Support information for

Ultrafast Transformation of PbI2 in Two-Step Fabrication of Halide Perovskite Film for Long-Term Performance and Stability via Nanosecond Laser Shock Annealing

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Methods

1. Materials: Methylammonium iodide (MAI, purity >99.5%), Lead (II) iodide (PbI_2, purity >99.99%) and Formamidinium iodide (FAI, purity >99.5%) purchased from Xi'an Polymer Light Technology Corp. Methylammonium chloride (MACl, purity>99.5%) and 2-Propanol (IPA, purity>99.5%) were purchased from Libra Tech and Aladdin, respectively. Dimethyl sulfoxide (DMSO, purity>99.5%) and N, N-Dimethylformamide (DMF, purity>99.8%) were purchased from Sigma-Aldrich. All materials are used as received.

2.$FA_{0.95}MA_{0.05}PbI_3$ film preparation: High transmittance fluorine-doped tin oxide (FTO) conductive glass substrate was ultrasonic cleaned with deionized water, acetone, and IPA for 20 minutes, then dried in a vacuum drying oven for 30 minutes. Subsequently, the FTO glass was cleaned with ultraviolet ozone for 20 min. 1.5 M of PbI_2 dissolve in DMF: DMSO (9:1) mixed solvent was dynamic spin-coated onto FTO glass at 2000 r.p.m. for 30s, then annealed at 70℃ for 1 min, and cooled to room temperature for 5 min. After that, a solution of FAI: MAI: MACl (90 mg: 6.39 mg: 9 mg dissolved in 1ml IPA) was dropped onto the surface of PbI_2, spin coating at 2000 r.p.m. for 30s. Nanosecond laser radiation is performed immediately after the wet film is spin-coated (1064 nm, 25 W, pulse duration: 20 ns, scanning speed 300-500 mm/s, beam diameter: 100 μm). For comparison, the film was subjected to thermal annealing, and the annealing temperature is 150℃ for 15 min.

3.Material characterization: The morphology of $FA_{0.95}MA_{0.05}PbI_3$ film was obtained by field emission scanning electron microscopy (FE-SEM, Zeiss SIGMA). The XRD patterns were obtained by Bruker D8 Venture. X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS) data were measured by Thermo Fisher Scientific

ESCALAB250Xi. Raman patterns of perovskite films were characterized by RM1000 for laser excitation at 514.5 nm. PE Lambd 950 was used to perform the ultraviolet-visible absorption (UV-Vis) to obtain the optical bandgap. Steady-state photoluminescence of perovskite films was characterized by FLS 980 for laser excitation at 532 nm. Time-resolved photoluminescence (TRPL) data were collected by FLS1000 using 532 nm laser excitation. A layer of gold was deposited on the surface of the perovskite film, the carrier concentration of the film, the carrier mobility in the film, and the resistivity of the film was obtained by the Hall test. Photocurrent measurement was carried out on the $FA_{0.95}MA_{0.05}PbI_3$ photodetector with the structure of Au/$FA_{0.95}MA_{0.05}PbI_3$/Au, which was measured by Keysight B2901A oscilloscope under 355 nm laser radiation (Power density, 5 mW/cm^2, effective area 1.2 x10^{-3} mm^2). Thermogravimetric analysis (TGA) of perovskite films has an important application in thermal stability analysis of perovskite by using Mettler-Toledo TGA2.

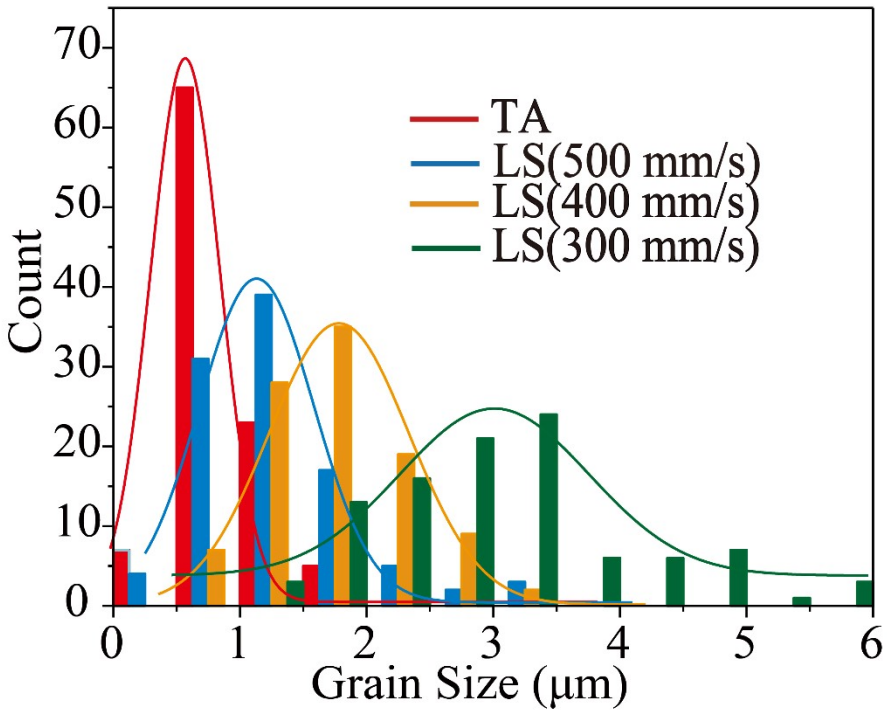

Figure S1. Grain size distribution of $FA_{0.95}MA_{0.05}PbI_3$ perovskite films.

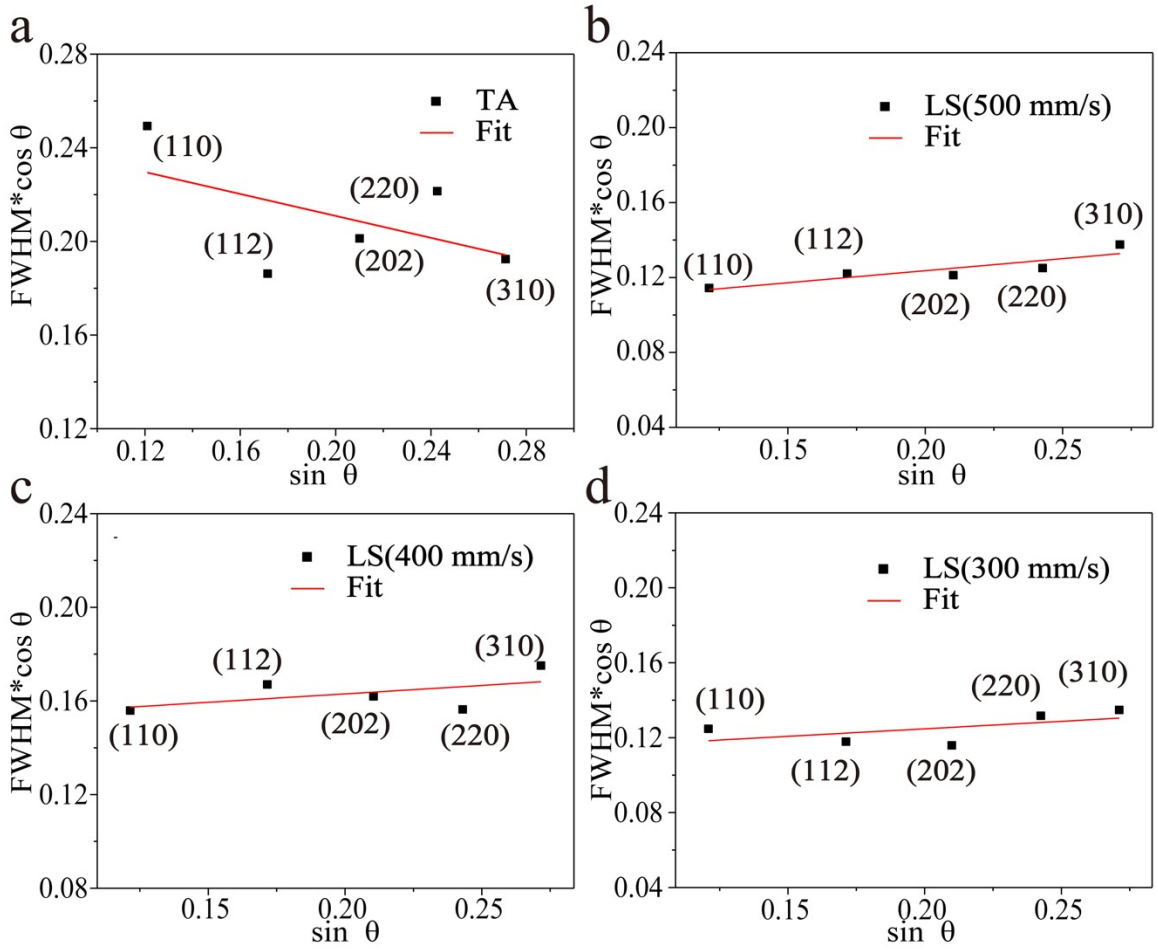

Figure S2. Williamson-Hall plot from X-ray diffraction patterns of perovskite film fabricated by thermal-annealing and laser scanning processes. a)Thermal annealing, b)Laser scanning speed 500 mm/s, c)Laser scanning speed 400 mm/s, d)Laser scanning speed 300 mm/s.

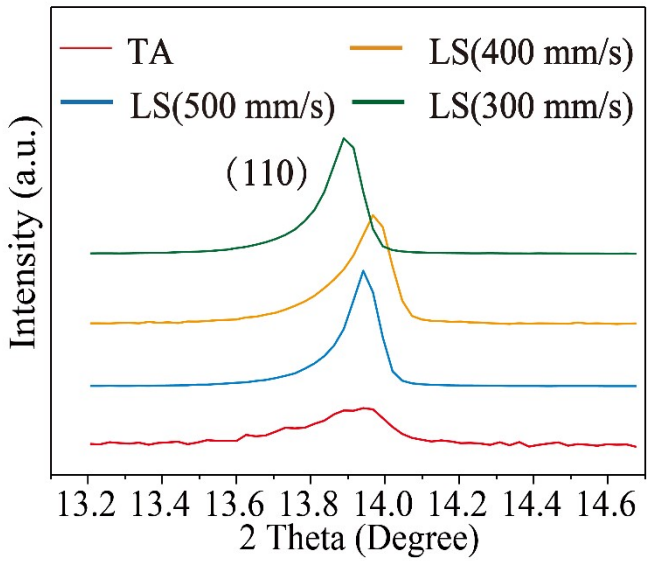

Figure S3. Magnified X-ray diffraction patterns between 13.2° and 14.6°.

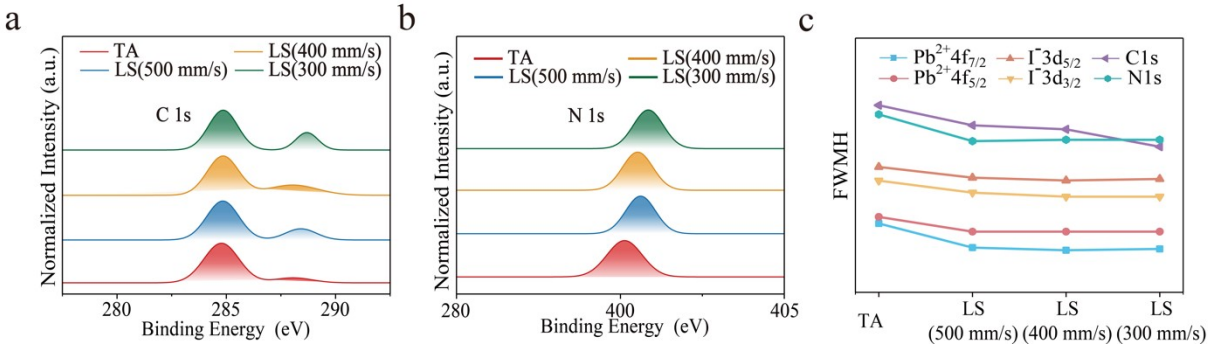

Figure S4. XPS pattern of $FA_{0.95}MA_{0.05}PbI_3$ films fabricated by thermal annealing comparison nanosecond laser scanning.

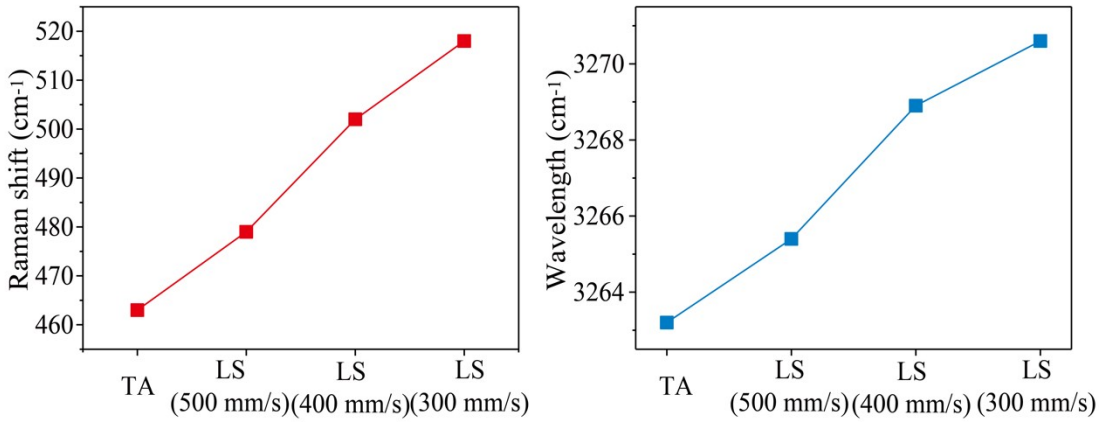

Figure S5. Raman and FTIR spectra appear redshift phenomenon. a)NCN bending region of Raman spectra,b)N-H stretching part of FTIR spectra.

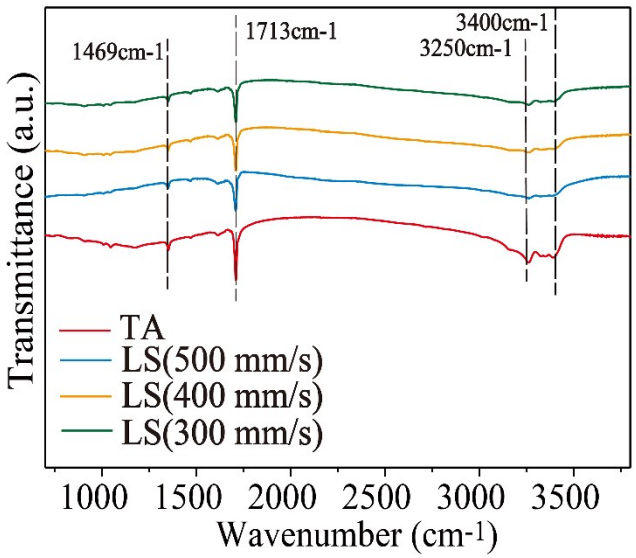

Figure S6. FTIR spectra of perovskite films fabricated by different conditions,1469 cm^{-1} represents C-N stretching; 1713 cm^{-1} represents C=N stretching; 3200cm^{-1} represents N-H stretching; 3400cm-1 represents C-H stretching.

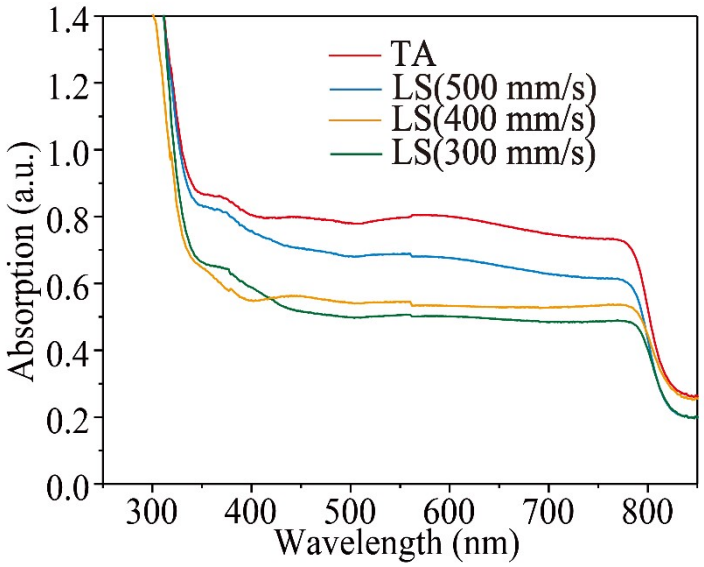

Figure S7. UV–VIS absorption spectra of perovskite film fabricated by thermal-annealing and laser scanning processes.

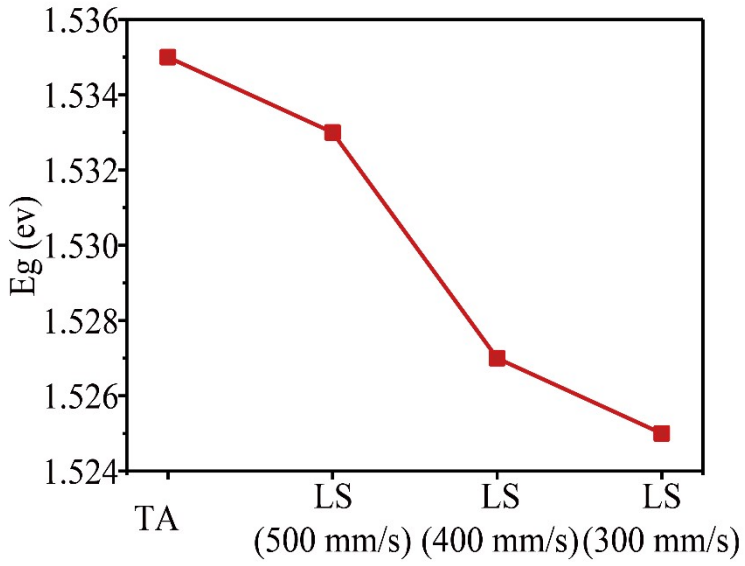

Figure S8. Eg of perovskite film fabricated by thermal-annealing and laser scanning processes.

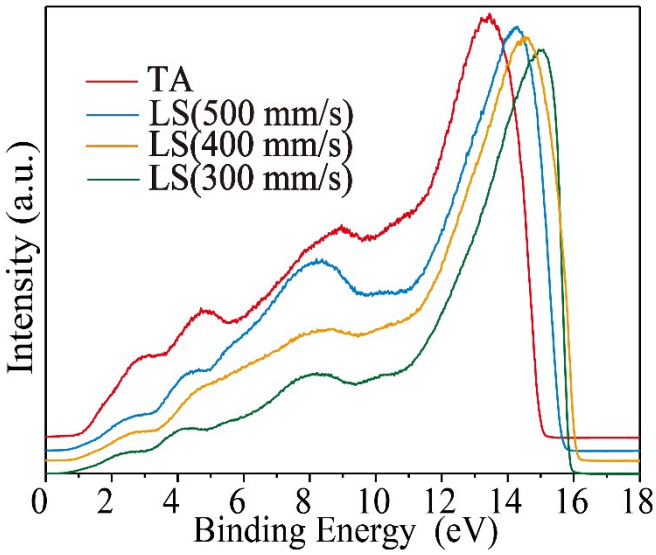

Figure S9. UPS spectra under different fabricate conditions of perovskite films.

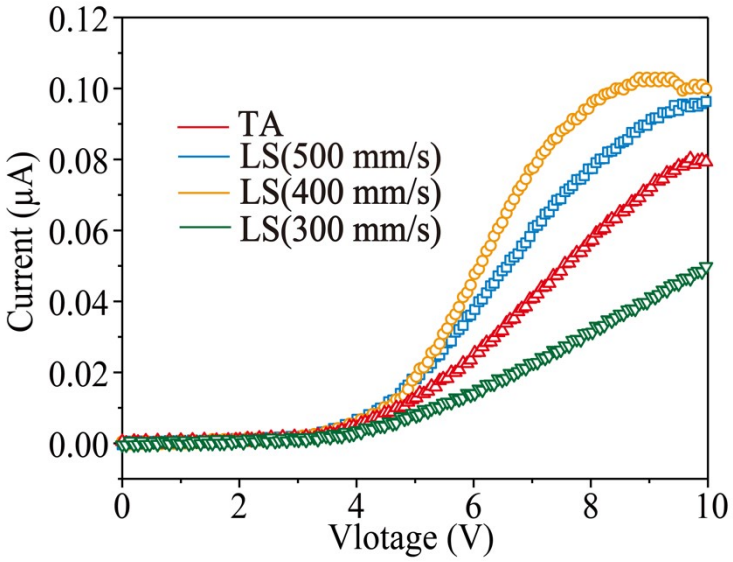

Figure S10. Dark current curve for $FA_{0.95}MA_{0.05}PbI_3$ photodetector.

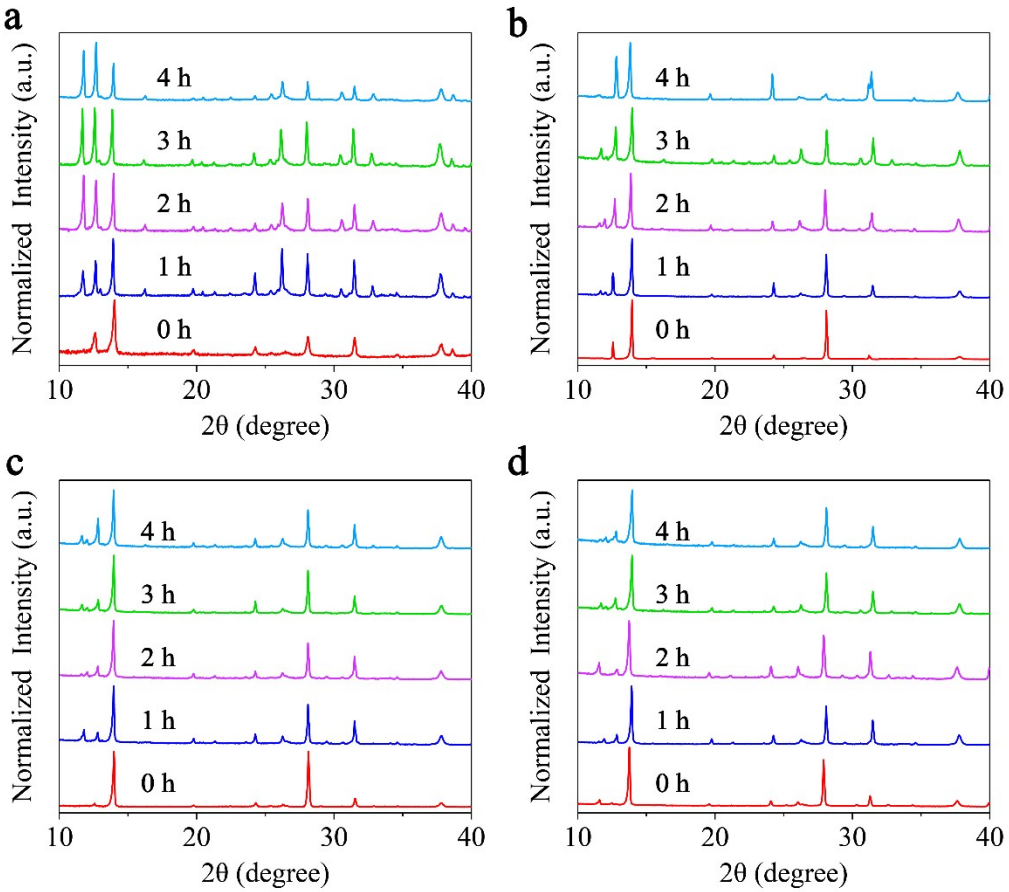

Figure S11. The XRD patterns of a) TA, b) LS(500 mm/s), c) LS(400 mm/s), d) LS(300 mm/s) were kept at 150 ℃ for different time, with in 85% relative humidity.