Supplementary Information

Coarsening of one-step deposited organolead triiodide perovskite films via Ostwald ripening for high efficiency planar-heterojunction solar cells

Weidong Zhu,[a] Chunxiong Bao,[a] Yangrunqian Wang,[a] Faming Li,[a] Xiaoxin Zhou,[a] Jie Yang,[a] Bihu Lv,[b] Xiaoyong Wang,[b] Tao Yu,[*abc] and Zhigang Zou [abc]

[a]*National Laboratory of Solid State Microstructures & Eco-Materials and Renewable Energy Research Center (ERERC) at Department of Physics, Nanjing University, Nanjing 210093, P. R. China. E-mail: yutao@nju.edu.cn*

[b]*Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, P. R. China.*

[c]*Jiangsu Key Laboratory for Nano Technology, Nanjing 210093, P. R. China.*

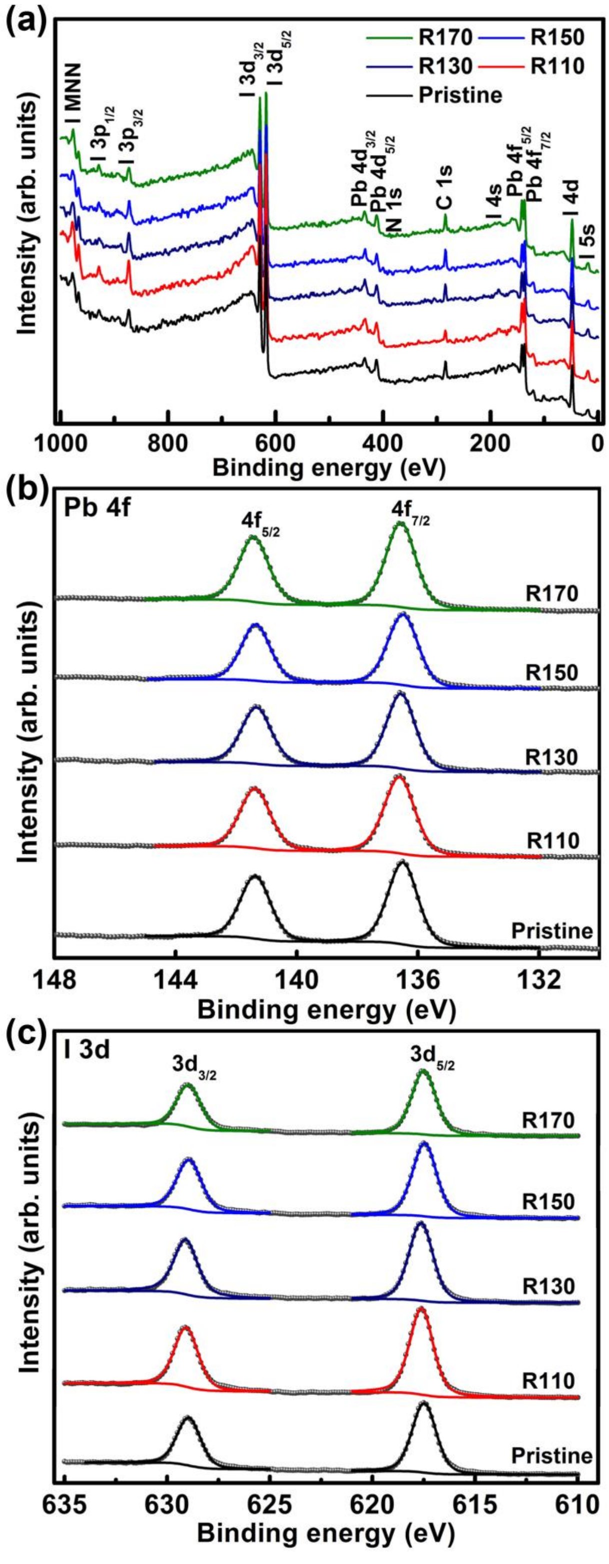

Fig. S1 (a) The low resolution XPS survey spectra of pristine OTP film (Pristine) and coarsened OTP films with the post-synthesis heating treatment temperatures of 110 °C (R110), 130 °C (R130), 150 °C (R150), and 170 °C (R170), respectively. (b) and (c) are the corresponding Pb 4f and I 3d core level XPS spectra.

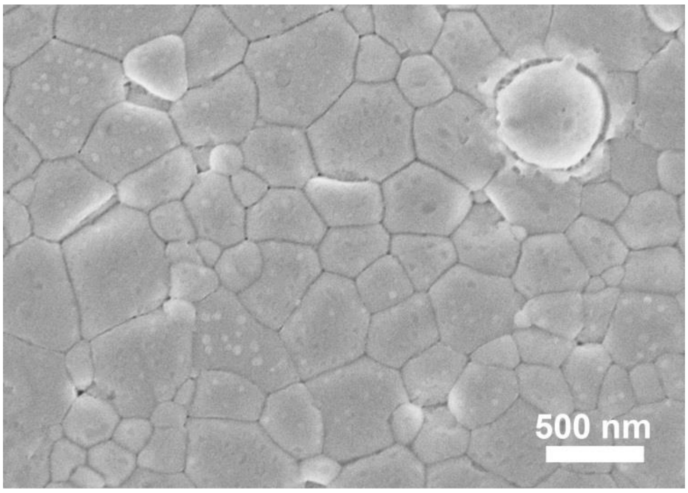

Fig. S2 The top-view SEM image of the pristine OTP film after deposition of CH_3NH_3I.

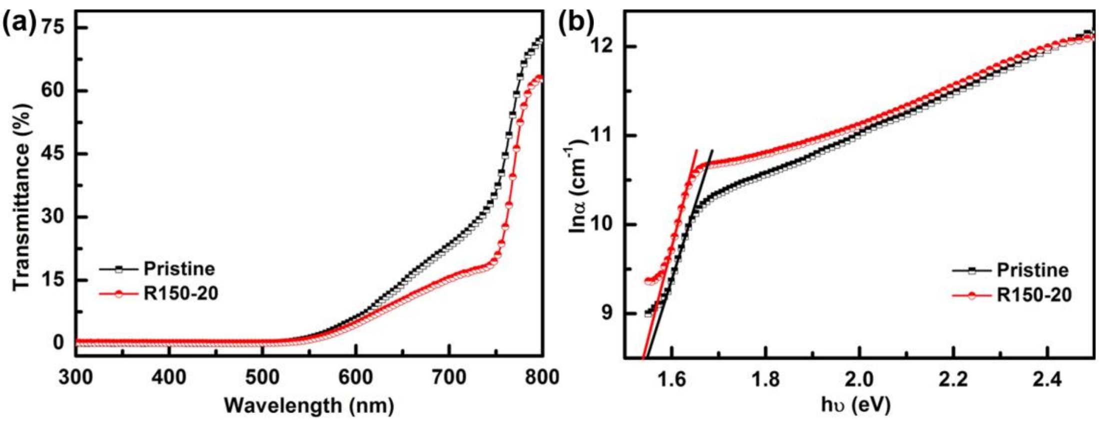

Fig. S3 (a) Transmittance spectra of pristine OTP film (Pristine) and coarsened OTP film with the post-synthesis heating treatment temperature and time of 150 °C and 20 min (R150-20). (b) Determination of the Urbach energy (E_u) of pristine OTP film and R150-20 sample using the empirical equation: $\ln\alpha = \frac{1}{E_u}h\nu + \ln\alpha_0 - \frac{E_g}{E_u}$, where α_0 is constant, $h\nu$ is the photon energy, E_g is the optical bandgap, E_u is the Urbach energy, and α is the absorption coefficient. The α can be calculated from the Transmittance spectrum using the equation of $\alpha = \frac{-\ln T}{d}$ (d is the film thickness). By fitting the linear part of plots of $\ln\alpha$ versus $h\nu$, the Urbach energy can be calculated from the slope of fitted straight line.

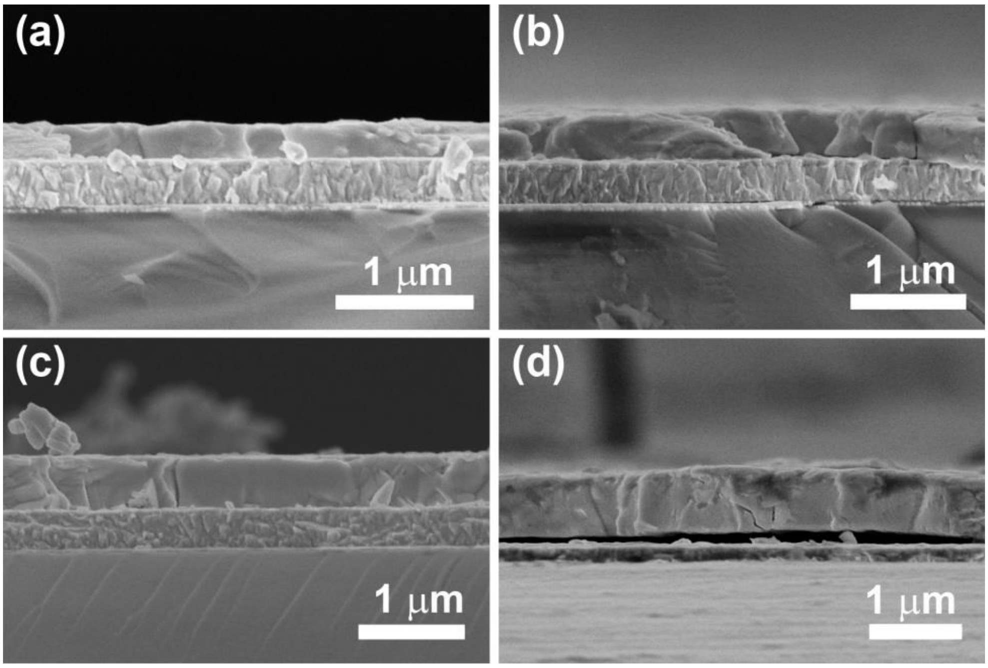

Fig. S4 The cross-section SEM images of R150-20 samples with different thicknesses of 270, 440, 520, and 690 nm, respectively. The film thickness was changed by controlling the rotation speed during deposition of pristine OTP films.

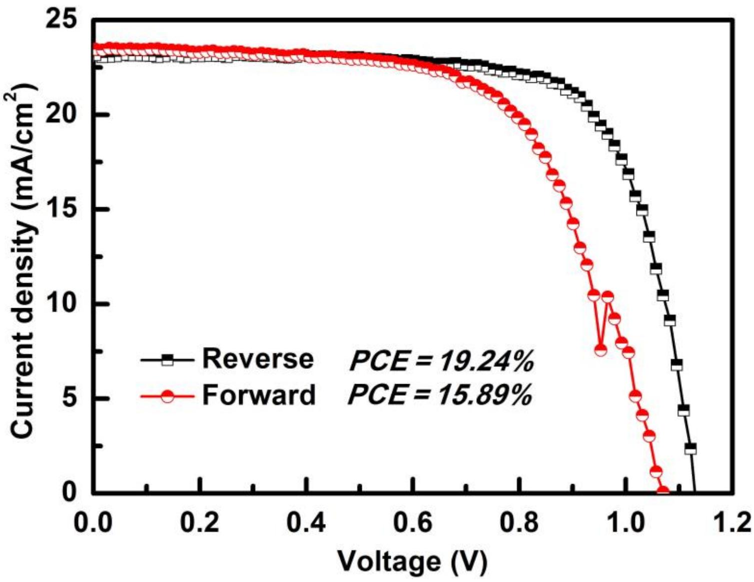

Fig. S5 J-V curves measured at forward scan (Forward, from -0.1 V to 1.2 V) and reverse scan (Reverse, 1.2 V to -0.1 V) for the best performing perovskite solar cell employed the R150-20 sample with thickness of 440 nm. It should be noted that there was a kink at 0.95 V in forward J-V curve of cell, which has been frequently observed in the previous works and was difficult to be eliminated by the modification OTP film quality.[1-3] The underlying reason for this phenomenon is mainly consisted in the migration of iodide ions/interstitials driven by an external electrical bias leading to shift in the effective work function at the respective electrodes of the cells.[1]

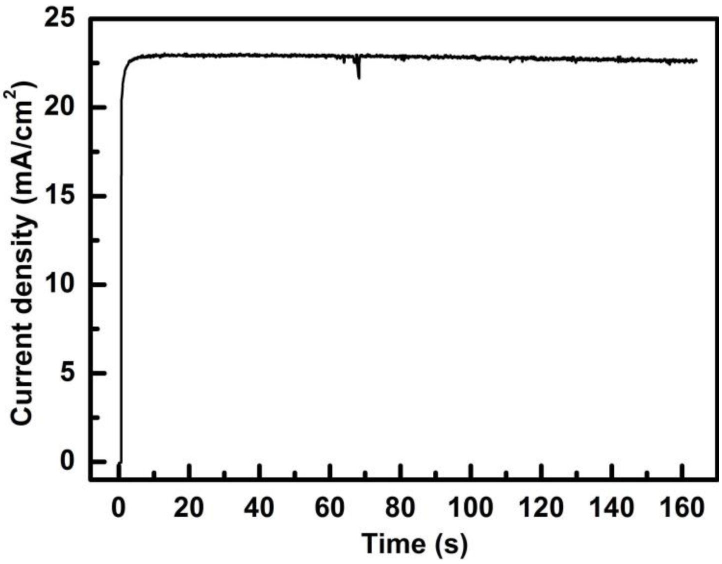

Fig. S6 Photocurrent density as a function of time for best performing perovskite solar cell employed the R150-20 sample with thickness of 440 nm held at a forward bias of 0.01 V. The cell was placed in the dark prior to the start of the measurement.

Table S1 PV parameters for the cells employed R150-20 samples with different thicknesses.

Thickessness (nm)	J_{sc} (mA cm^{-2})	V_{oc} (V)	FF	PCE (%)
270	22.15	1.13	0.674	16.87
440	23.18	1.12	0.710	18.43
520	23.80	1.10	0.691	18.09
690	23.85	1.09	0.648	16.85

Refercences

[1] C. Li, S. Tscheuschner, F. Paulus, P. E. Hopkinson, J. Kießling, A. Köhler, Y. Vaynzof and S. Huettner, *Adv. Mater.*, 2016, **28**, 2446-2454.

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[3] R. S. Sanchez, V. Gonzalez-Pedro, J.-W. Lee, N.-G. Park, Y. S. Kang, I. Mora-Sero and J. Bisquert, *J. Phys. Chem. Lett.*, 2014, **5**, 2357-2363.