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Supporting Information

Construction of Chiral-2D/3D Perovskite Heterojunction Films for Efficient Circularly Polarized Light Detection

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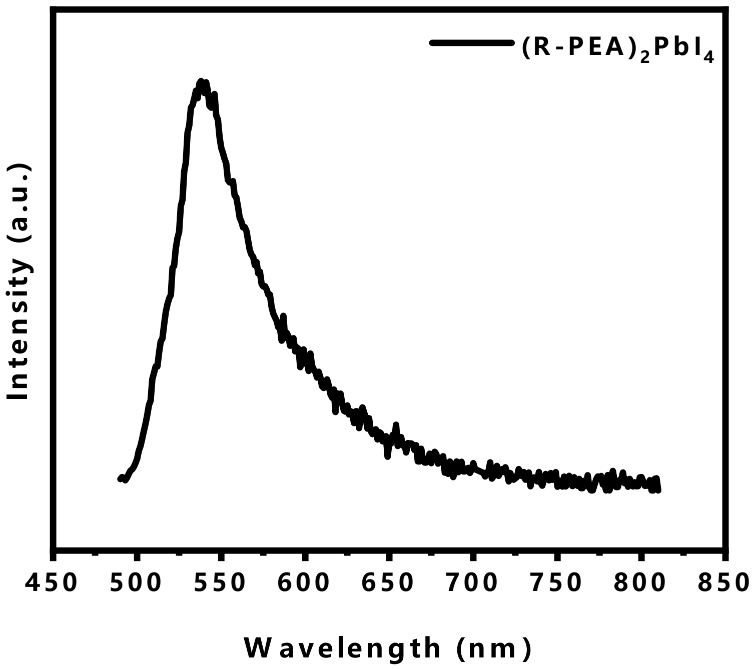

Figure S1. Photoluminescence spectrum of (R-α-PEA)$_2$PbI$_4$ film.

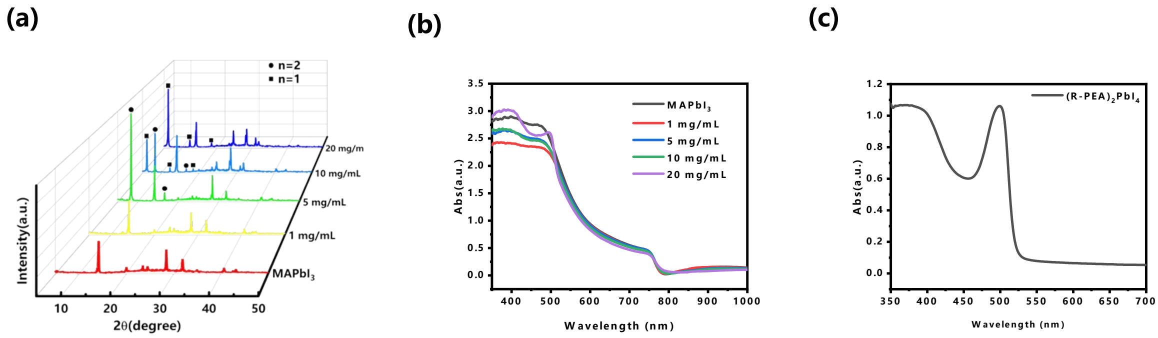

Figure S2. (a,b) XRD patterns (a) and absorption spectra (b) of 3D and (R-α-PEA)I-treated samples with four different concentrations (1, 5, 10, 20 mg/mL). (c) The absorption spectrum of (R-α-PEA)$_2$PbI$_4$ film.

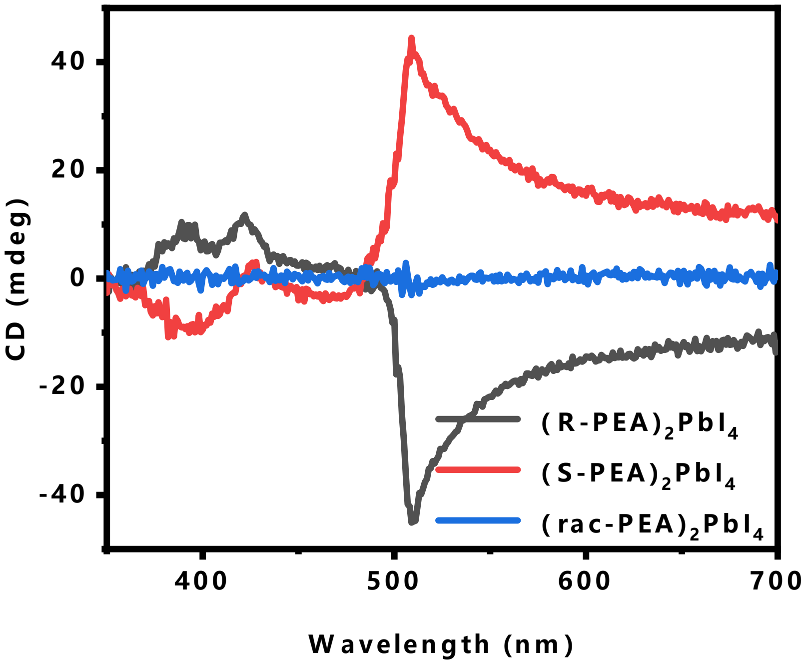

Figure S3. CD spectra of (R-α-PEA)$_2$PbI$_4$, (S-α-PEA)$_2$PbI$_4$ and (rac-α-PEA)$_2$PbI$_4$ films.

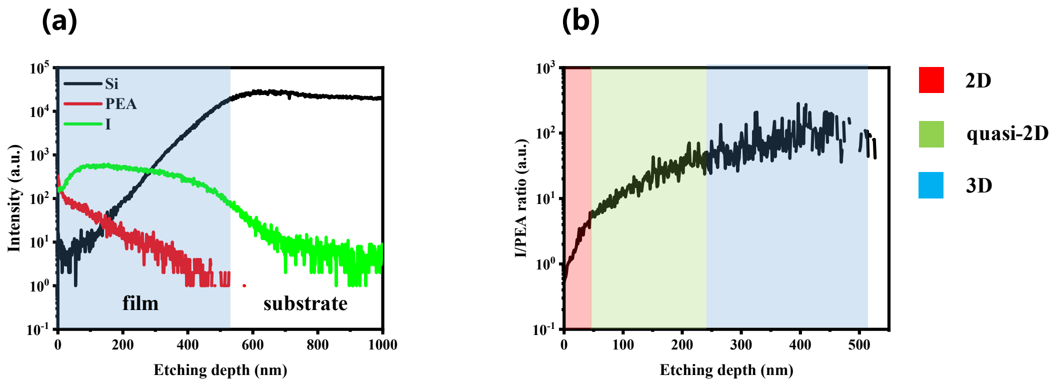

Figure S4. (a) Raw intensity profiles of Si, PEA, I in the 120s-cycle1 sample measured by TOF-SIMS. (b) Relative intensity ratio of I/PEA with different etching times.

Based on the I to PEA relative ratio as a function of etching depth shown above, the phase distribution along the vertical direction can be divided into three regions. The quasi-2D phases with small n values are mainly presented on the surface of the films with the thickness of ~50 nm, while the next is the transition zone from small to large n phases with the thickness of about 200 nm. And the bottom part of the film shows large and nearly constant I/PEA ratio, which can be assigned to the 3D perovskite phase with the thickness of around 250 nm.

Figure S5. The optical microscope images of chiral-2D/3D films spin-coated with 1-5 cycles of (R-α-PEA)I solution.

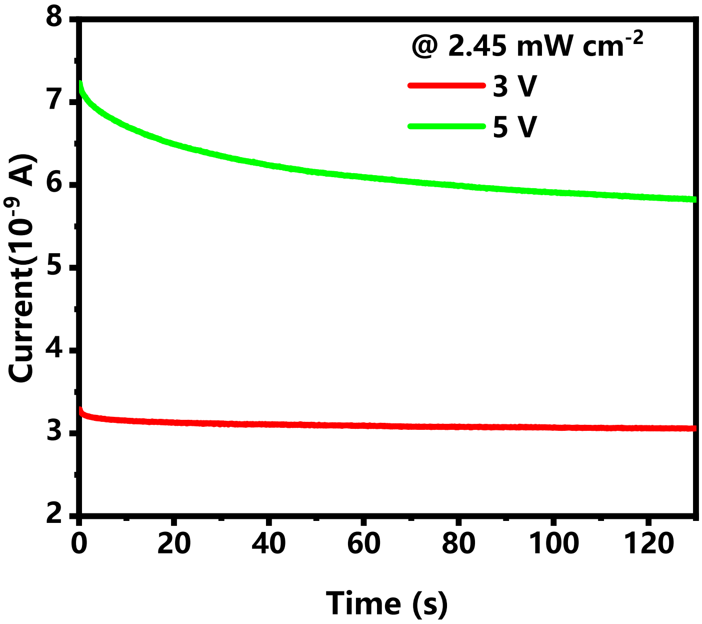

Figure S6. The photocurrent output of chiral-2D/3D detectors under the bias of 3 V and 5 V, respectively, with the incident light intensity of 2.45 mW cm^{-2}.

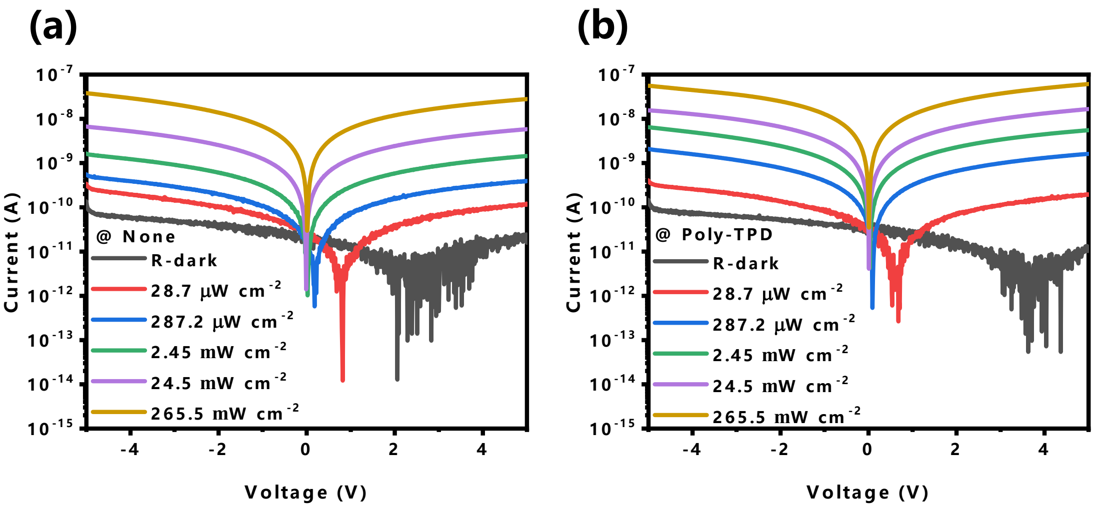

Figure S7. (a, b) *I*-*V* curves of the devices without (a) and with Poly-TPD interlayer (b) under illumination of a 505 nm laser with different intensities at a bias of 3 V.