Direct Observation of Competition between Amplified Spontaneous Emission and Auger Recombination in Quasi-Two-Dimensional Perovskites

Minghuan Cui[†#], Chaochao Qin[†#], Yuanzhi Jiang[‡], Mingjian Yuan[‡], Liuhong Xu[†], Didi Song[†], Yuhai Jiang[§]* & Yufang Liu[†]**

[†]Henan Key Laboratory of Infrared Materials & Spectrum Measures and Applications, Henan Normal University, Xinxiang 453007, P. R. China

[‡]Key Laboratory of Advanced Energy Materials Chemistry (Ministry of Education), College of Chemistry, Nankai University, Tianjin 300071, P. R. China

[§]Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201210, P. R. China

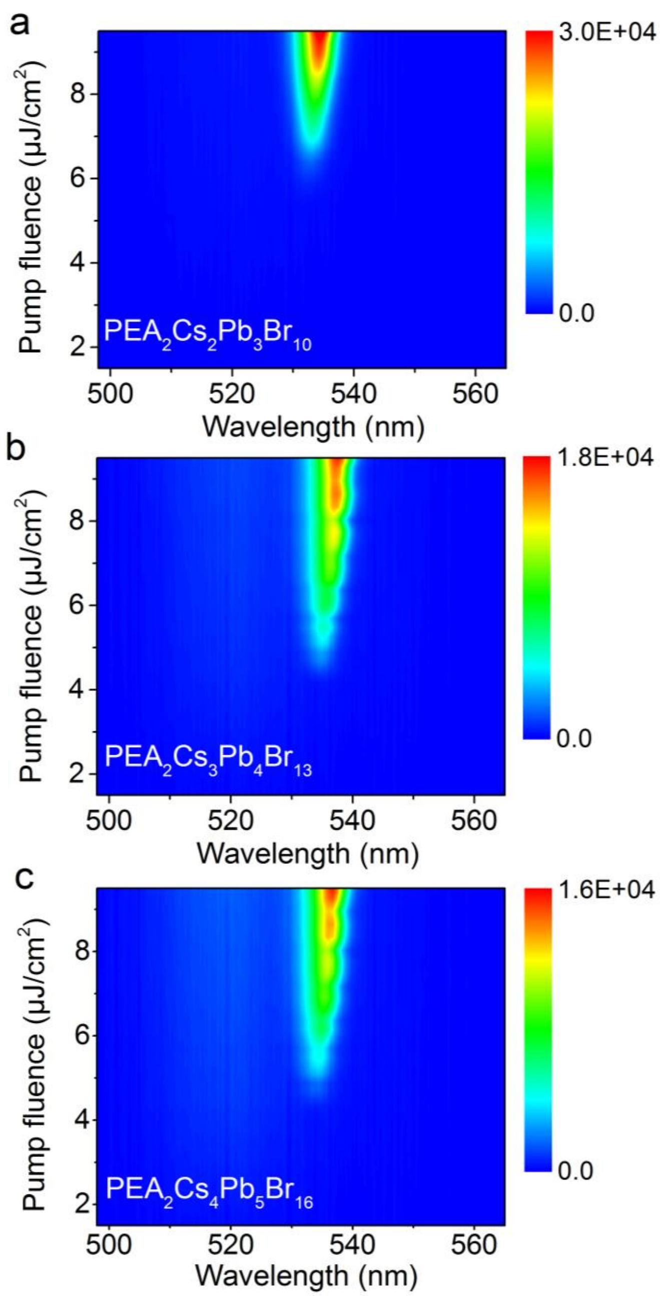

Fig. S1 Vis-pseudocolor representation of pump-fluence dependence of the emission from **a** $(PEA)_2Cs_2Pb_3Br_{10}$ films (<*n*> = 3) **b** $(PEA)_2Cs_3Pb_4Br_{13}$ films (<*n*> = 4) and **c** $(PEA)_2Cs_4Pb_5Br_{16}$ films (<*n*> = 5) under 365 nm laser pulse excitation.

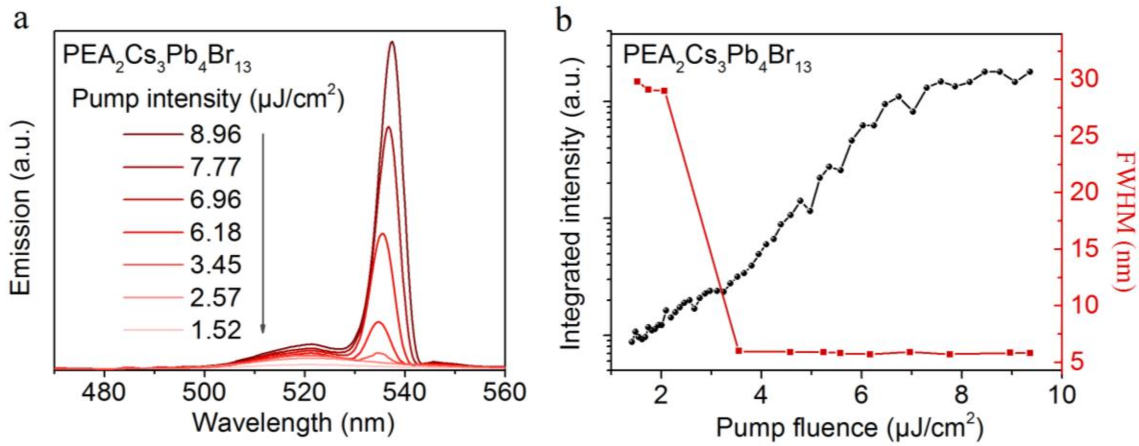

Fig. S2 a Pump-fluence dependence of the emission from $(PEA)_2Cs_3Pb_4Br_{13}$ films (<*n*> = 4) under 365 nm laser pulse excitation. **b** The FWHM of the emission peak with the change of pump fluence shows abrupt narrowing and integrated PL and ASE intensity with the change of pump fluence reveals threshold property of $(PEA)_2Cs_3Pb_4Br_{13}$ films (<*n*> = 4).

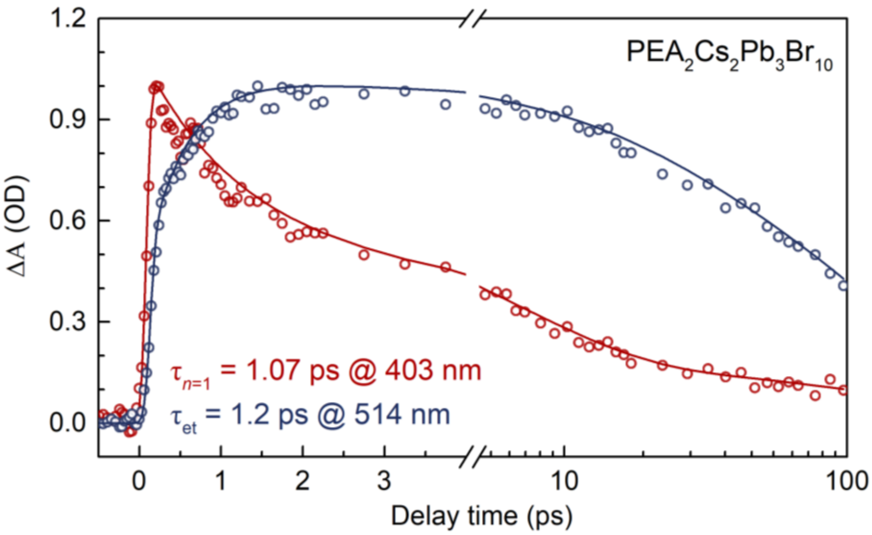

Fig. S3 Normalized GSB kinetics for <*n*> = 3 films at 403 and 513 nm corresponding to $n = 1$ and $n \geq 5$ QWs, respectively. Excitation, 365 nm (1 kHz, 100 fs); pump fluence, 101.9 μJ cm^{-2}.

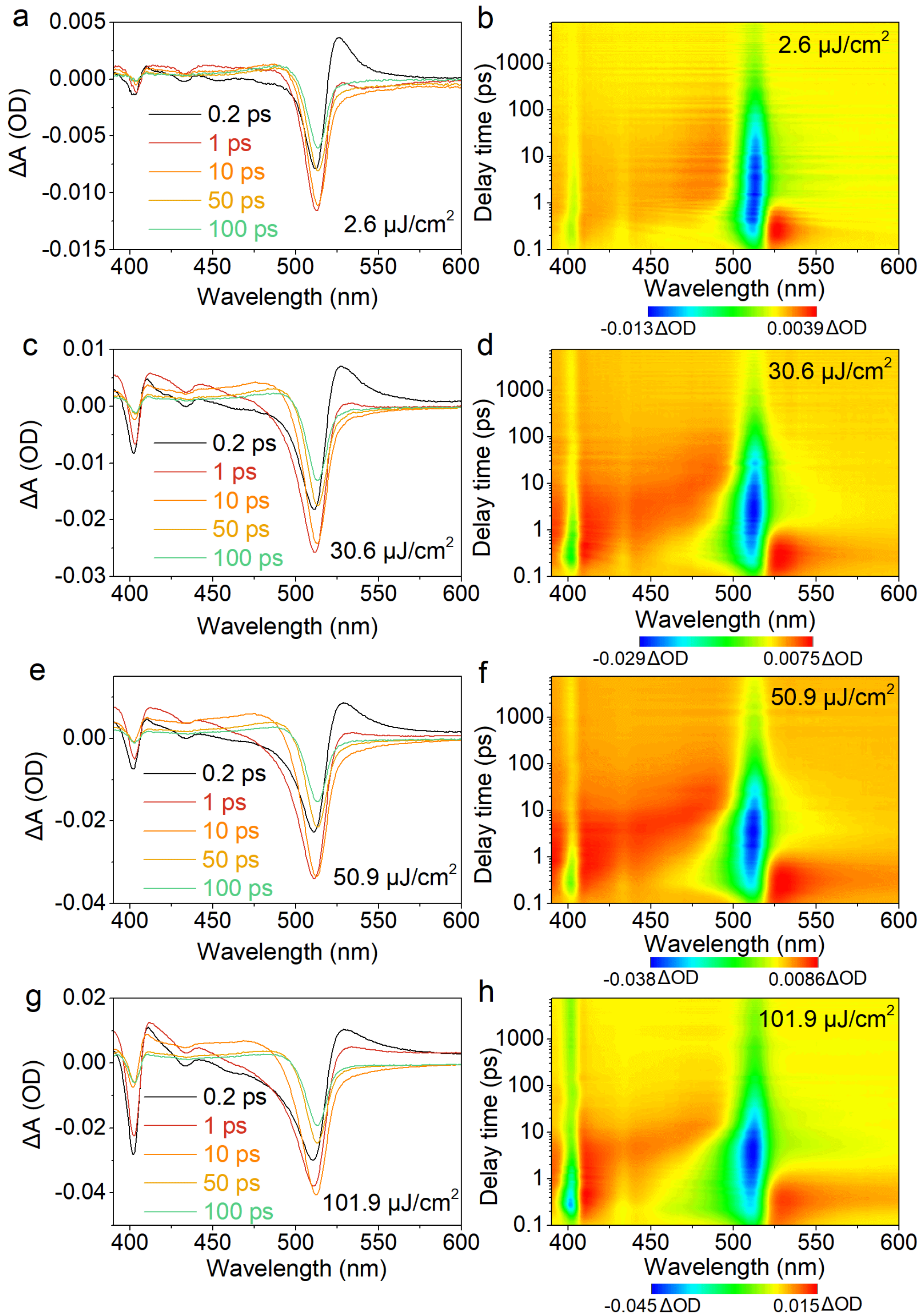

Fig. S4 TA measurements for $(PEA)_2Cs_2Pb_3Br_{10}$ films (<*n*> = 3) thin films at different pump fluence. a, c, e, g TA spectra at selected timescales and **b, d, f, h** Vis-pseudocolor representation of TA spectra at 2.6, 30.6, 50.9 and 101.9 μJ cm^{-2}.

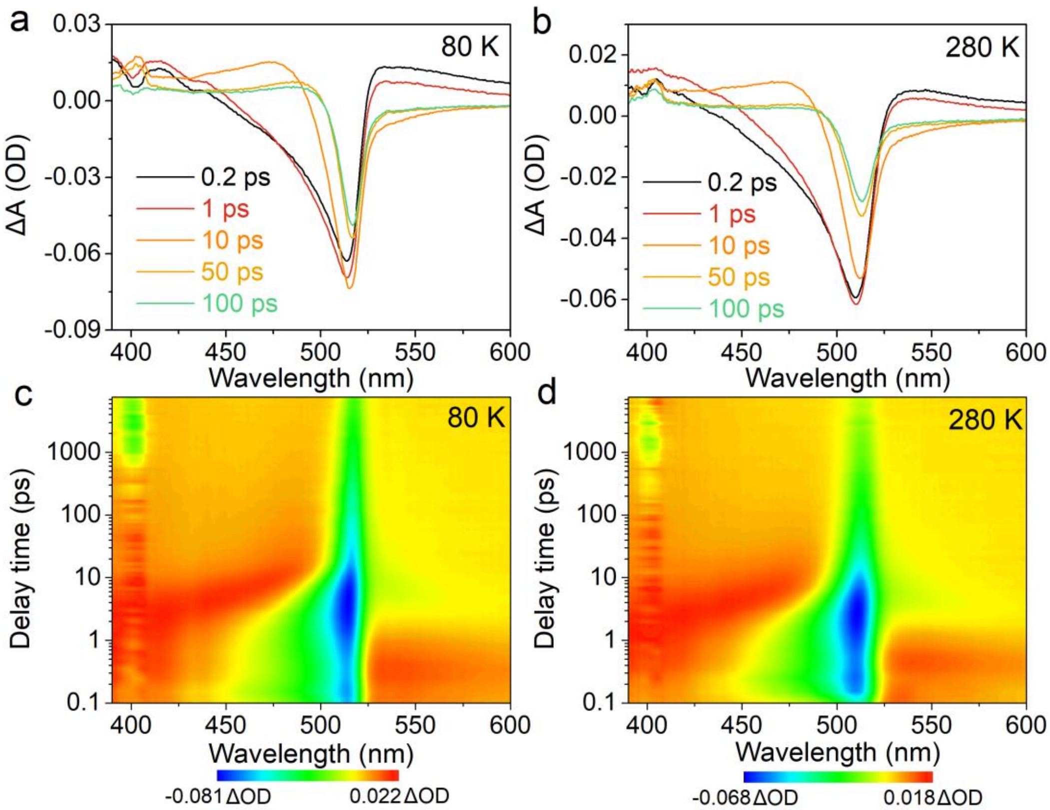

Fig. S5 TA measurements for $(PEA)_2Cs_4Pb_5Br_{16}$ films (<*n*> = 5) thin films at different temperatures. a, c TA spectra at selected timescales and **b, d** Vis-pseudocolor representation of TA spectra at 80 and 280 K.

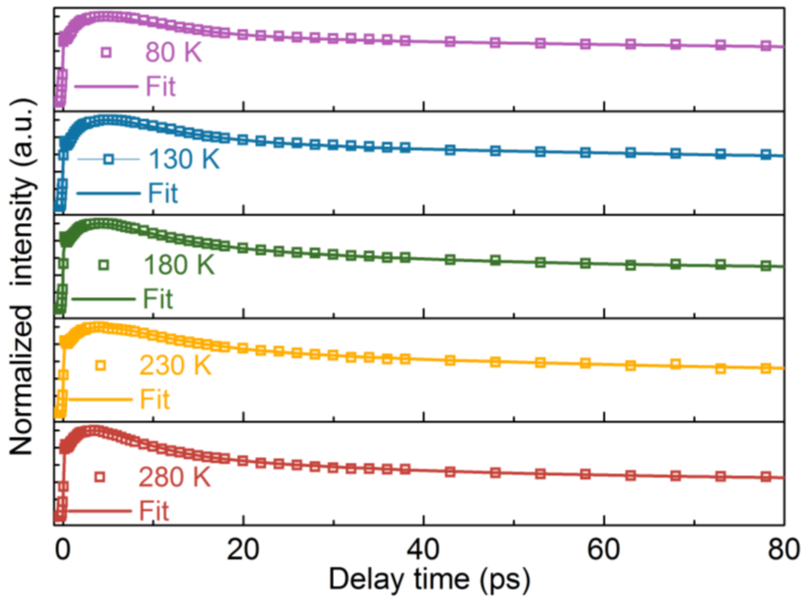

Fig. S6 Comparison of $n \geq 5$ QWs bleach recovery kinectics of $<n> = 5$ quasi-2D RPPs films at indicated temperature. Kinetics traces have been normalized to the same value at the maximum.

Table S1. Fitting parameters of the TA kinetics of <*n*> = 3 quasi-2D RPP films ($n \geq 5$ QWs).

Pump fluence	τ_{et}/ps (Weights/%)	τ_1/ps (Weights/%)	τ_2/ps (Weights/%)
2.6 μJ cm^{-2}	0.2 ± 0.06 (100)	--	72.3 ± 19 (100)
30.6 μJ cm^{-2}	0.9 ± 0.21 (100)	6.7 ± 2.2 (17.4)	65.2 ± 9.5 (82.6)
50.9 μJ cm^{-2}	1.2 ± 0.11 (100)	9.5 ± 1.8 (20.3)	53.5 ± 6.7 (79.7)
101.9 μJ cm^{-2}	1.2 ± 0.3 (100)	9.5 ± 2.1 (32.2)	50.8 ± 6.3 (67.8)

Table S2. Fitting parameters of the TA kinetics of <*n*> = 5 quasi-2D RPP films (*n* ≥ 5 QWs) at the indicated temperatures.

Temperatures	τ_{et}/ps (Weights/%)	τ_1/ps (Weights/%)	τ_2/ps (Weights/%)
80 K	2.6 ± 0.6 (100)	7.8 ± 0.71 (69.6)	164.2 ± 12.8 (30.4)
130 K	2.5 ± 0.51 (100)	8.0 ± 1.2 (68.8)	132.8 ± 14.3 (31.2)
180 K	1.9 ± 0.38 (100)	9.9 ± 2.3 (68.4)	111.9 ± 13.7 (31.6)
230 K	1.2 ± 0.25 (100)	8.4 ± 1.5 (59.2)	95.6 ± 5.5 (40.8)
280 K	1.3 ± 0.3 (100)	8.4 ± 0.8 (32.2)	78.5 ± 7.7 (67.8)

Table S3. Fitting parameters of the TA kinetic curves of the <*n*> = 3–5 quasi-2D RPP films (*n* ≥ 5 QWs) at a 101.9 μJ/cm^2 excitation fluence.

<*n*>	τ_{et}/ps (Weights/%)	τ_1/ps (Weights/%)	τ_2/ps (Weights/%)
<*n*>=3	1.2 ± 0.3 (100)	9.5 ± 2.1 (32.2)	50.8 ± 6.3 (67.8)
<*n*>=4	1.2 ± 0.32 (100)	7.3 ± 0.89 (38.1)	61.6 ± 3.4 (61.9)
<*n*>=5	1.4 ± 0.39 (100)	7.3 ± 0.6 (43.4)	76.1 ± 5.1 (56.6)