

Supporting Information

Formation of Intermediate-phase Free Ultrasmooth Quasi-two-dimensional Perovskite Films for High Quality Lasers

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Table. S1 The relative polarity and boiling points of the antisolvents.^{1,2}

Antisolvent	Relative polarity	Boiling point (°C)
EA	0.228	77
CB	0.188	132
DE	0.117	34.5
DIE	0.110	69
TOL	0.100	118

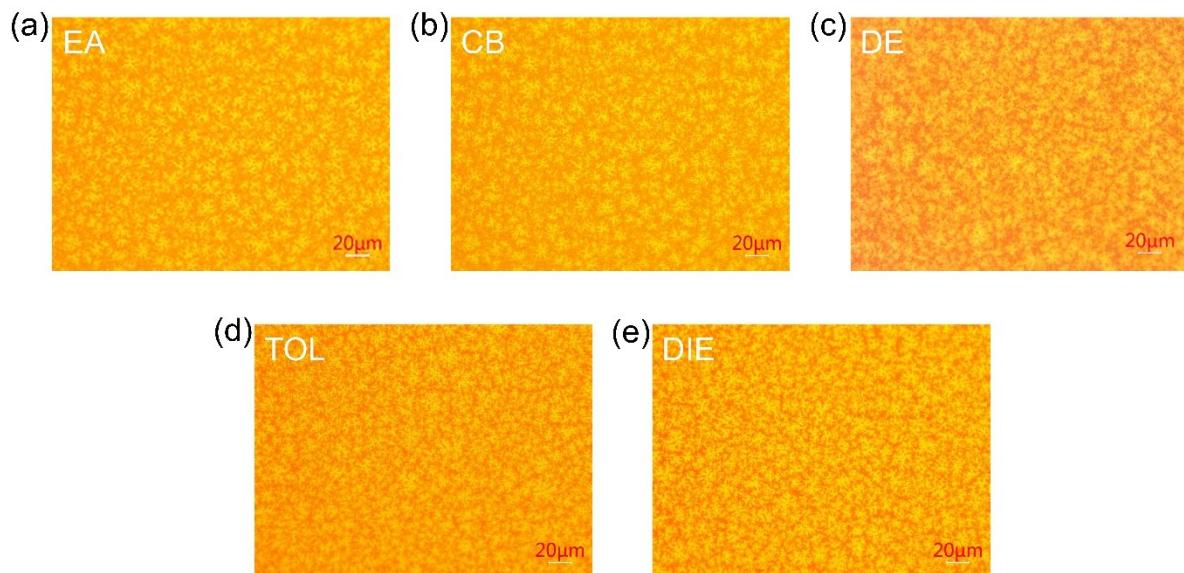


Fig. S1 Optical microscope images of the P2F8 films: (a) EA-film; (b) CB-film; (c) DE-film; (d) TOL-film; (e) DIE-film.

Table S2. Single fitting parameters of TRPL traces for the P2F8 films.

Antisolvent	EA	CB	DE	TOL	DIE
A	0.28±0.002	0.21±0.002	0.21±0.002	0.26±0.003	0.27±0.003
t (ns)	1.11±0.02	1.18±0.02	1.19±0.02	1.21±0.02	1.24±0.02

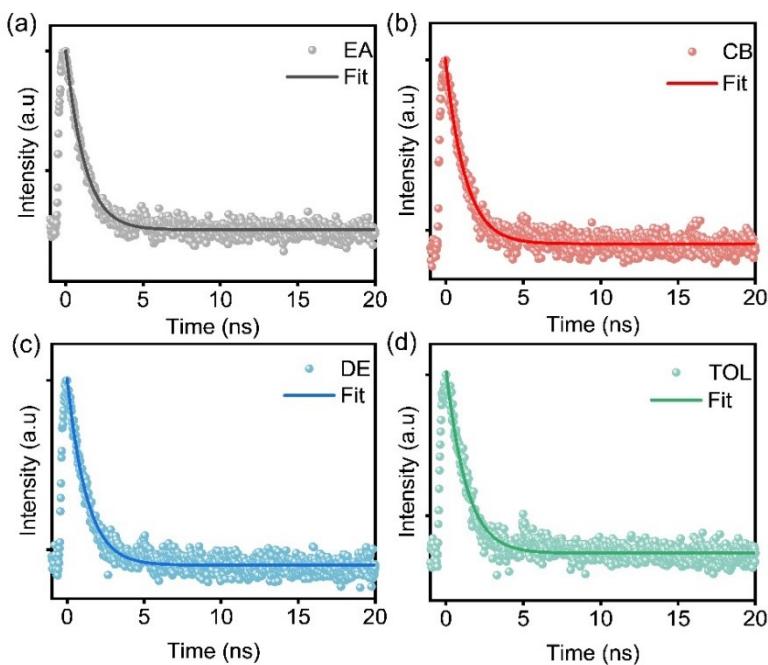


Fig. S2 TRPL spectra of the P2F8 films: (a) EA-film; (b) CB-film; (c) DE-film; (d) TOL-film.

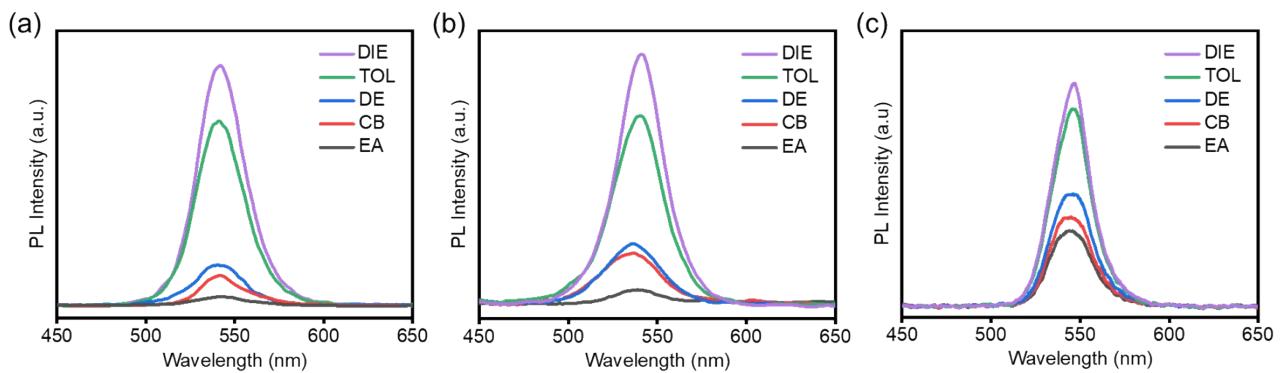


Fig. S3 (a) PL spectra under CW laser excitation at a pump density of 19.9 W cm^{-2} . (b) PL spectra under ps pulsed laser excitation at a pump density of $1.3 \mu\text{J cm}^{-2}$. (c) PL spectra under fs pulsed laser excitation at a pump density of $7.4 \mu\text{J cm}^{-2}$.

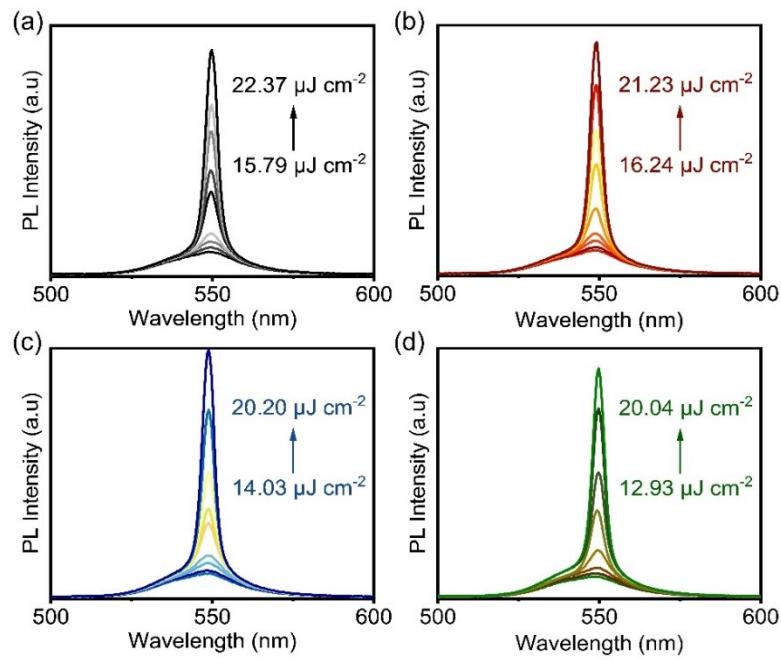


Fig. S4 PL emission of the P2F8 films at different pump densities: (a) EA-film; (b) DE-film; (c) CB-film; (d) TOL-film.

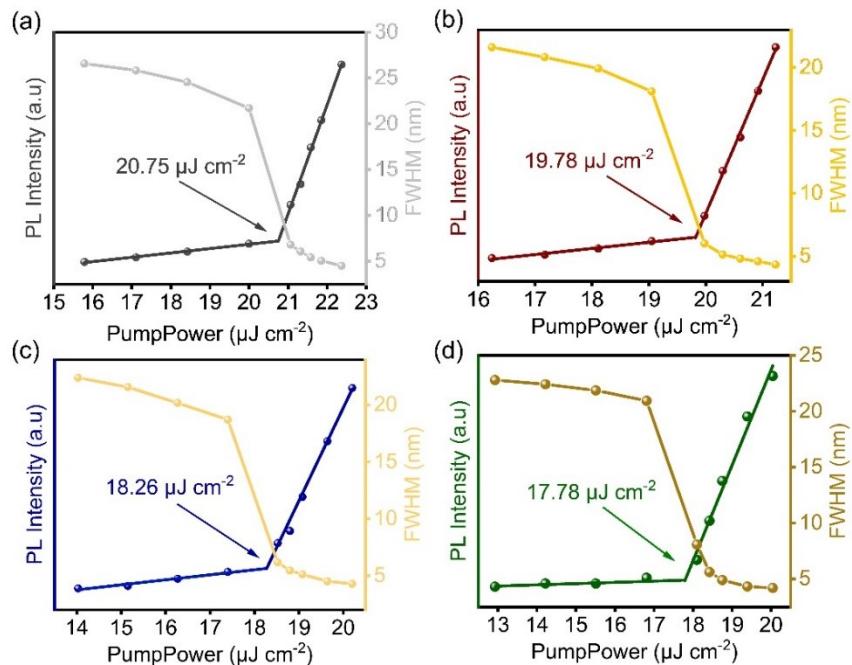


Fig. S5 FWHM and Light output intensity of PL emission of the P2F8 films: (a) EA-film; (b) CB-film; (c) DE-film; (d) TOL-film.

SUPPLEMENTARY REFERENCES

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2. L. Wang, X. Wang, L.-L. Deng, S. Leng, X. Guo, C.-H. Tan, W. C. H. Choy and C.-C. Chen, *Mater. Horiz.*, 2020, **7**, 934-942.