Supplementary Information:

Unveiling hot carrier relaxation and carrier transport mechanisms in quasi-two-dimensional layered perovskites

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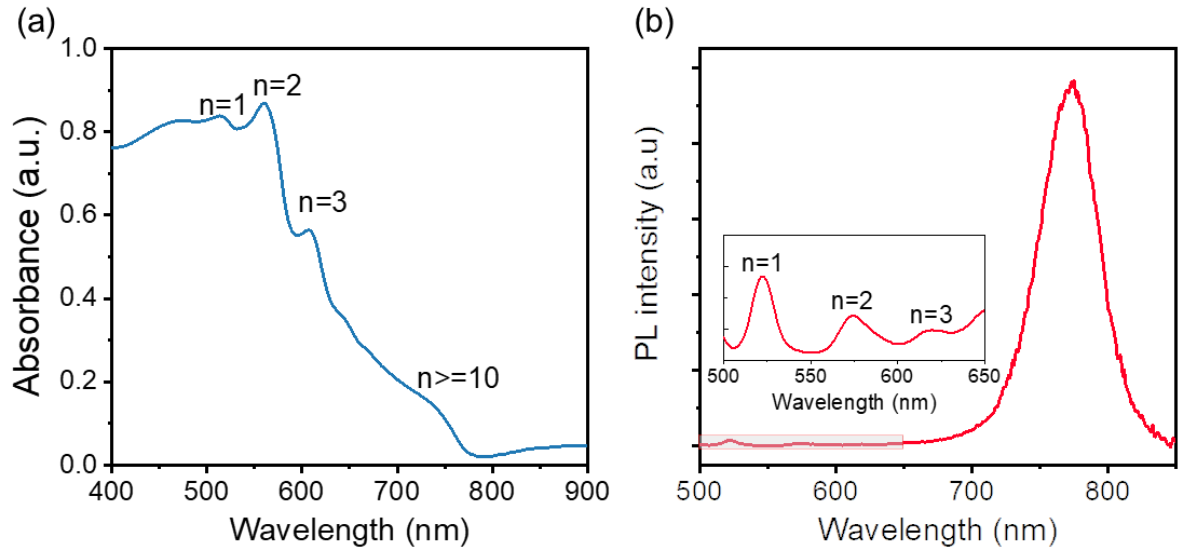

Figure S1. Steady-state (a) absorption and (b) photoluminescence (PL) spectra of $(PEA)_2(MA)_2Pb_3I_{10}$ 2D layered perovskite film. The PL is excited from the glass substrate side under excitation at 460 nm.

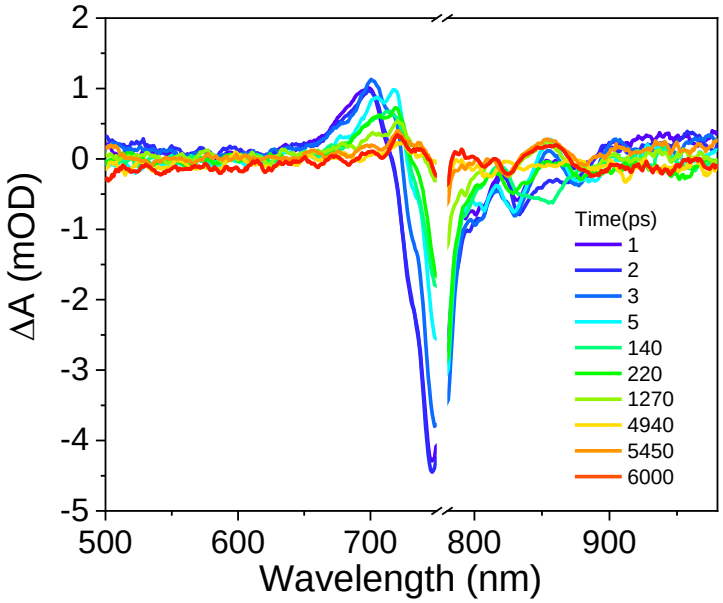

Figure S2. Transient absorption spectra of $(PEA)_2(MA)_2Pb_3I_{10}$ excited from the front face (air side) under 770 nm excitation (The scattering signal due to excitation light is removed for clarity).

Table S1. Fitting parameters of TA kinetics under various excitation wavelengths.

λ_{exc}	λ_{probe}	τ_0, ps	A_0	τ_1, ps	A_1	τ_2, ps	A_2	τ_3, ps	A_3
400 nm	525 nm (n=1)	0.22 ±0.04	54%	2.0 ±1	23%	11 ±4	23%	--	--
	573 nm (n=2)	0.40 ±0.01	25%	9.0 ±0.3	35%	100 ±4	29%	2600 ±200	11%
	617 nm (n=3)	0.20 ±0.002	-100%	13 ±0.6	39%	107 ±9	39%	850 ±90	22%
	760 nm (n>=10)	0.53 ±0.08	-100%	9.0 ±0.8	-23%	280 ±30	38%	2100 ±200	62%
530 nm	573 nm (n=2)	0.32 ±0.08	15%	9.7 ±0.8	50%	130 ±20	35%	--	--
	617 nm (n=3)	0.56 ±0.03	26%	20 ±1	36%	280 ±20	38%	--	--
	760 nm (n>=10)	0.46 ±0.01	-100%	17 ±3	-20%	--	--	1000 ±200	38%
570 nm	573 nm (n=2)	0.38 ±0.04	-12%	4.8 ±0.3	32%	46 ±2	50%	590 ±50	18%
	617 nm (n=3)	0.26 ±0.08	34%	4.5 ±0.3	17%	52 ±2	33%	650 ±40	16%
	760 nm (n>=10)	0.41 ±0.01	-89%	5.1 ±0.8	-11%	260 ±10	55%	4200 ±200	45%
610 nm	573 nm (n=2)	0.06 ±0.01	-100%	--	--	230 ±20	100%	--	--
	617 nm (n=3)	0.30 ±0.02	30%	5.9 ±0.3	31%	97 ±6	31%	4000 ±800	8%
	760 nm (n>=10)	--	--	2.0 ±0.8	-80%	--	--	15000 ±1000	100%

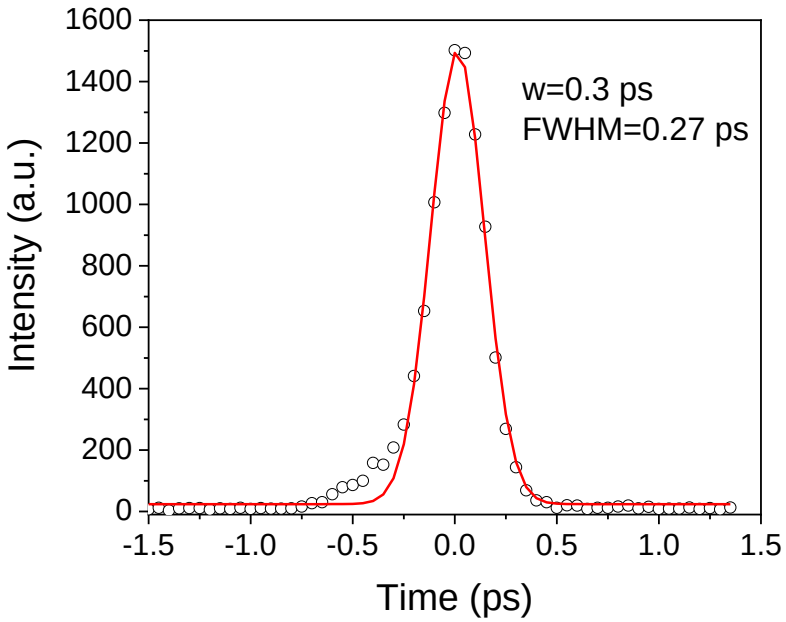

Figure S3. Instrument response function (IRF) of fluorescence up-conversion setup determined from the Gaussian fit of Raman scattering signal in ethanol solvent.

Table S2. Fitting parameters of time-resolve PL kinetics at different emission wavelengths measured via femtosecond fluorescence up-conversion under 400 nm excitation.

λ_{PL}	τ_0, ps	A_0	τ_1, ps	A_1	τ_2, ps	A_2	τ_3, ps	A_3	τ_4, ps	A_4
522 nm (n=1)	0.39 ± 0.05	-100%	2.5 ±0.4	73%	18 ±3	27%	--	--	--	--
573 nm (n=2)	0.22 ±0.04	-100%	3.0 ±0.4	57%	31 ±8	31%	250 ±100	12%	--	--
620 nm (n=3)	0.23 ±0.06	-100%	3.5 ±0.7	53%	47 ±17	30%	370 ±160	31%	--	--
720 nm (n>=10)	IRF	-16%	2.4 ±0.8	-35%	--	--	250 ±60	-49%	25000 fixed	100%

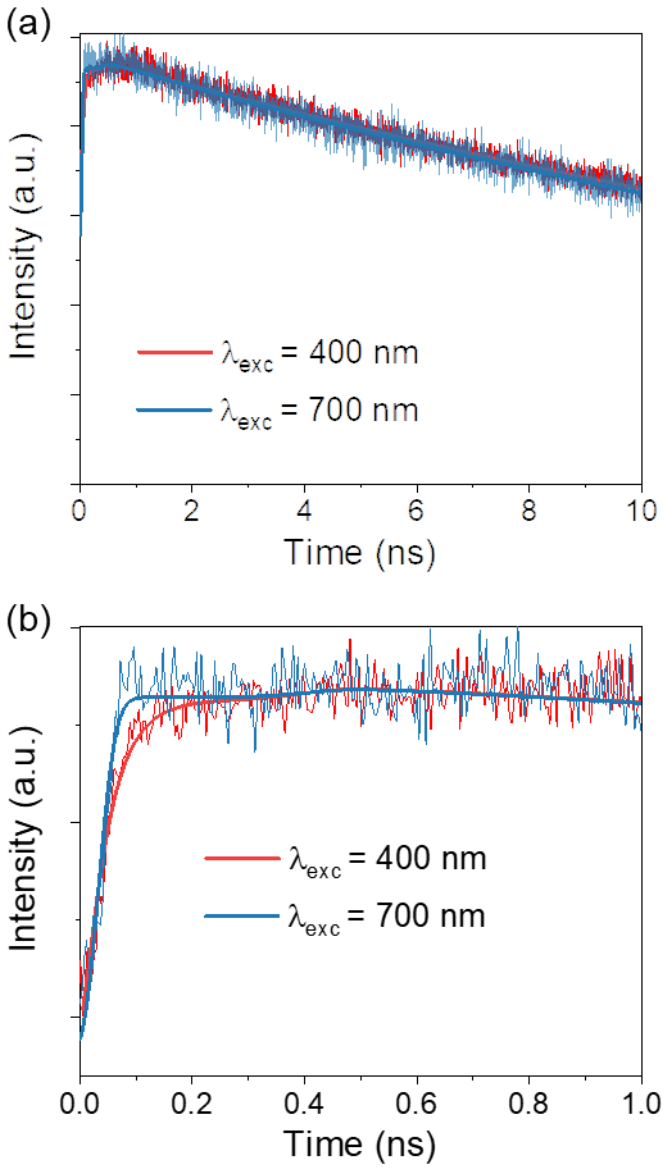

Figure S4. Time-resolved PL kinetics of $(PEA)_2(MA)_2Pb_3I_{10}$ at ns scale at 750 nm measured by time correlated singlet photon counting (TCSPC) technique under excitation at 400 nm (red) and 700 nm (blue).