Tech Co. Ltd. Glycine was purchased by Aladdin (China). All chemicals and reagents were used as received from chemical companies without any further purification.

Supplementary note 2

Fabrication of perovskite solar cells: SnO_2 was used as ETL, which was coated on the ITO glass sheet by spin-coating. The deposited ITO glass is transferred to a nitrogen-filled glove box (H_2O and $\text{O}_2 < 1$ ppm) to spin-coating the perovskite film which was prepared by 693 mg of PbI_2 , 245 mg of MAI, and different concentrations BDP dissolving in a mixed solvent of Dimethyl sulfoxide (DMSO) and Dimethyl formamide (DMF) ($v:v = 3:7$). The 80 μL of the MAPbI_3 precursor solution was deposited onto the ITO/ SnO_2 layer by a spin coating process, i.e., 500 rpm for 10 s, 4000 rpm for 30s. 400 μL of chlorobenzene (CB) was poured on the rotating substrate 28 s after the start of the procedure. Subsequently, the obtained films were dried at 60°C for 5 min and at 100°C for 10 min. The 75 μL of the mixture hole transport layer solution prepared by mixing 72.3 mg of Spiro-OMeTAD, 18.5 μL with a solution of 500 mg/mL Li-TFSI in acetonitrile and 28.5 μL of 4-tertbutylpyridine in 1 mL of CB was spin-coated on the prepared MAPbI_3 films at 3000 rpm for 30 s. Finally, 100 nm Ag was thermally deposited under vacuum condition.

Fabrication of JLMT: Firstly, 0.1 mg glycine was dissolved in 1 ml deionized water, then coated on SnO₂ at 2000 RMP and annealed at 100 °C for 10 min. Subsequently, 0.1 mg of MoS₂ was dissolved in 1 ml of deionized water and dispersed in ultrasonic machine for 30 min. And then we spun on glycine at 2000 RMP and annealed at 100 °C for 10 min.

Supplementary note 3

Characterization: The scanning electron microscope (SEM) images and energy dispersive spectroscopy (EDS) linear-scan were taken using a Hitachi S-4800. NMR was measured by Bruker Avance III 400MHz, American. TEM and HRTEM images were measured by JEOL 2100F, Japan. KPFM were measured by Park NX20, Korea. The optical properties of the films were analyzed using an UV-Vis-near-infrared. Fourier transform infrared spectroscopy (FTIR) was measured by Nicolet iS10, America. The crystal structure was carried out by X-ray diffraction (XRD) (Japan Rigaku D/max-ga X-ray diffractometer) using Cu K α ($\lambda = 0.15406$ nm) source. PL spectra were obtained using a PL microscopic spectrometer (FLS1000, China) with a 385 nm CW laser excitation source. The TRPL (FLS1000, China) were measured by using an excitation wavelength of 385nm. The main corresponding setup consisted of perovskite, mica-flakes. X-ray photoelectron spectroscopy (XPS) was used to obtain the information on the chemical states of the control and BDP-perovskite film (measured by Escalab250Xi, Germany). Grazing incidence wide-angle X-ray scattering (GIWAXS) was tested at Shanghai BL14B1 station with a light intensity of 10 keV. *J-V* characterizations was carried out was carried out under AM 1.5 G simulated sunlight illuminations (100 mW cm⁻², Model 94043A, Oriel, American). The spectral responses were obtained from an EQE measurement system (Newport PV measurement, American).

Supplementary note 4

Theoretical calculation: In this paper, the theoretical calculations were performed by density function (DFT) methods. The calculations of electrostatic potential (ESP) were conducted on the Gaussian 09 program by using B3PW91 and the all-electron double- ξ valence basis sets of 3-21G*. The calculations of highest occupied molecular orbital (HOMO) and the lowest occupied molecular orbital (LUMO) were conducted on the Gaussian 09 program by using B3PW91 and the all-electron double- ξ valence basis sets of 3-21G*.

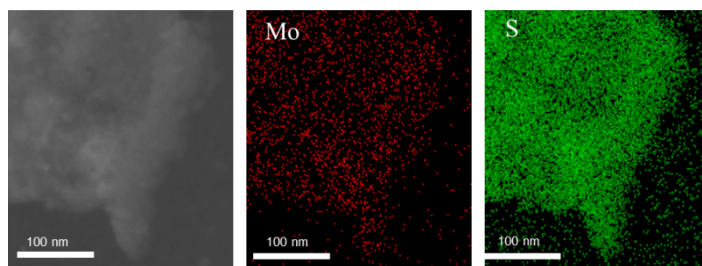


Fig. S1 The EDS image of Mo and S for the MoS₂ nanosheet.

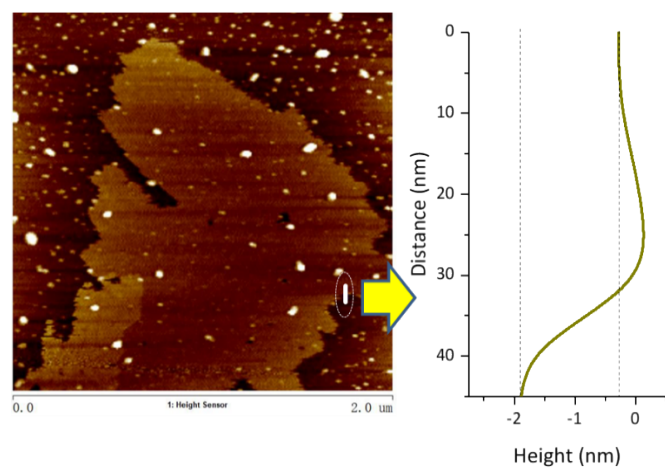


Fig. S2 AFM image of MoS₂ nanosheet with ~1.5 nm thickness.

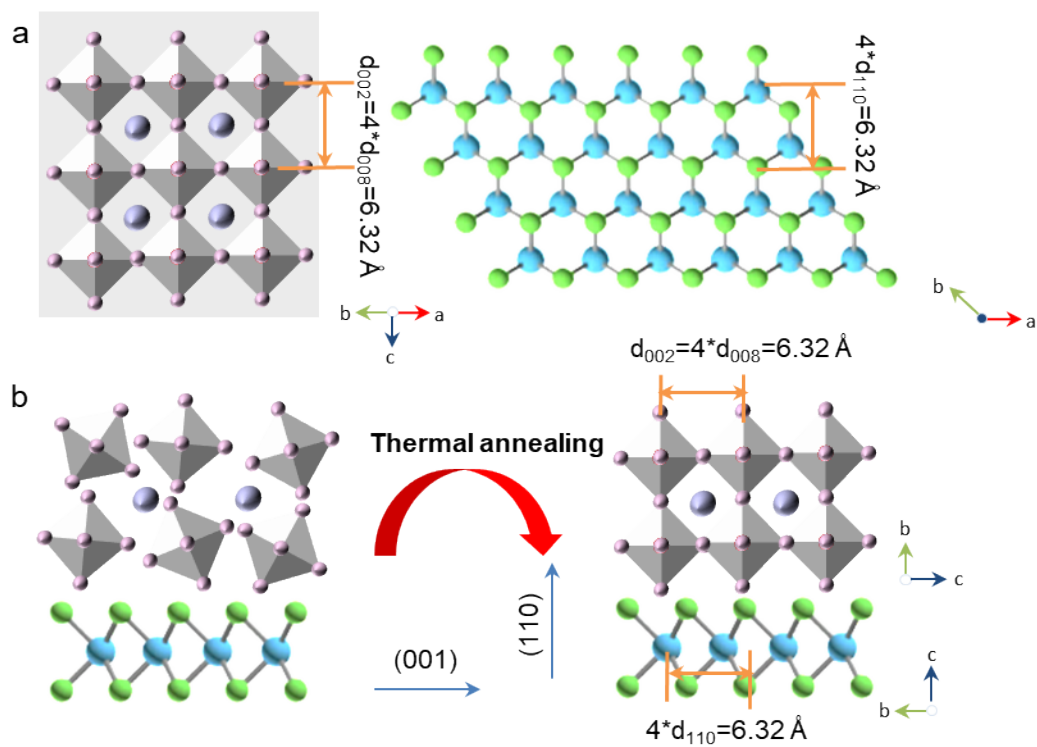


Fig. S3 (a) Atomic crystal structure of the (001) plane of MoS₂ and the (110) plane of the perovskite. (b) Schematic diagram of the vdW epitaxial growth of a MAPbI₃ grain on a MoS₂ surface.

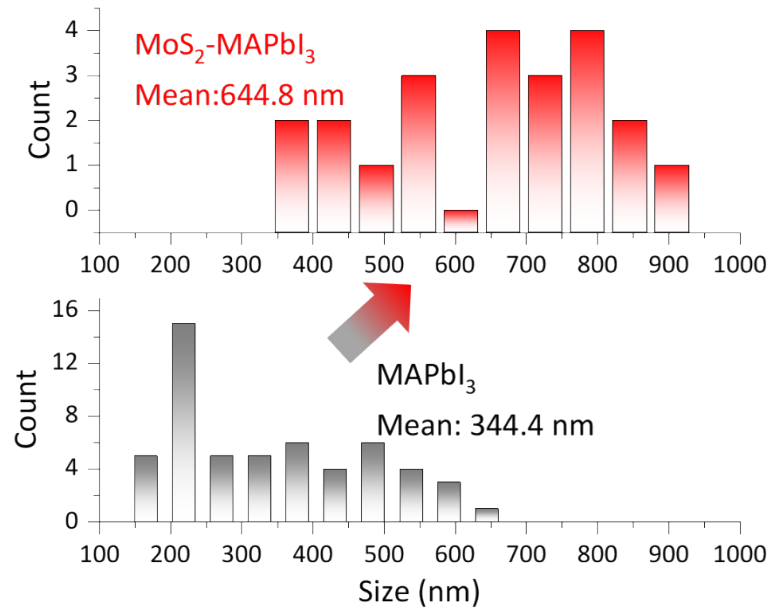


Fig. S4 Histogram for the MAPbI₃ and the MoS₂-MAPbI₃ film with different grain sizes.

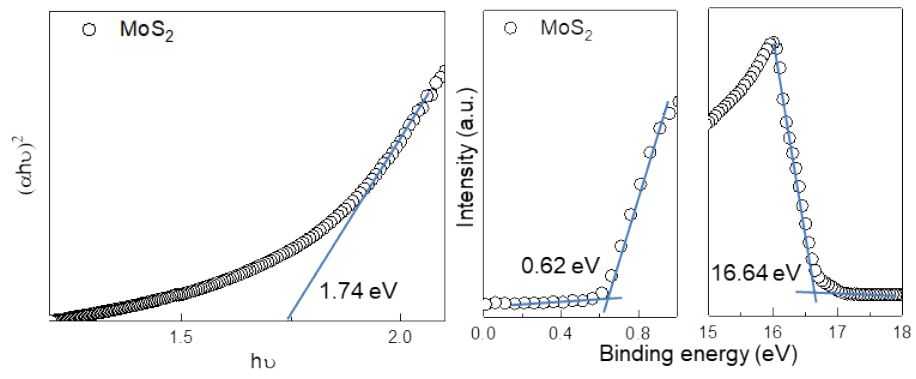


Fig. S5 Optical bandgap and UPS of MoS₂.

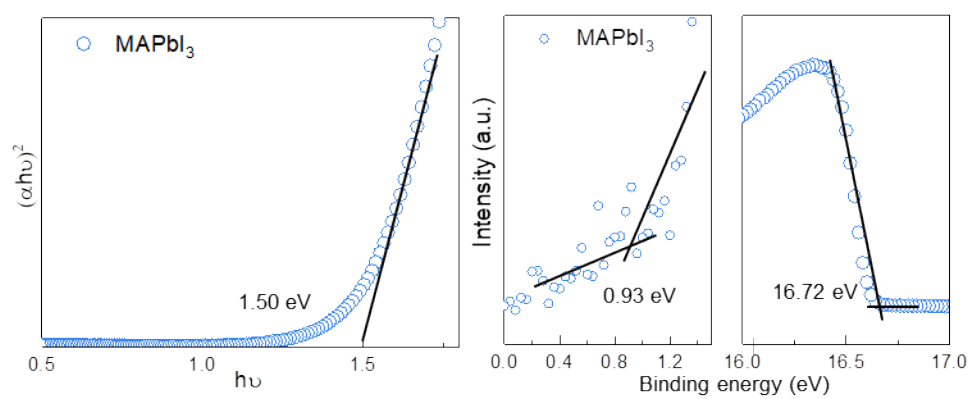


Fig. S6 Optical bandgap and UPS of MAPbI₃.

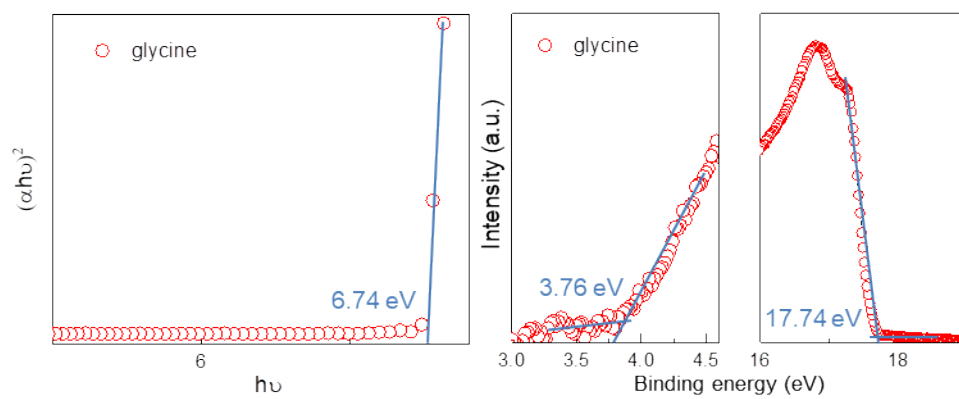


Fig. S7 Optical bandgap and UPS of glycine. Based on this data, the LUMO, E_F , and HOMO levels of glycine are determined as -0.50 eV, -3.48 eV, and -7.24 eV.

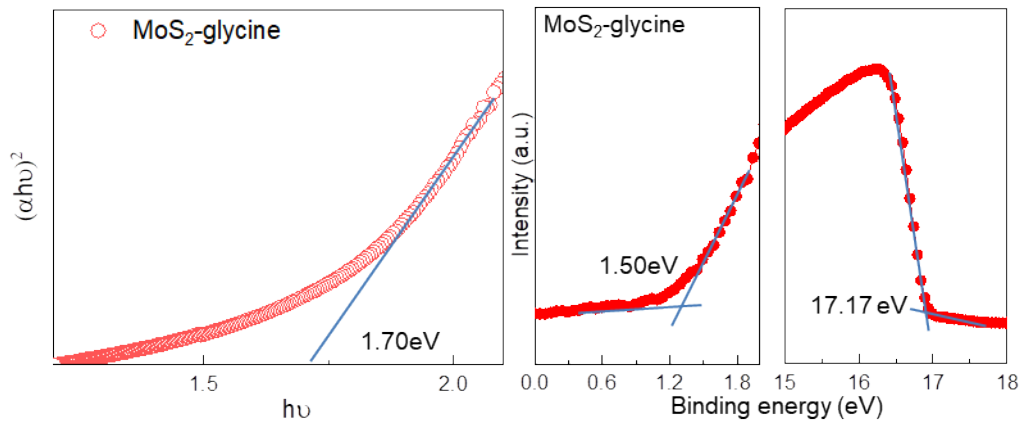


Fig. S8 Optical bandgap and UPS MoS₂-glycine.

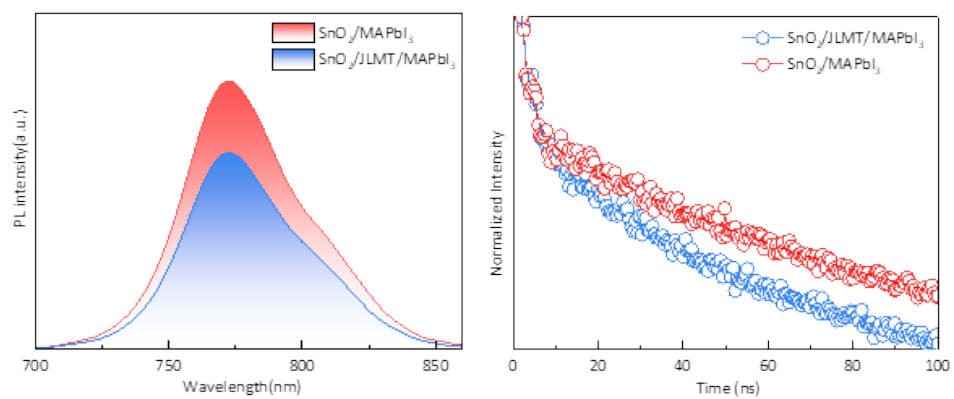


Fig. S9 PL and TRPL of the control- and JLMT-perovskite film.

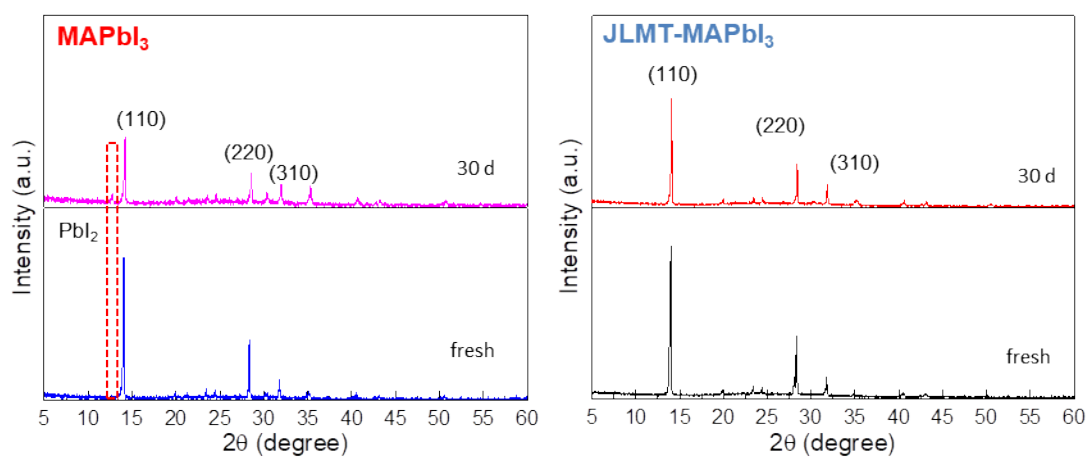


Fig. S10 XRD patterns of the fresh and aged films (exposure to ~40% RH, ~25°C for 30 days) of the MAPbI₃ and the JLMT-MAPbI₃ films.

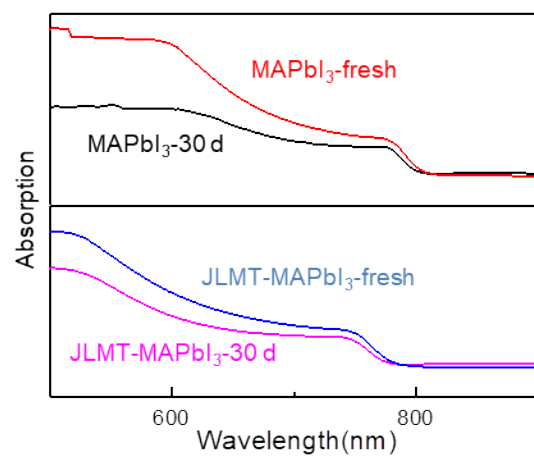


Fig. S11 The UV-vis absorption spectrum of the MAPbI₃ and JLMT-MAPbI₃ films (exposure to ~40% RH, ~25°C for 30 days).

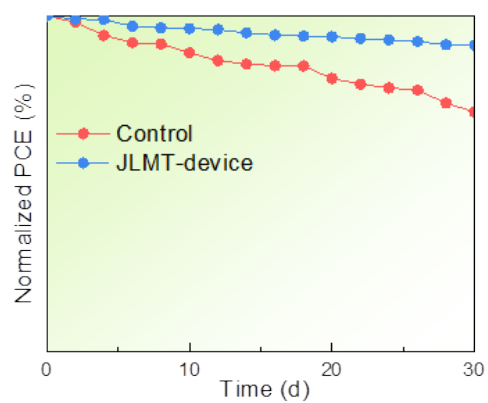


Fig. S12 *PCE* changes of the control- and the JLMT-device.

Table S1. TRPL lifetimes of the MAPbI₃ and JLMT-MAPbI₃ film.

Sample	τ_1 (ns)	τ_2 (ns)
MAPbI ₃	2.24	112.08
JLMT-MAPbI ₃	2.76	69.79

Table S2. The illuminated output parameters of PSCs

	J_{SC} (mA cm ⁻²)	V_{OC} (V)	FF (%)	PCE (%)
Control-R	21.85	1.13	74.81	18.48
Control-F	21.11	1.12	72.35	17.11
JLMT-device-R	22.83	1.20	79.44	21.76
JLMT-device-F	22.79	1.19	79.26	21.50

Table S3. The mean and standard deviation of parameters of the PSCs.

		V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
Mean	MAPbI ₃	1.13	21.06	72.45	17.58
	JLMT-MAPbI ₃	1.10	22.68	75.60	21.26
Standard deviation	MAPbI ₃	0.08	0.72	3.40	0.75
	JLMT-MAPbI ₃	0.03	0.48	3.08	0.25

Table S4. Fitting results from the EIS of PSCs based on the control and BDP-device.

	R_s ($\Omega \text{ cm}^2$)	R_{tr} ($\Omega \text{ cm}^2$)	R_{rec} ($\Omega \text{ cm}^2$)
Control	155	8292	72600
JLMT-device	119	7465	101700