Probing Encapsulation-Induced Changes in 2D/3D Perovskite Heterostructures for Perovskite Solar Cells

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1. EXPERIMENTAL SECTION

1.1 Materials

All chemicals used in this study were purchased from Xi'an Qili Optoelectronic Materials Co., Ltd., unless otherwise specified, and were used without further purification. The materials included lead iodide (PbI_2, 99.99%), formamidinium iodide (FAI, 99.9%), cesium bromide (CsBr, 99.5%), octylammonium bromide (OAI, 99.5%+), 2-phenylethylammonium iodide (PEAI, 99.5%), phenylammonium chloride (PACl, 99%), lead chloride ($PbCl_2$, 98%), lead bromide ($PbBr_2$), zinc bromide ($ZnBr_2$), 2,2′,7,7′-Tetrakis(N,N-di-p-methoxyphenylamine)-9,9′-spirobifluorene (spiro-OMeTAD, 99%), tert-butylpyridine (tBP), methoxypropyl sulfoxide (MPSO), and dimethyl sulfoxide (DMSO). The purities of $PbBr_2$, $ZnBr_2$, tBP, TMSO, MPSO, and DMSO were not specified.

1.2 Device Fabrication

1.2.1 Preparation of Solution

Mixed-orientation perovskite ($FA_{0.9}Cs_{0.1}PbI_3$): FAI, CsI, and PbI_2 powders were weighed according to the stoichiometric ratio and dissolved in a DMF: TMSO (9:1 v/v) solvent mixture to form the perovskite precursor solution. For the (100)-oriented perovskite ($FAPbI_3$), FAI, MACl, RbCl, PACl, $PbCl_2$, and PbI_2 were weighed and dissolved in a DMF: MPSO (10:1, v/v) solvent mixture. For the (111)-oriented perovskite ($FA_{0.85}Cs_{0.1}MA_{0.05}Pb(I_{0.95}Br_{0.05})_3$), FAI, MABr, CsI, and PbI_2 were weighed and dissolved in a DMF: DMSO (8:1, v/v) solvent mixture to form the precursor solution. As obtaining highly pure (100)- and (111)-oriented films under an identical composition remains experimentally challenging, the precursor composition was slightly adjusted while preserving the key perovskite framework, namely an FA-dominated A-site, Pb as the B-site cation, and I as the dominant halide, to enable the formation of films with well-defined orientations.

For 2D passivation materials, PEAI, PBAI, BAI, and OAI were dissolved in isopropanol (IPA) at an identical molar concentration of 0.0183 mmol and stirred overnight at room temperature.

1.2.2 Perovskite Solar Cell Fabrication

Fluorine-doped tin oxide (FTO) conductive glass substrates (20 mm × 16 mm) were sequentially cleaned in an ultrasonic bath with deionized water containing a glass detergent, deionized water, anhydrous ethanol, and acetone (20 min each). After drying, the substrates were treated with UV-ozone for 15 min. The diluted SnO_2 colloidal dispersion was then spin-coated onto the cleaned substrates and annealed at 150 °C for 30 min. Subsequently, the filtered perovskite precursor solution was deposited by spin coating, followed by thermal annealing at 150 °C for 20 min. Passivation layer was deposited by spin-coating at 3000 rpm for 30 s with an acceleration of 3000 rpm s^{-1}, without further annealing. The hole transport layer was formed by spin coating a filtered spiro-OMeTAD solution in chlorobenzene, which was oxidized in pure oxygen at 40 °C for 4 h. Finally, an 80 nm Au electrode was deposited by thermal evaporation under high vacuum to complete device fabrication.

1.2.3 Encapsulation

The encapsulant used in this study is polyisobutylene (PIB, HelioSeal PVS101). To facilitate delamination of the encapsulated thin films a polyethylene terephthalate (PET) barrier is inserted in between the film/device and PIB. The whole structure was encapsulated by thermal-lamination using a laminator setting at 95°C, with first-step 5 min pressed at 0.5 mbar and second-step 5 min pressed at 500 mbar. After encapsulation, the film sample are separated by removing the barrier layer and the device sample are used directly for I-V test.

1.3 Characterization of Devices and Films

The optical and structural properties of the perovskite films were characterized using various techniques. Steady-state and time-resolved photoluminescence (PL) spectra were measured using an FLS980 spectrometer (Edinburgh Instruments Ltd., UK). The crystal structure of the films was analyzed by X-ray diffraction (XRD) using an X'Pert PRO MPD diffractometer (PANalytical, Netherlands). The surface morphology and cross-sectional structure of the films were examined using a field-emission scanning electron microscope (FE-SEM, Inspect F50, Thermo Fisher Scientific). UV–visible absorption spectra were recorded using a UV-2600 spectrophotometer (Shimadzu Corporation, China). Photovoltaic performance was evaluated under simulated AM 1.5G solar illumination (100 mW/cm²) using a solar simulator (SS-F5-3A, Enlitech Co., Ltd.).

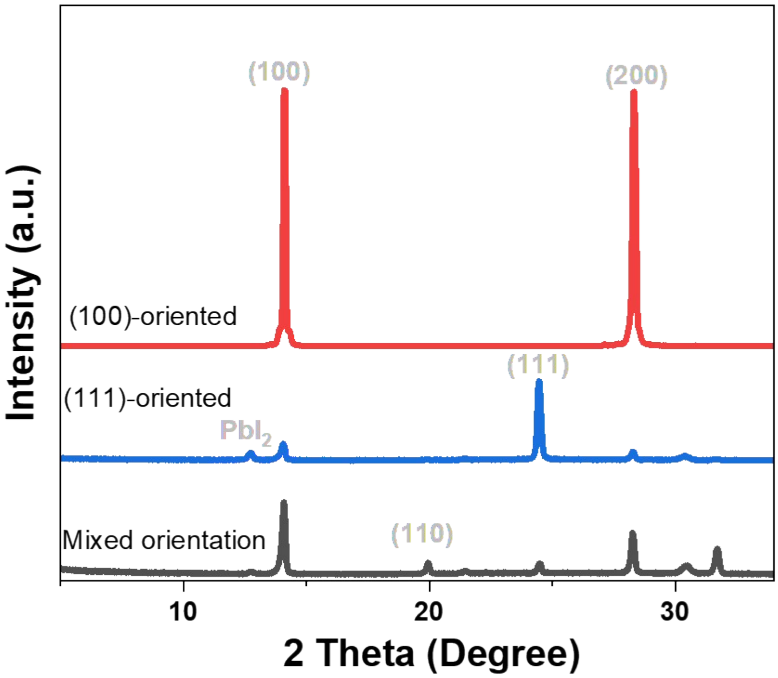

Figure S1. XRD patterns of perovskite films with mixed, (100)-, and (111)-orientations.

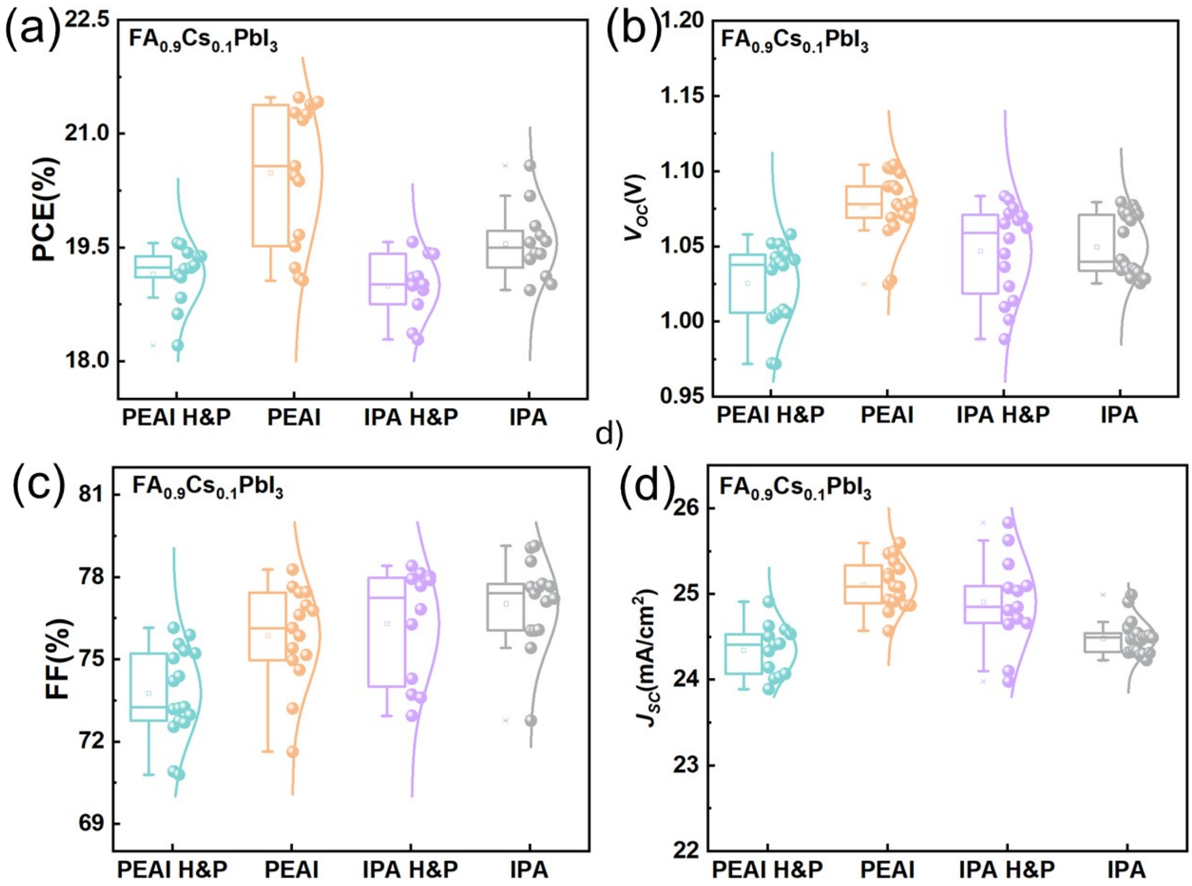

Figure S2. Performance of Mixed oriented PSCs with and without PEAI treatment before and after hot-lamination: (a) Power conversion efficiency (PCE), (b) open-circuit voltage (V_{OC}), (c) fill factor (FF), and (d) short-circuit current density.

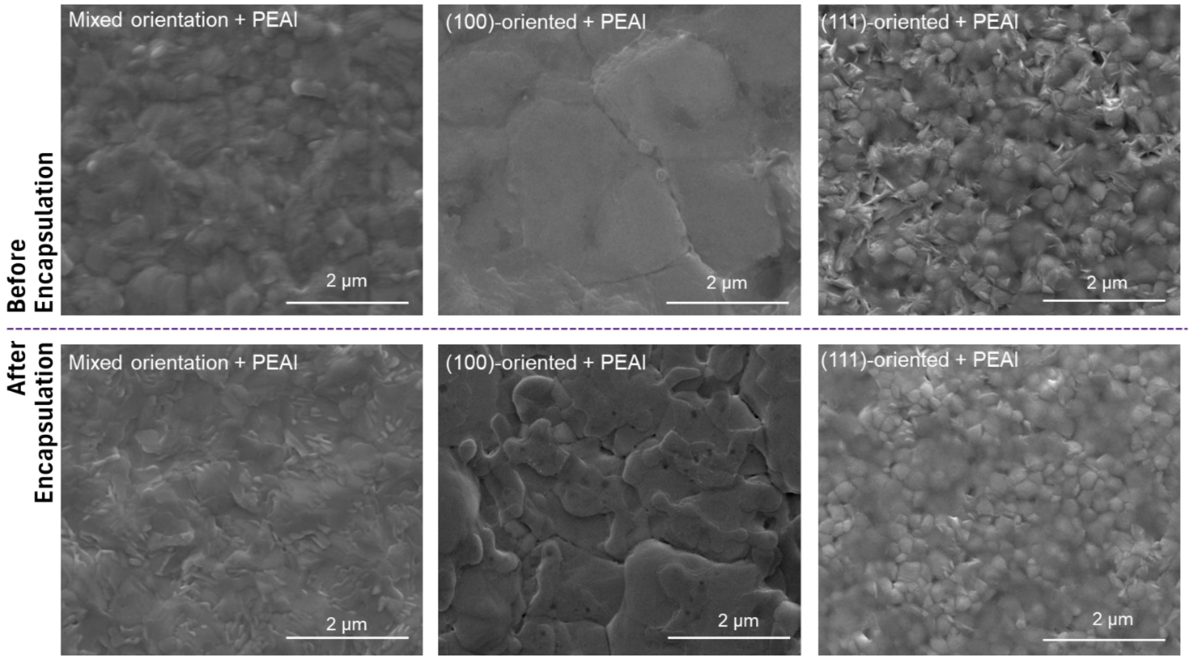

Figure S3. SEM images of PEAI treated perovskite films before and after thermal-lamination-based encapsulation.

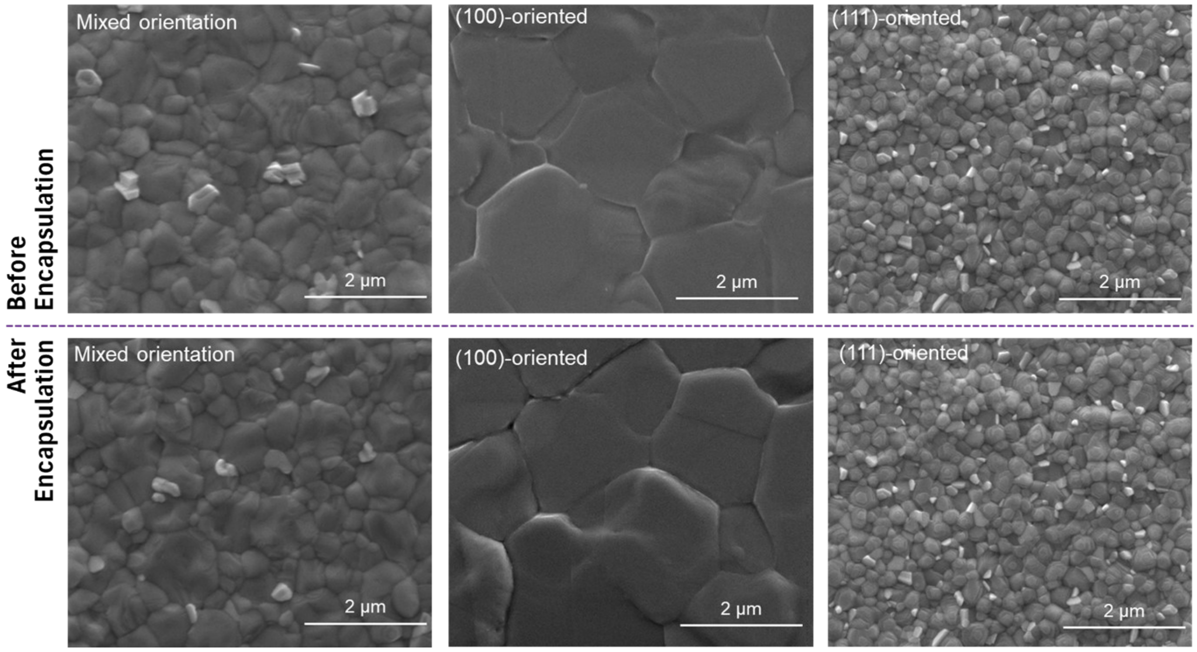

Figure S4. SEM images of unpassivated perovskite films before and after thermal-lamination-based encapsulation.

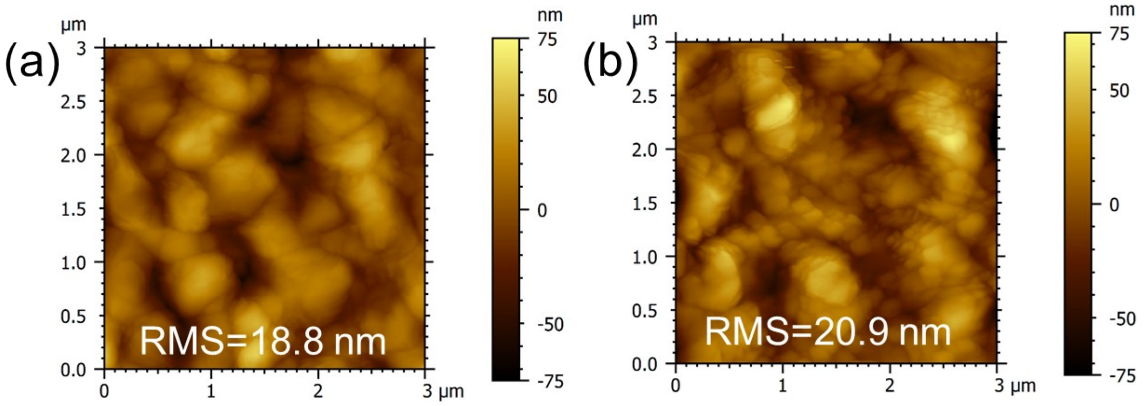

Figure S5. AFM images of PEAI-treated perovskite films (a) before and (b) after thermal lamination.

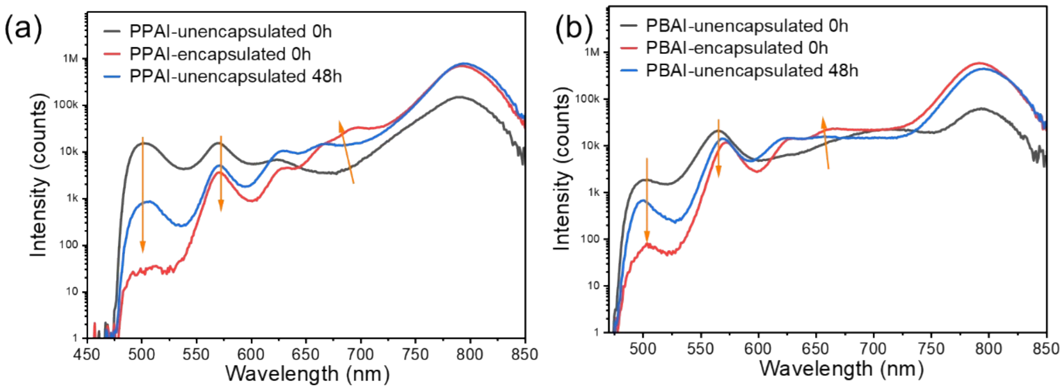

Figure S6. PL spectra of (a) PPAI- and (b) PBAI-treated (111)-oriented perovskite films before and after thermal lamination.

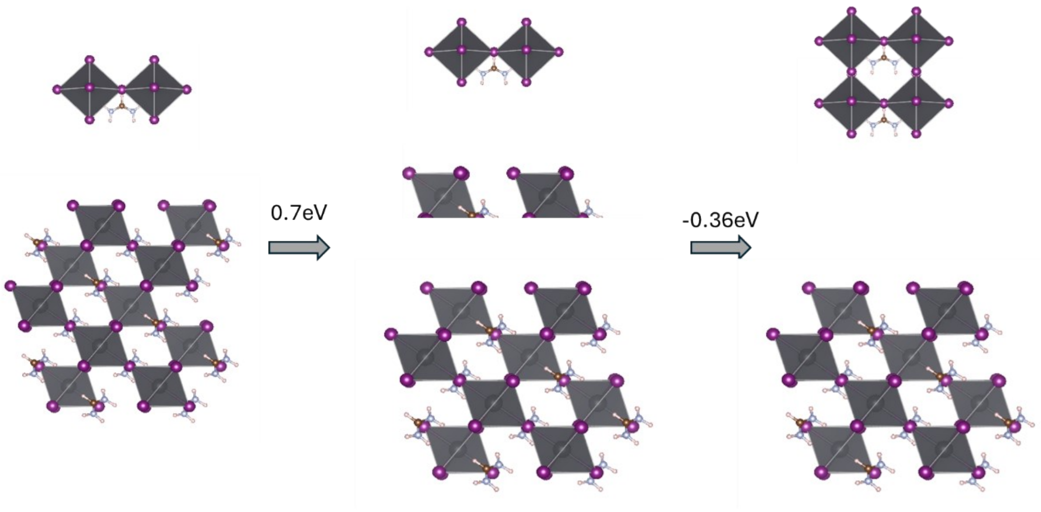

Figure S7. Schematic illustration of 2D perovskite formation on the (111) facet.