SUPPLEMENTARY INFORMATION

Dual-source vacuum deposition of pure and mixed halide 2D perovskites: thin film characterization and processing guidelines.

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Experimental Section

Materials. $C_8H_{12}IN$ (PEAI) and $C_8H_{12}BrN$ (PEABr) were purchased from Lumtec. PbI_2 and $PbBr_2$ were purchased from Tokyo Chemical Industry CO (TCI). All materials were used as received.

Thin-films preparation. ITO-coated glass and glass substrates were subsequently cleaned with soap, water and isopropanol in an ultrasonic bath, followed by UV-ozone treatment. They were transferred to a vacuum chamber integrated into a nitrogen-filled glovebox (H_2O and O_2 < 0.1 ppm) and evacuated to a pressure of 10^{-6} mbar. The vacuum chamber used to sublimate the organic halides PEAI and PEABr and lead salts PbI_2 and $PbBr_2$ was equipped with temperature-controlled evaporation sources (Creaphys) fitted with ceramic crucibles and independent temperature controllers and shutters. Three quartz crystal microbalance (QCM) sensors were used: two monitoring the deposition rate of each evaporation source and a third one close to the substrate holder monitoring the total deposition rate. All the sources were individually calibrated for its respective material.

XRD characterization. X-ray diffractograms were collected with a Panalytical Empyrean diffractometer equipped with CuKα anode operated at 45 kV and 40 mA and a PIXcel 1D detector

in scanning line mode. Single scans were acquired in the 2Θ = 5° to 50° range, in Bragg-Brentano geometry in air. All XRD analysis was carried out with Fullprof software. Whole-pattern Le Bail fits assuming a Thompson-Cox-Hastings pseudo-Voigt lineshape are performed to refine lattice parameters.

Optical Characterization. Absorption spectra were collected using a fiber optics based Avantes Avaspec 2048 Spectrometer in air. Photoluminescence was measured using a compact fluorescence lifetime spectrometer C11367, Quantaurus- Tau, with continuous wave 340 nm LED light source, in air.

Morphological Characterization. The surface morphology of the thin films was analyzed using atomic force microscopy (AFM,Multimode SPM, Veeco, USA). Electron microscopy characterization (SEM and EDX) was performed using a Hitachi S-4800 microscope operating at an accelerating voltage of 10 kV.

Time-resolved microwave conductivity. For the TRMC measurements, the 2D perovskite films were mounted in a sealed microwave resonance cavity within a nitrogen glovebox. The TRMC technique monitors the change in reflected microwave power by the loaded microwave cavity upon pulsed laser excitation at various wavelengths (403 nm, 445 nm, and 510 nm). The photo-conductance (ΔG) of the sample was deduced from the laser-induced change in normalized microwave power (ΔP/P) by

$$-K\Delta G(t) = \frac{\Delta P(t)}{P}$$

where K is the sensitivity factor. The yield of generated free charges φ and mobility $\sum\mu = (\mu_e + \mu_h)$ comprise the photoconductance, ΔG_{max}:

$$\eta\sum\mu = \frac{\Delta G_{max}}{I_0 e\beta}$$

Normalized TRMC traces can specifically show comparison of samples on charge carrier decay

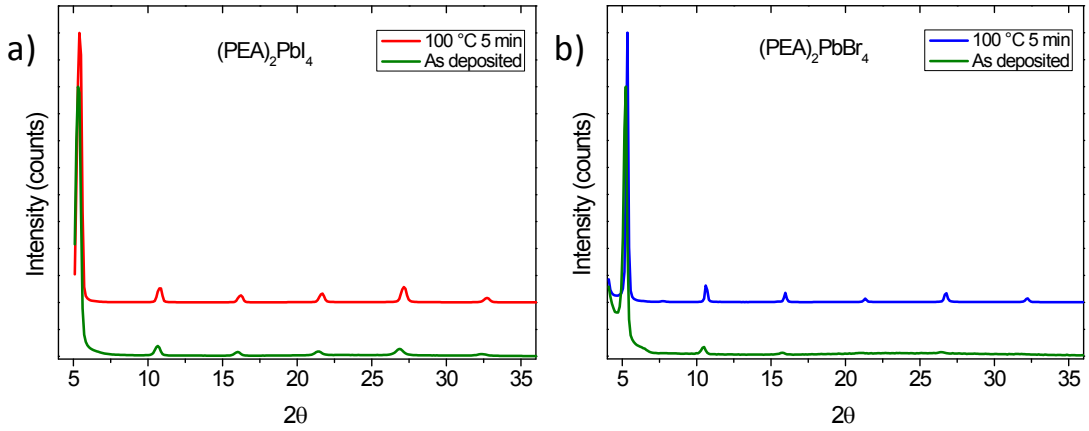

Figure S1. XRD patterns of a vacuum deposited a) PEA_2PbI_4 and b) PEA_2PbBr_4 measured as prepared

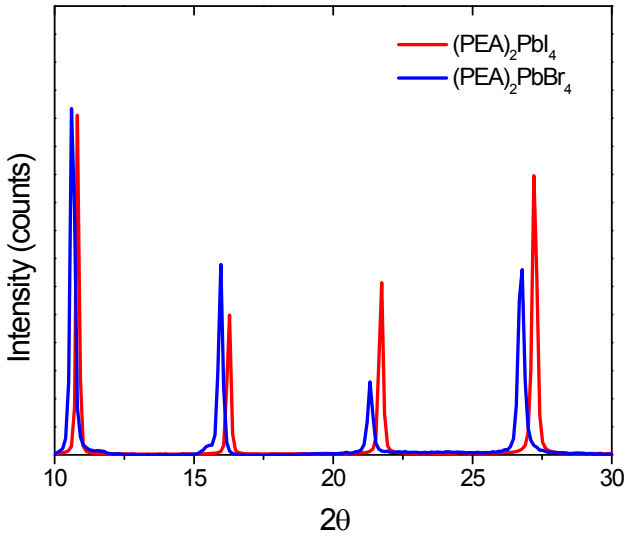

Figure S2. XRD patterns of a vacuum deposited PEA_2PbI_4 and PEA_2PbBr_4

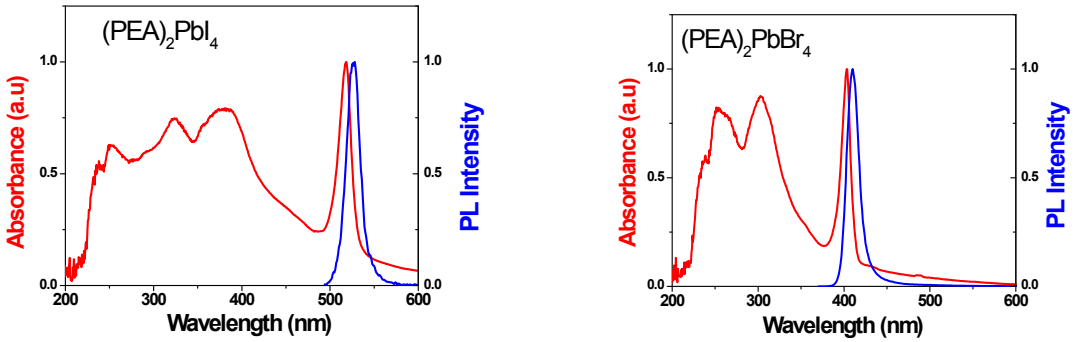

Figure S3. Optical absorption and photoluminescence of 2D PEA_2PbI_4 (λ_{abs} 518 nm, FWHM 20 nm and λ_{em} 528 nm, FWHM 19 nm) and 2D PEA_2PbBr_4 (λ_{abs} 403 nm, FWHM 14 nm and λ_{em} 410 nm, FWHM 17 nm) perovskites thin films prepared from solution process.

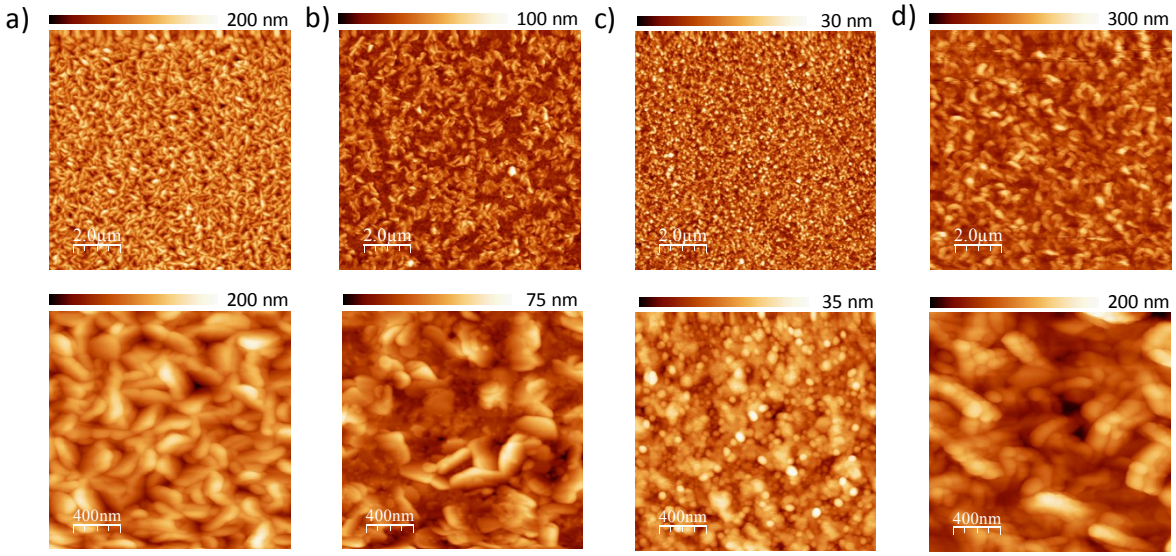

Figure S4. AFM topographies at different magnifications of a) PEA_2PbI_4, b) PEA_2PbBr_4 c) $(PEABr)_2PbI_2$ and d) $(PEAI)_2PbBr_2$ 2D perovskite thin films.

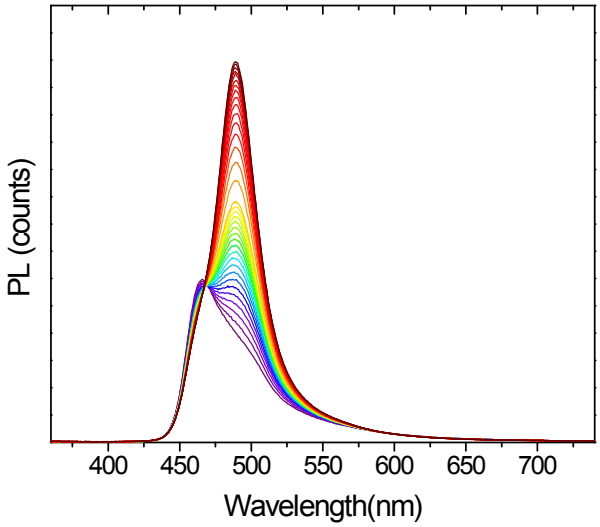

Figure S5. Evolution of the photoluminescence spectra of the $(PEABr)_2PbI_2$ thin film under continuous excitation with a CW laser at 375 nm. Each spectrum is collected every 10 s, going from purple to dark red.