

**Supplementary Information for:**

# High-Efficiency, Flexible and Large-area Red/Green/Blue All-Inorganic Metal Halide Perovskite Quantum Wires-Based Light-Emitting Diodes

*Yang Bryan Cao<sup>1,2,3</sup>, Daquan Zhang<sup>1,2,3</sup>, Qianpeng Zhang<sup>1,2,3</sup>, Xiao Qiu<sup>1,2,3</sup>, Yu Zhou<sup>1,2,3</sup>, Swapnadeep Poddar<sup>1,2,3</sup>, Yu Fu<sup>1,2,3</sup>, Yudong Zhu<sup>1,4</sup>, Jin-Feng Liao<sup>5</sup>, Lei Shu<sup>1,2,3</sup>, Beita Ren<sup>1,2,3</sup>, Yucheng Ding<sup>1,2,3</sup>, Bing Han<sup>4</sup>, Zhubing He<sup>4</sup>, Dai-Bin Kuang<sup>5</sup>, Kefan Wang<sup>6</sup>, Haibo Zeng<sup>7\*</sup>, Zhiyong Fan<sup>1,2,3\*</sup>*

<sup>1</sup>Department of Electronic & Computer Engineering, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong SAR, China.

<sup>2</sup>State Key Laboratory of Advanced Display and Optoelectronics Technologies HKUST, Clear Water Bay, Kowloon, Hong Kong SAR, China.

<sup>3</sup>Guangdong-Hong Kong-Macau Joint Laboratory for Intelligent Micro-Nano Optoelectronic Technology, HKUST, Clear Water Bay, Kowloon, Hong Kong SAR, China.

<sup>4</sup>Department of Materials Science and Engineering, Southern University of Science and Technology, No. 1088, Xueyuan Rd., Shenzhen, 518055, Guangdong, P. R. China.

<sup>5</sup>MOE Key Laboratory of Bioinorganic and Synthetic Chemistry, Lehn Institute of Functional Materials, School of Chemistry, Sun Yat-sen University, Guangzhou 510275, P. R. China.

<sup>6</sup> Henan Provinces Key Laboratory of Photovoltaic Materials, Henan University, Kaifeng 475004, Henan, China.

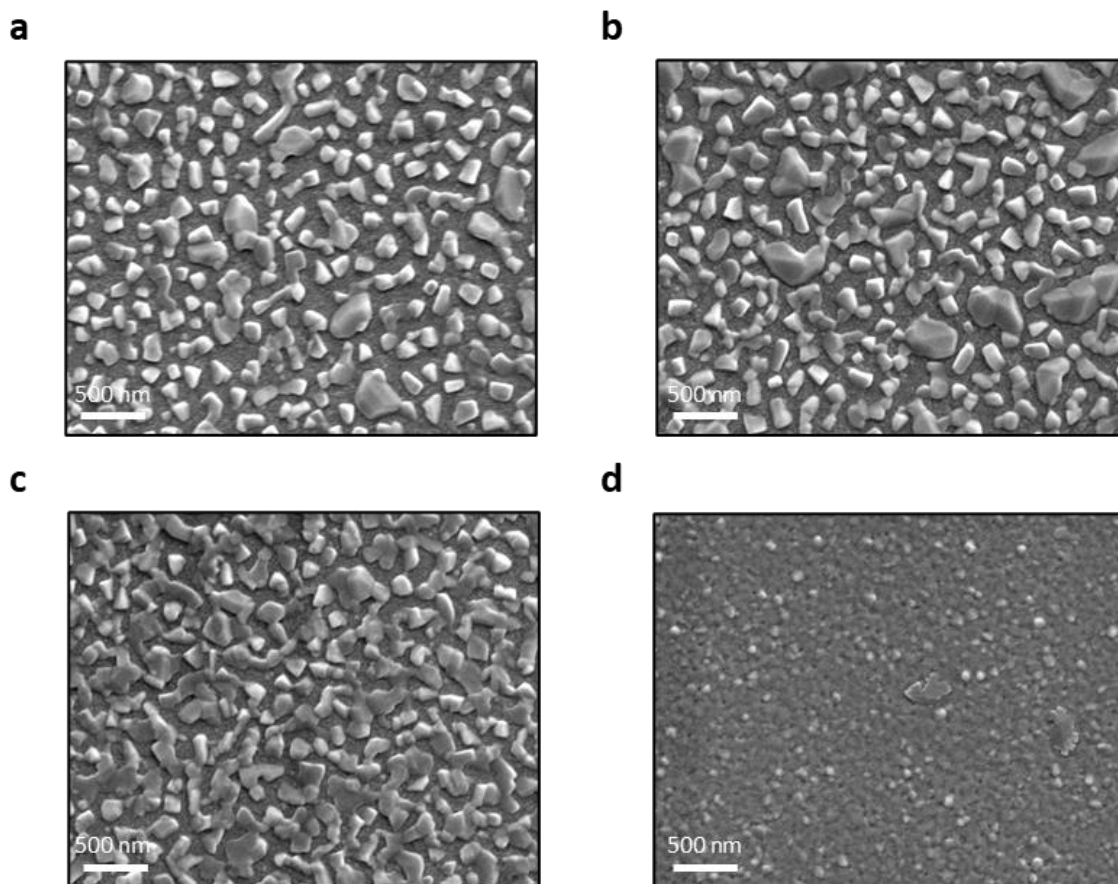
<sup>7</sup>MIT Key Laboratory of Advanced Display Materials and Devices, Institute of Optoelectronics & Nanomaterials, School of Materials Science and Engineering, Nanjing University of Science and Technology, Nanjing 210094, China.

\*Corresponding author: [eezfan@ust.hk](mailto:eezfan@ust.hk) (Z.F.), [zeng.haibo@njust.edu.cn](mailto:zeng.haibo@njust.edu.cn) (H.Z.).

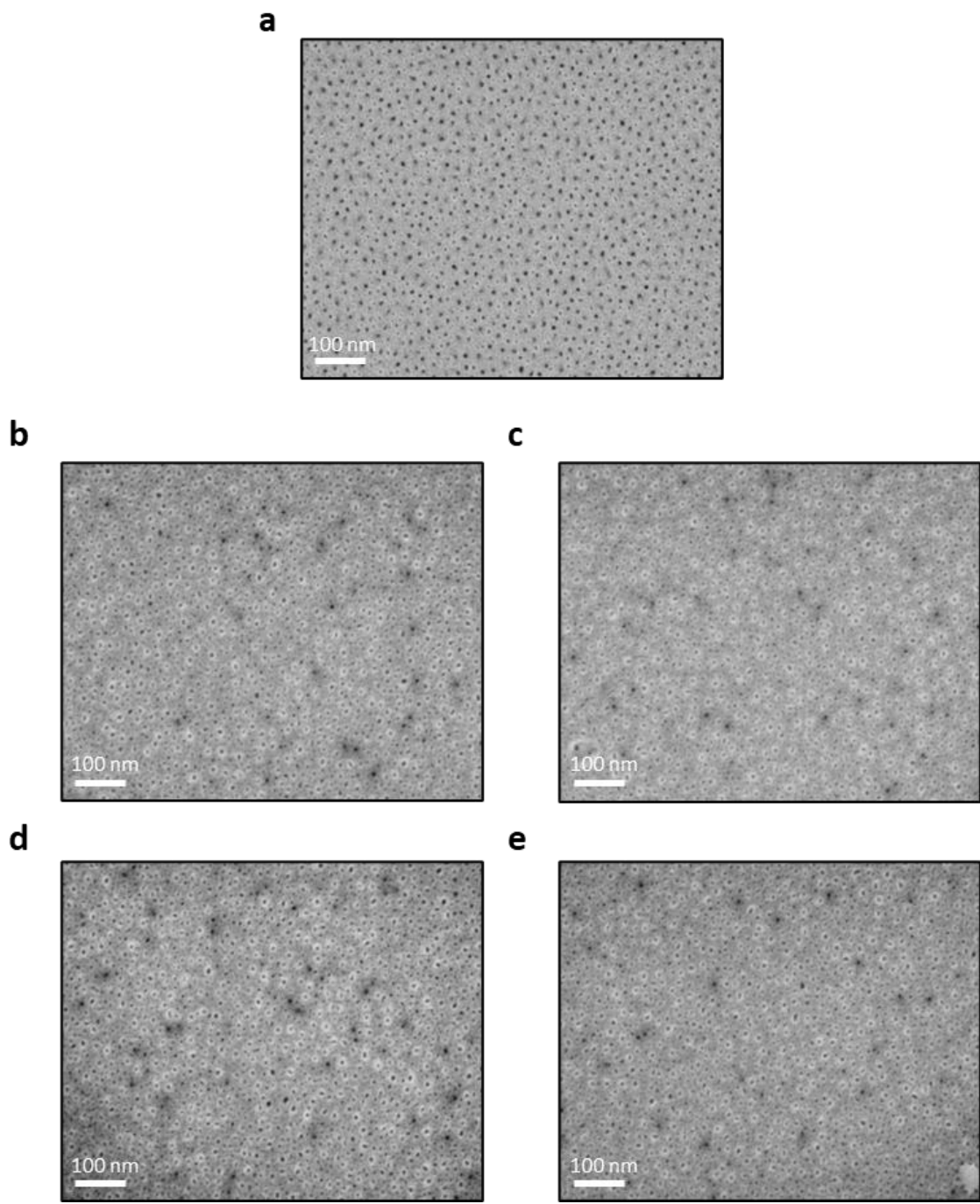
This file include:

Supplementary Figure 1-22

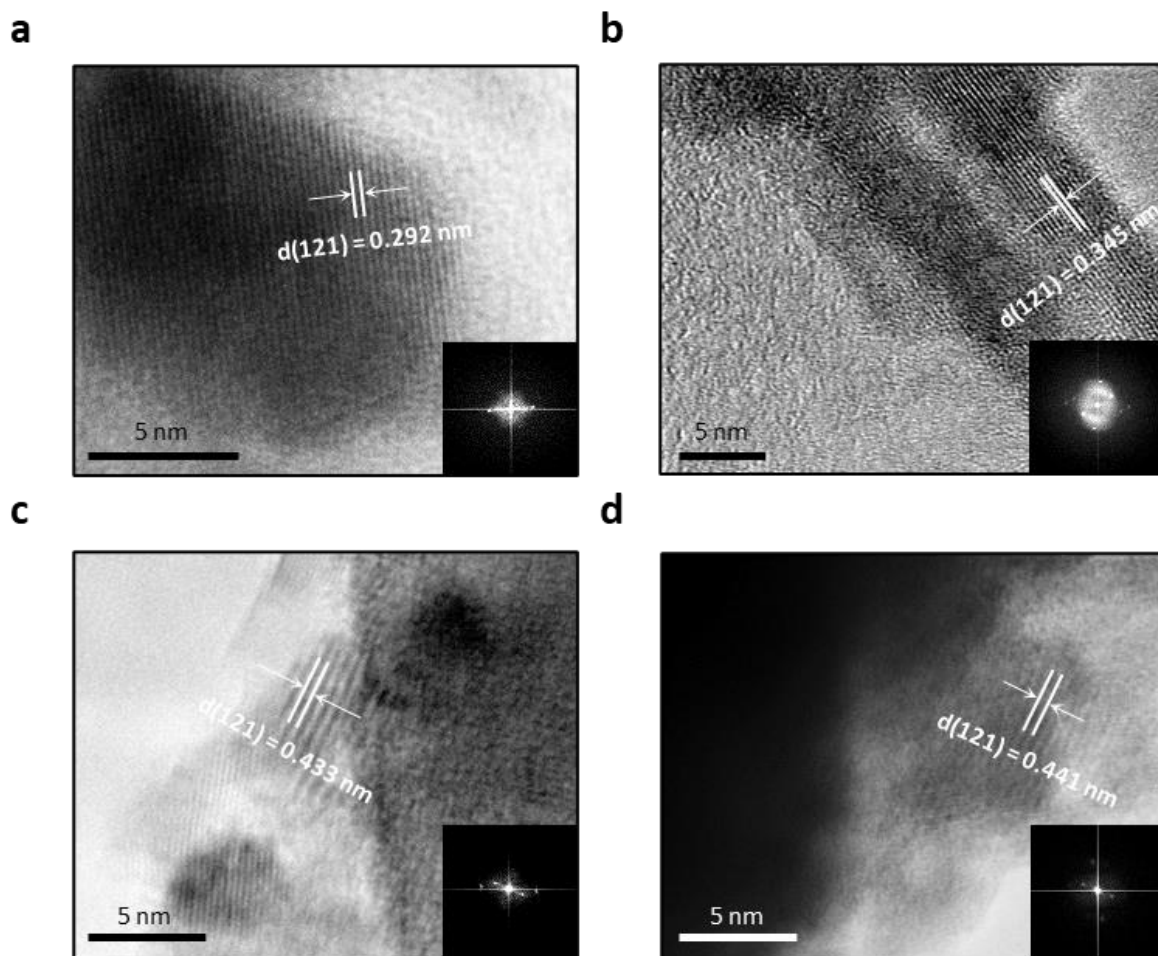
Supplementary Table 1-3



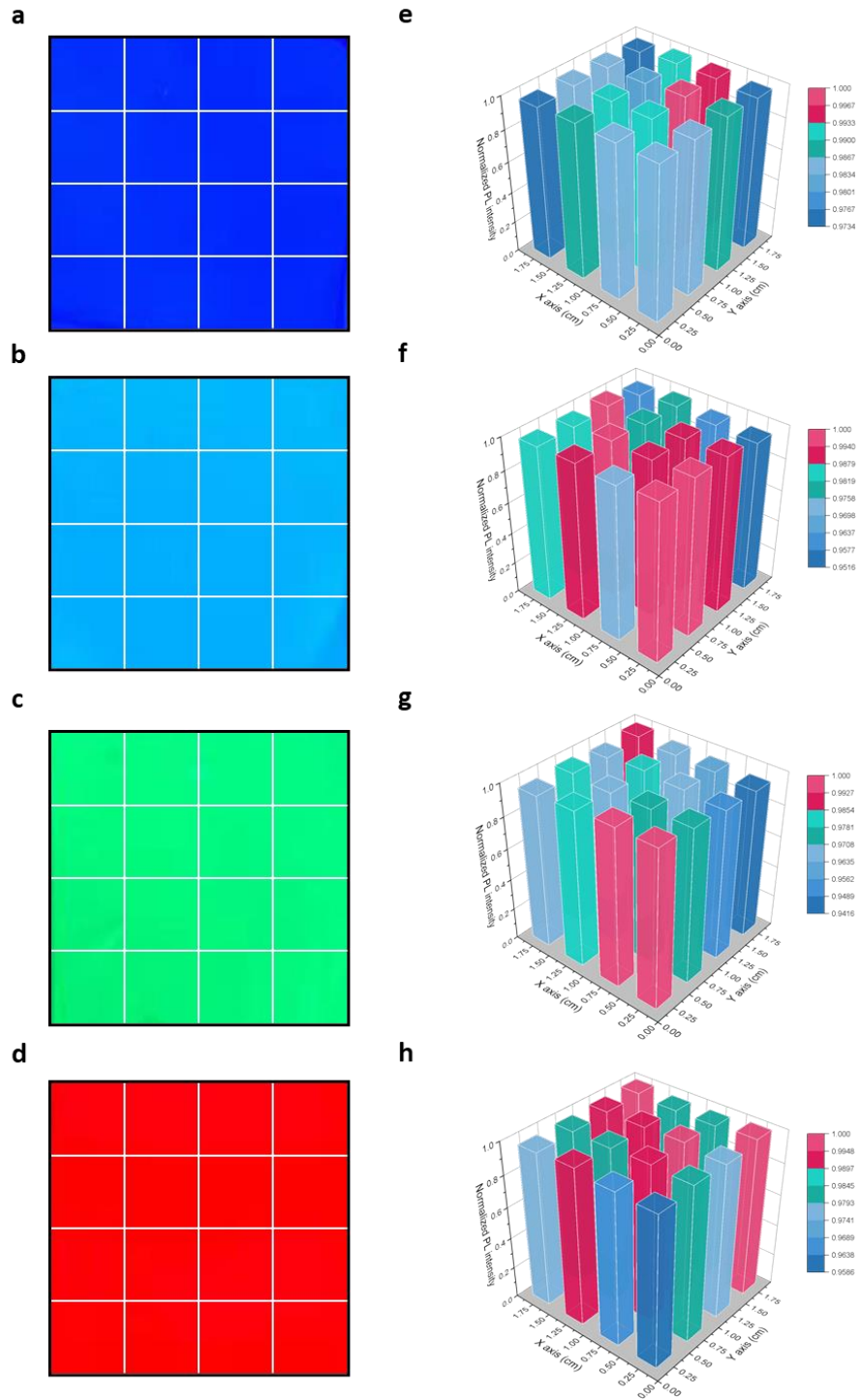
**Supplementary Figure 1. Top-view scanning electron microscopy (SEM) images of perovskite films deposited. a, 6-4 Cl-Br perovskite. b, 4-6 Cl-Br perovskite. c, CsPbBr<sub>3</sub>. d, I-Br perovskite. All the films were deposited on ITO glass.**



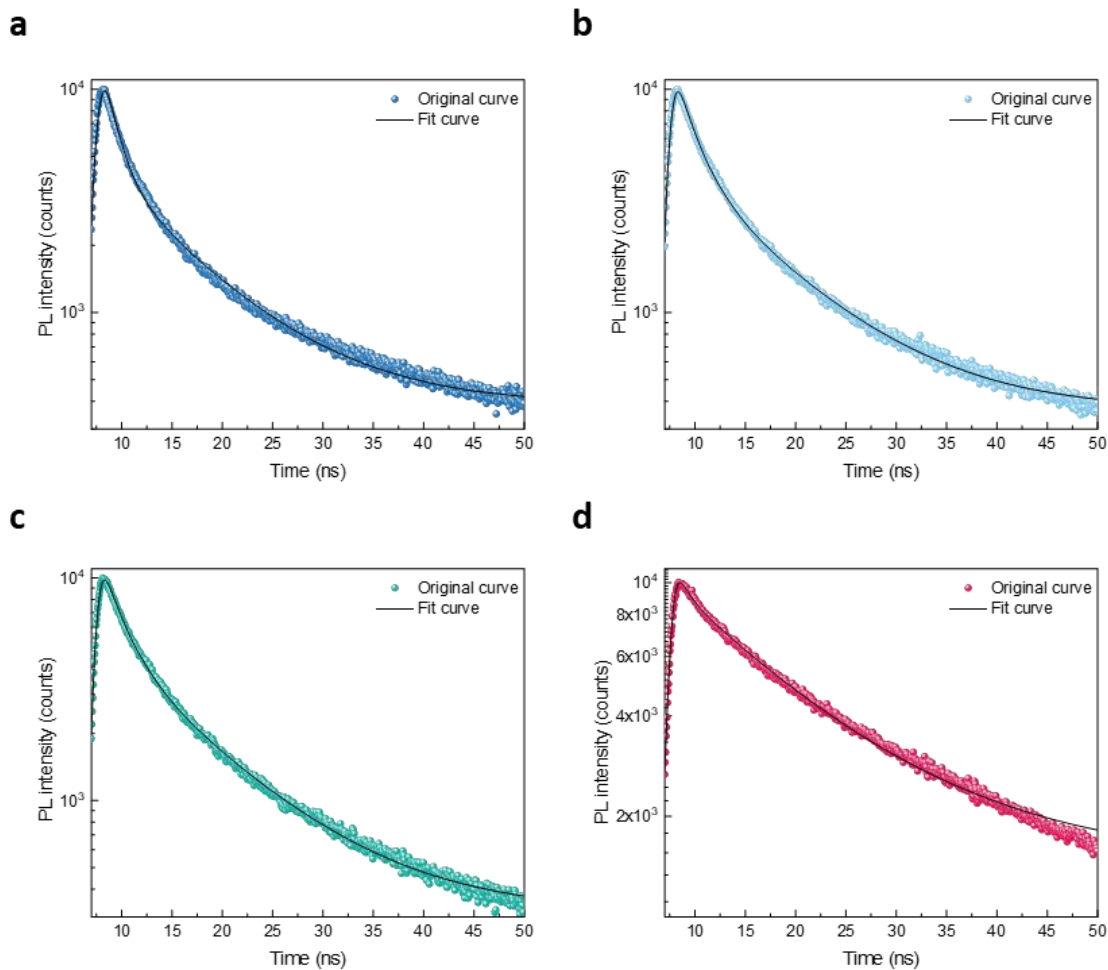
**Supplementary Figure 2. Top-view SEM images of perovskite QWs. a,** blank PAM substrate **b,** 6-4 Cl-Br PeQWs. **c,** 4-6 Cl-Br PeQWs. **d,** CsPbBr<sub>3</sub> QWs. **e,** I-Br PeQWs.



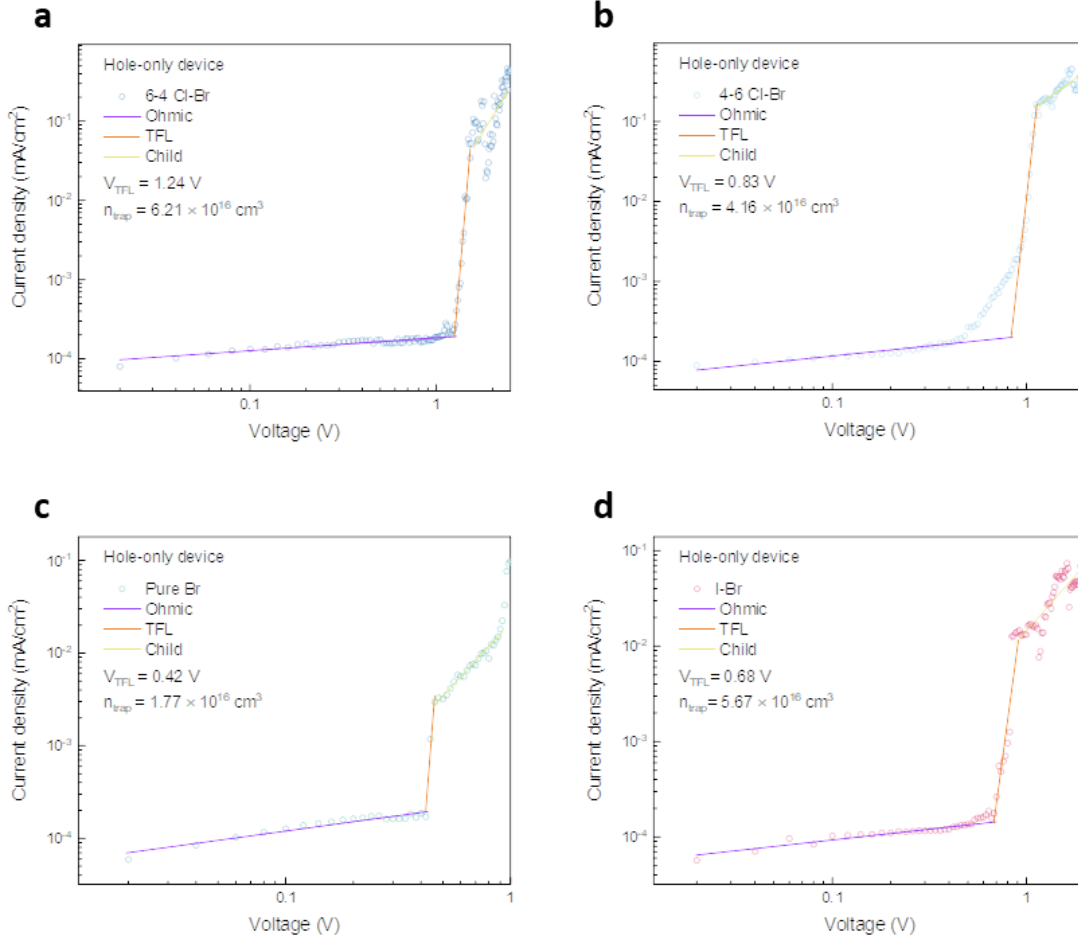
**Supplementary Figure 3. High-resolution transmission electron microscopy (HRTEM) image of perovskite QWs. a, 6-4 Cl-Br perovskite. b, 4-6 Cl-Br perovskite. c, CsPbBr<sub>3</sub>. d, I-Br perovskite.**



**Supplementary Figure 4. Photoluminescent intensity distribution of PeQWs arrays. a-d,** Photograph of blue, sly-blue, green and pure-red light emitting PeQWs with an area of  $2 \times 2 \text{ cm}^2$  arrays. **e-h,** Normalized photoluminescent intensity of the PeQWs arrays with an area of  $2 \times 2 \text{ cm}^2$ . The normalized photoluminescent intensity is presented by a  $4 \times 4$ -pixel 3D column chart.



**Supplementary Figure 5. Photoluminescence lifetime characteristics.** Unfitted and fitted time-resolved photoluminescence decay curves for **a**, 6-4 Cl-Br PeQWs, **b**, 4-6 Cl-Br PeQWs, **c**, CsPbBr<sub>3</sub> QWs and **d**, I-Br PeQWs with and without TAPC.



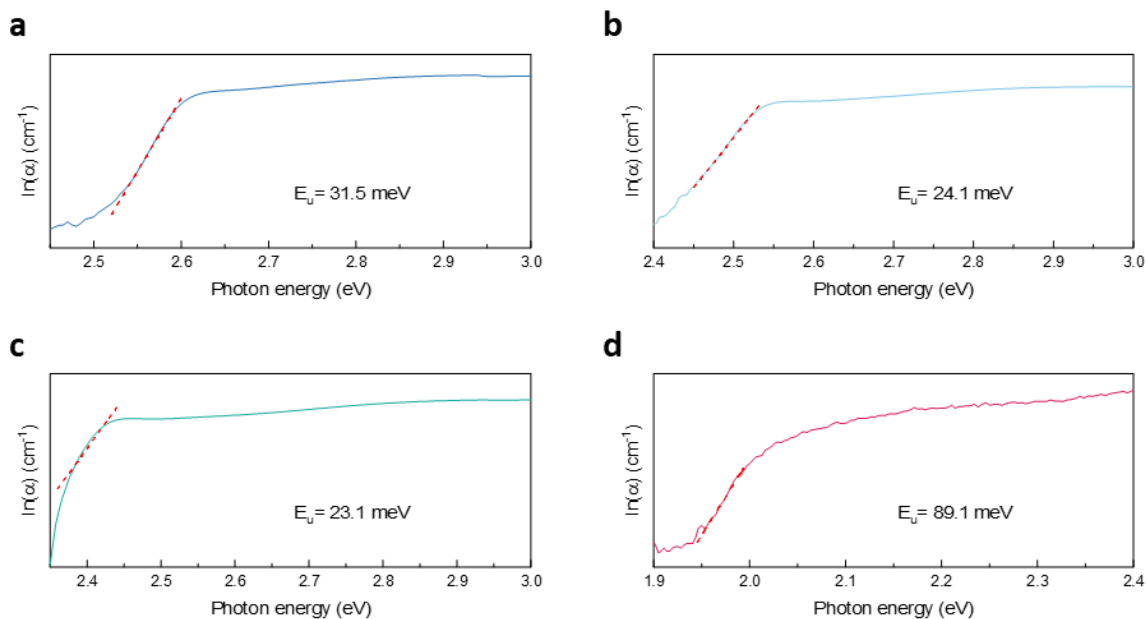
**Supplementary Figure 6. The J–V characteristics of hole devices.** The space-charge-limited current (SCLC) curves of a, 6-4 Cl-Br PeQWs. b, 4-6 Cl-Br PeQWs. c, CsPbBr<sub>3</sub> QWs. d, I-Br PeQWs.

To analyze the hole trap density of our PeQWs, Space charge-limited current (SCLC) measurement using a hole-only device structure is carried out. The total hole trap state density inside the PeQWs is calculated by the equation as below:

$$n_t = \frac{2\varepsilon\varepsilon_0V_{TFL}}{eL^2}$$

where  $n_t$  is the trap state density,  $V_{TFL}$  is the trap-filled limit voltage,  $L$  is the thickness of the perovskites,  $e$  is the elementary charge,  $\varepsilon_0$  and  $\varepsilon$  are the vacuum permittivity and relative permittivity, respectively.



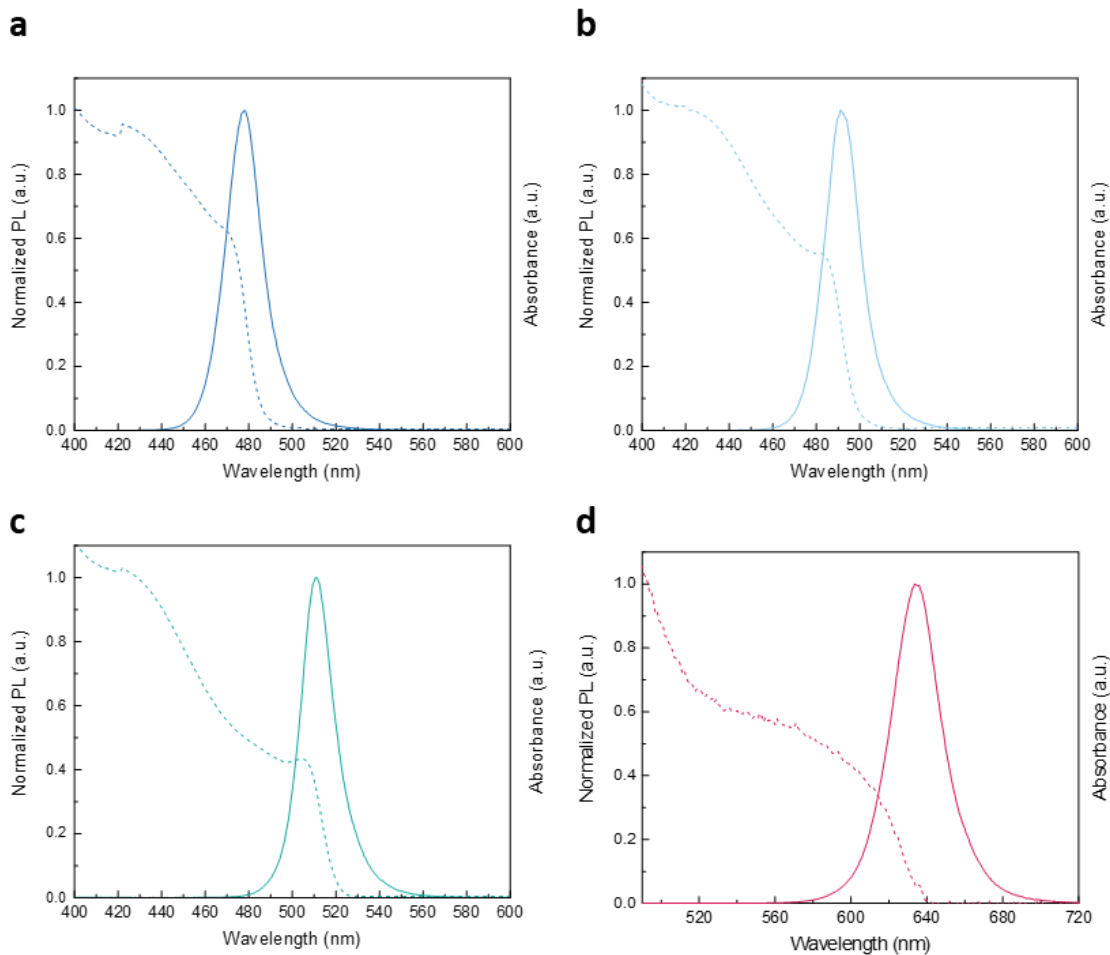


**Supplementary Figure 7. The Urbach energy of perovskite QWs.** Absorption coefficient spectra for **a**, 6-4 Cl-Br PeQWs. **b**, 4-6 Cl-Br PeQWs. **c**, CsPbBr<sub>3</sub> QWs. **d**, I-Br PeQWs.

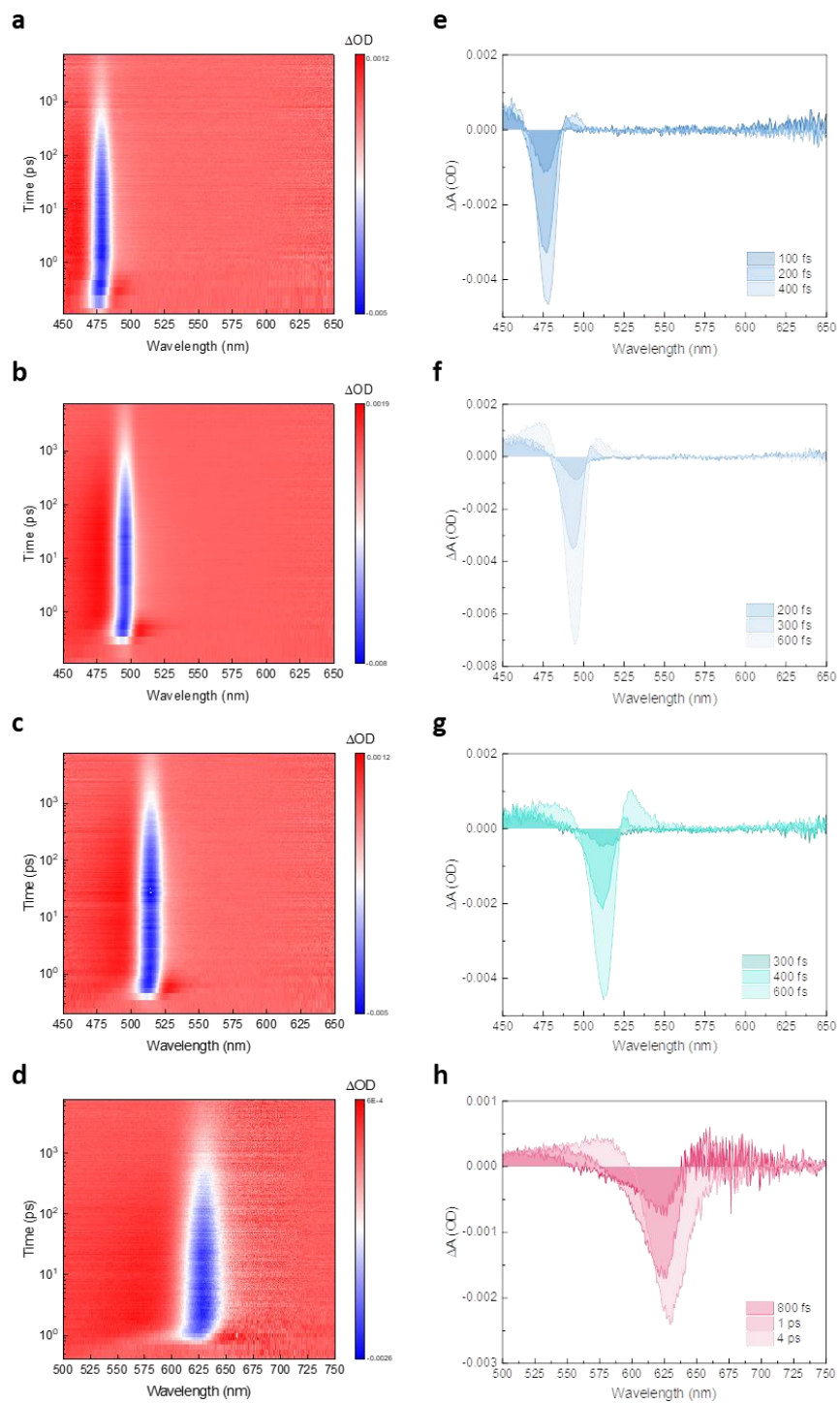
The absorption coefficient spectrum is extracted from the absorbance spectrum. In this way, the Urbach energy can be calculated as follow:

$$\alpha = \alpha_0 e^{\frac{h\nu}{E_u}}$$

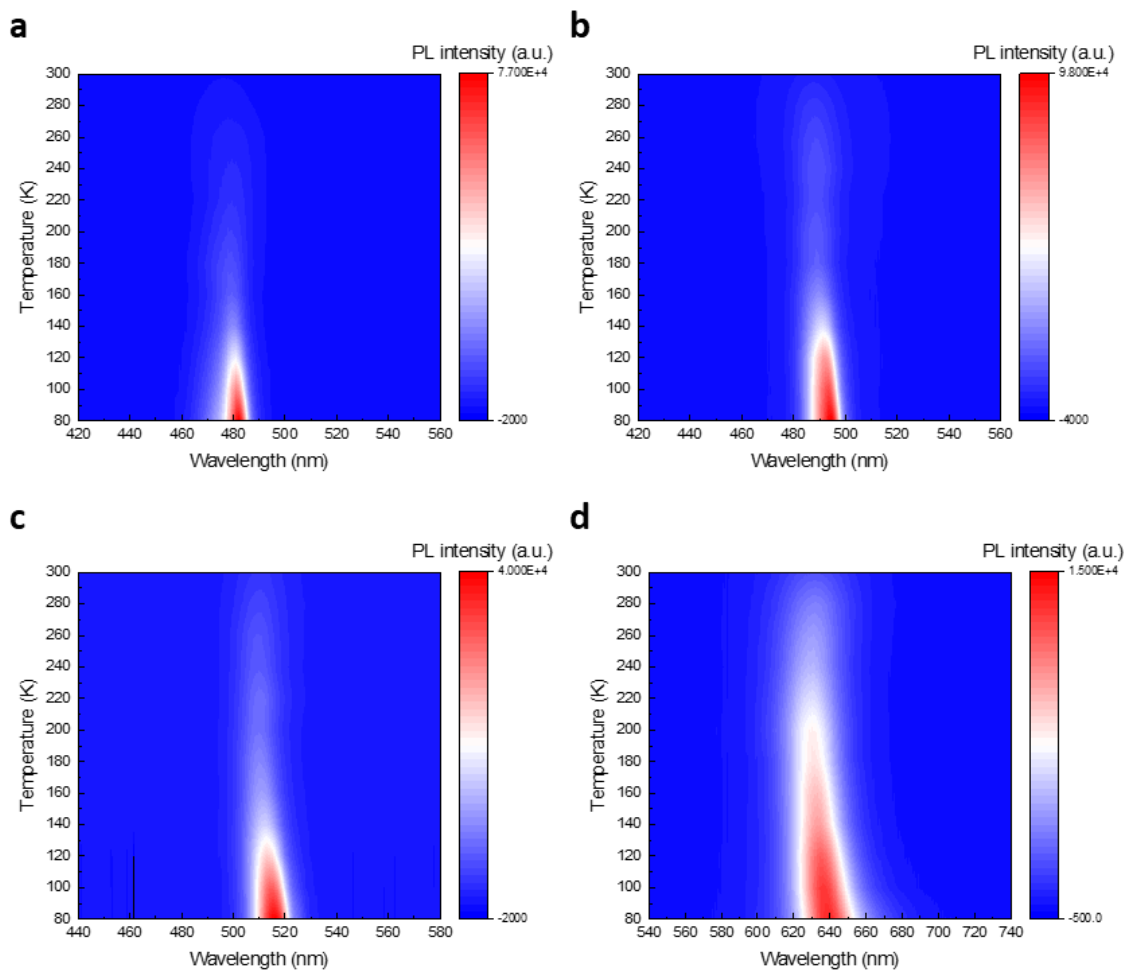
where  $\alpha$  is the absorption coefficient and  $h\nu$  is the photon energy. The Urbach energy was found to be 23.1 meV, 24.1 meV, 31.5 meV and 89.1 meV for 6-4 Cl-Br, 4-6 Cl-Br, CsPbBr<sub>3</sub> and I-Br PeQWs, respectively. Urbach energy is a parameter used to quantify energetic disorder in the band edges of a semiconductor. The low Urbach energy indicates a low electronic disorder in the band edges for our PeQWs.



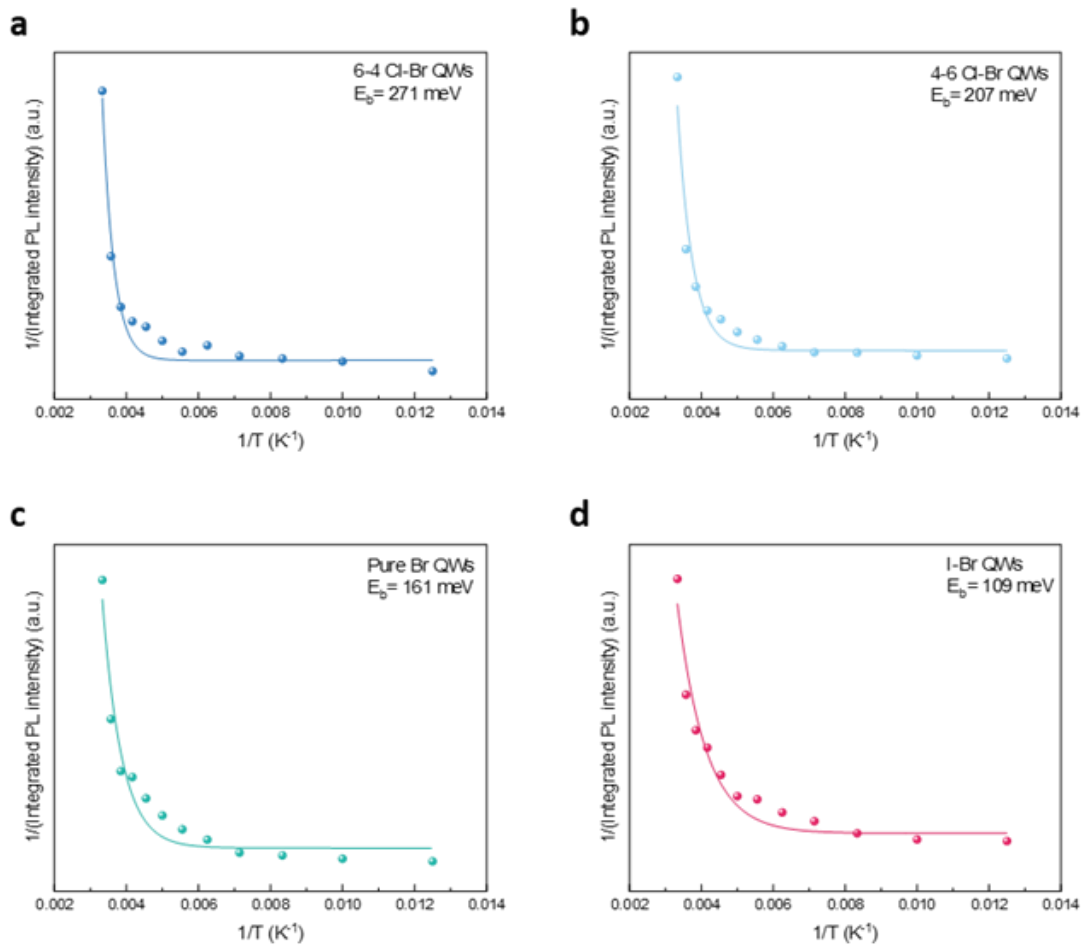
**Supplementary Figure 8. Photoluminescence and absorbance of perovskite QWs.** Photoluminescence and absorbance spectrum of **a**, 6-4 Cl-Br PeQWs. **b**, 4-6 Cl-Br PeQWs. **c**, CsPbBr<sub>3</sub> QWs. **d**, I-Br PeQWs.



**Supplementary Figure 9. Transient absorption (TA) spectra of perovskite QWs.** Transient absorption of **a,e**, 6-4 Cl-Br PeQWs. **b,f**, 4-6 Cl-Br PeQWs. **c,g**, CsPbBr<sub>3</sub> QWs. **d,h**, I-Br PeQWs.



**Supplementary Figure 10. Temperature-dependent PL spectra of perovskite QWs.**  
 Temperature-dependent PL spectra of **a**, 6-4 Cl-Br PeQWs. **b**, 4-6 Cl-Br PeQWs. **c**, CsPbBr<sub>3</sub> QWs. **d**, I-Br PeQWs.



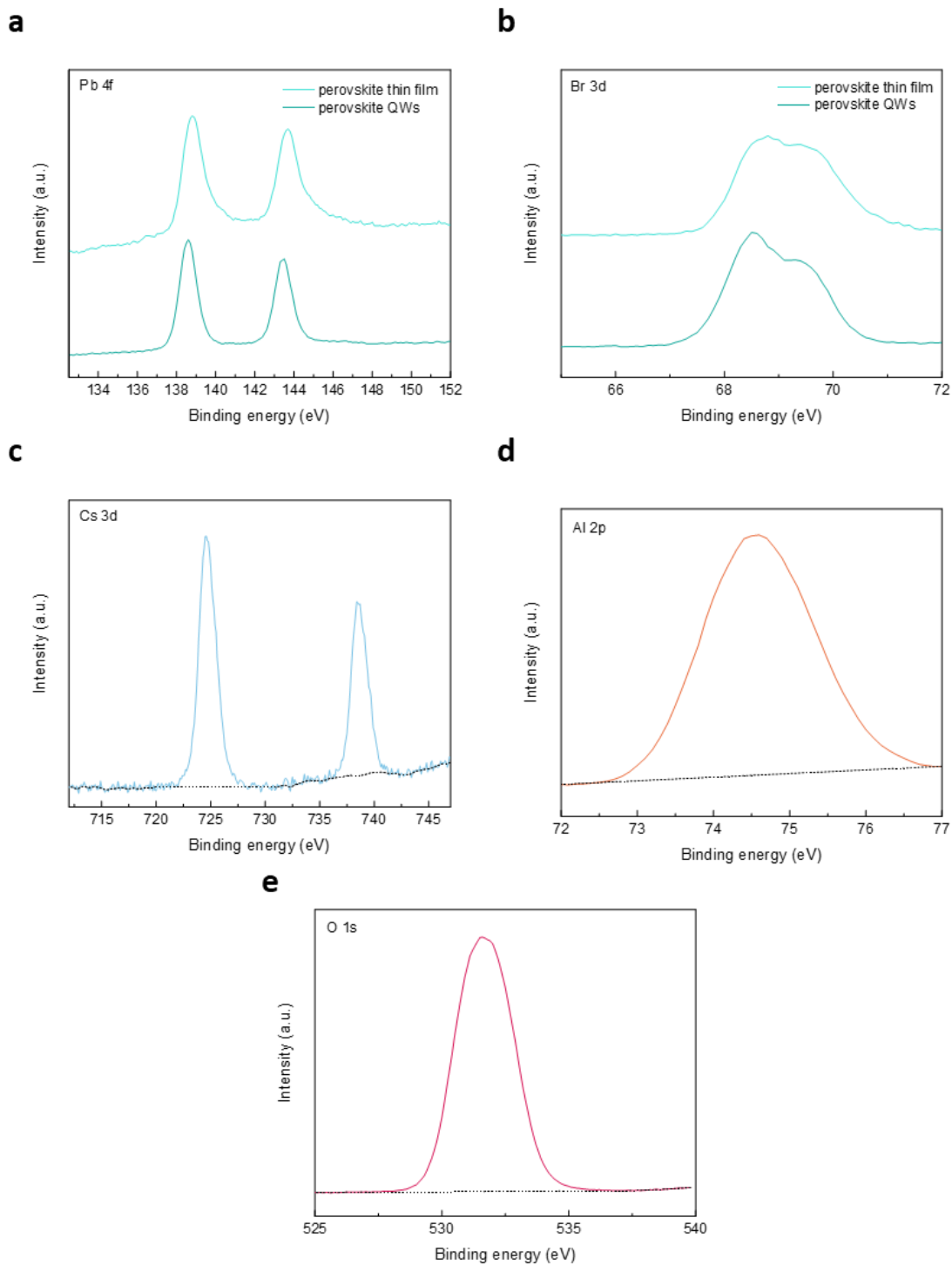
**Supplementary Figure 11. The exciton binding energy of perovskite QWs.** The calculated exciton binding energy of **a**, 6-4 Cl-Br PeQWs. **b**, 4-6 Cl-Br PeQWs. **c**, CsPbBr<sub>3</sub> QWs. **d**, I-Br PeQWs.

The estimate of exciton binding energy by the temperature dependence of integrated PL signal is obtained by the equation as below:

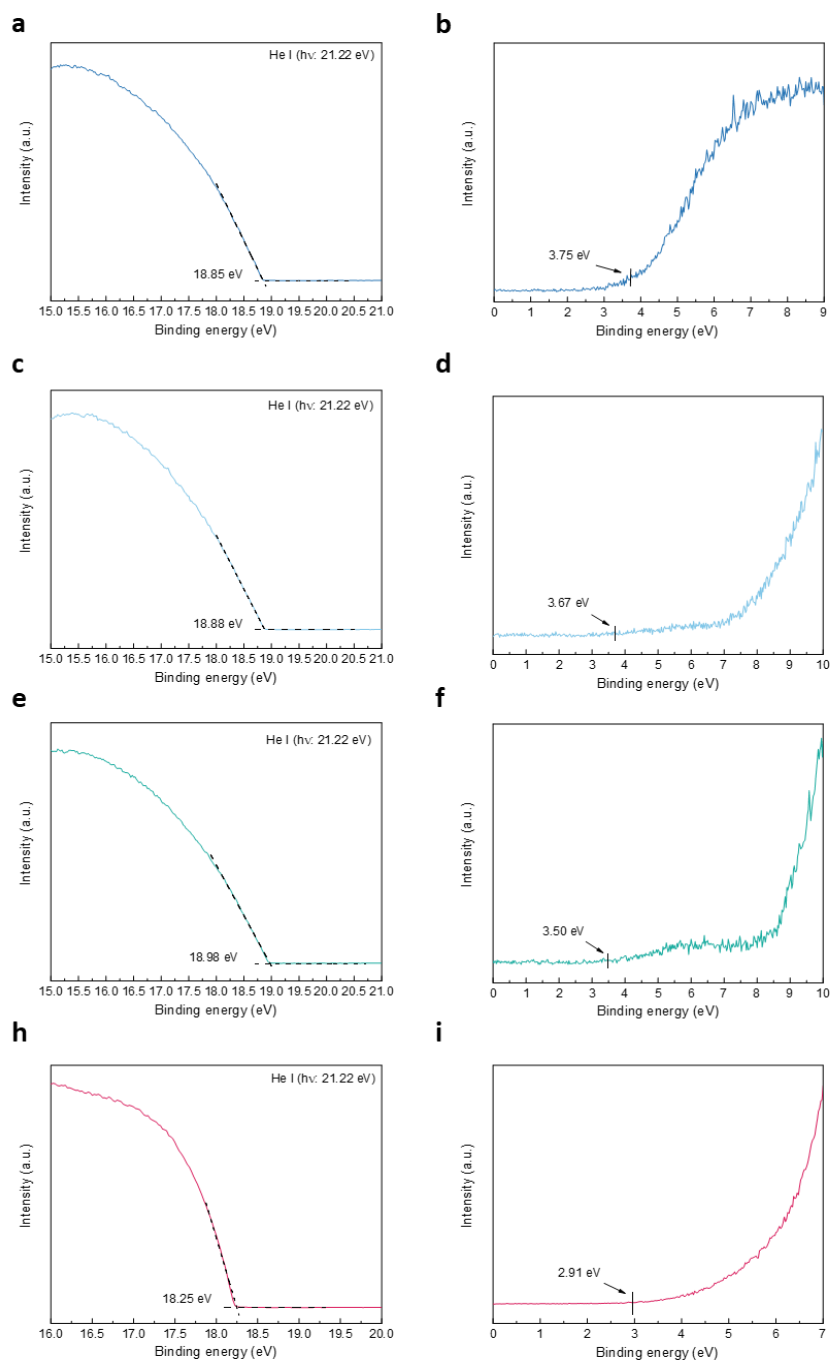
$$I(T) = \frac{I_0}{1 + Ae^{-\frac{E_b}{k_B T}}}$$

where  $I_0$  is the integrated PL intensity extrapolated at 0 K,  $A$  is a constant,  $E_b$  is the exciton binding energy, and  $k_b$  is the Boltzmann constant. Note that the exciton binding energy of perovskite might be over-estimated due to the possibility of structural transition at low temperature.

As a result, the exciton binding energy is approximately 271 meV, 207 meV, 161 meV and 106 meV for 6-4 Cl-Br, 4-6 Cl-Br, CsPbBr<sub>3</sub> and I-Br PeQWs, respectively.



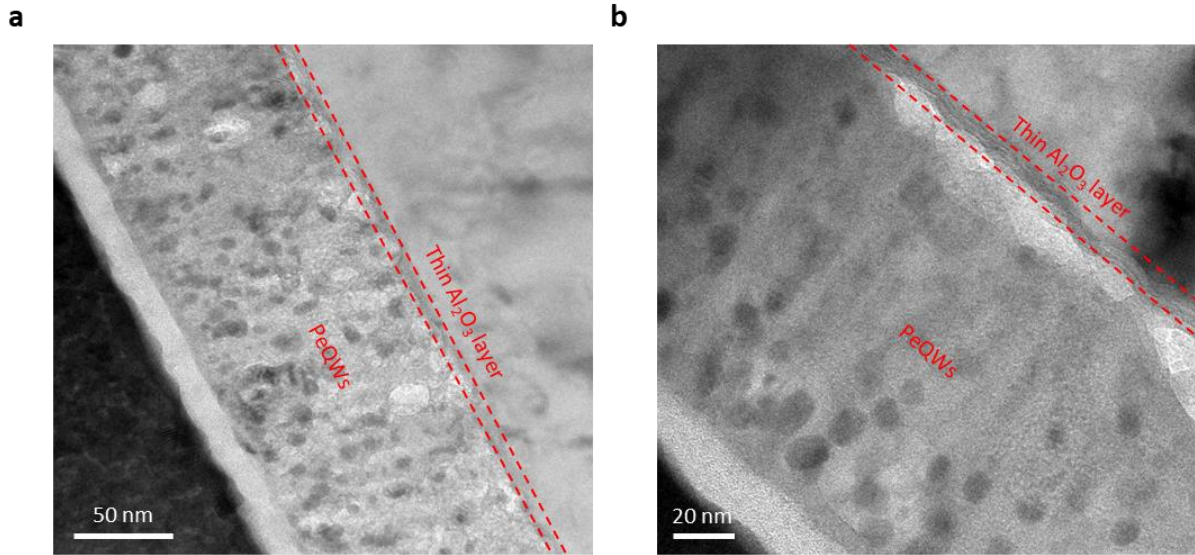
**Supplementary Figure 12. X-ray photoelectron spectroscopy (XPS) spectra of perovskite thin films and QWs. a,b,** Pb 4f and Br 3d spectra of perovskite thin films and QWs. XPS spectra for **c,** Cs 3d, **d,** Al 2p and **e,** O 1s.



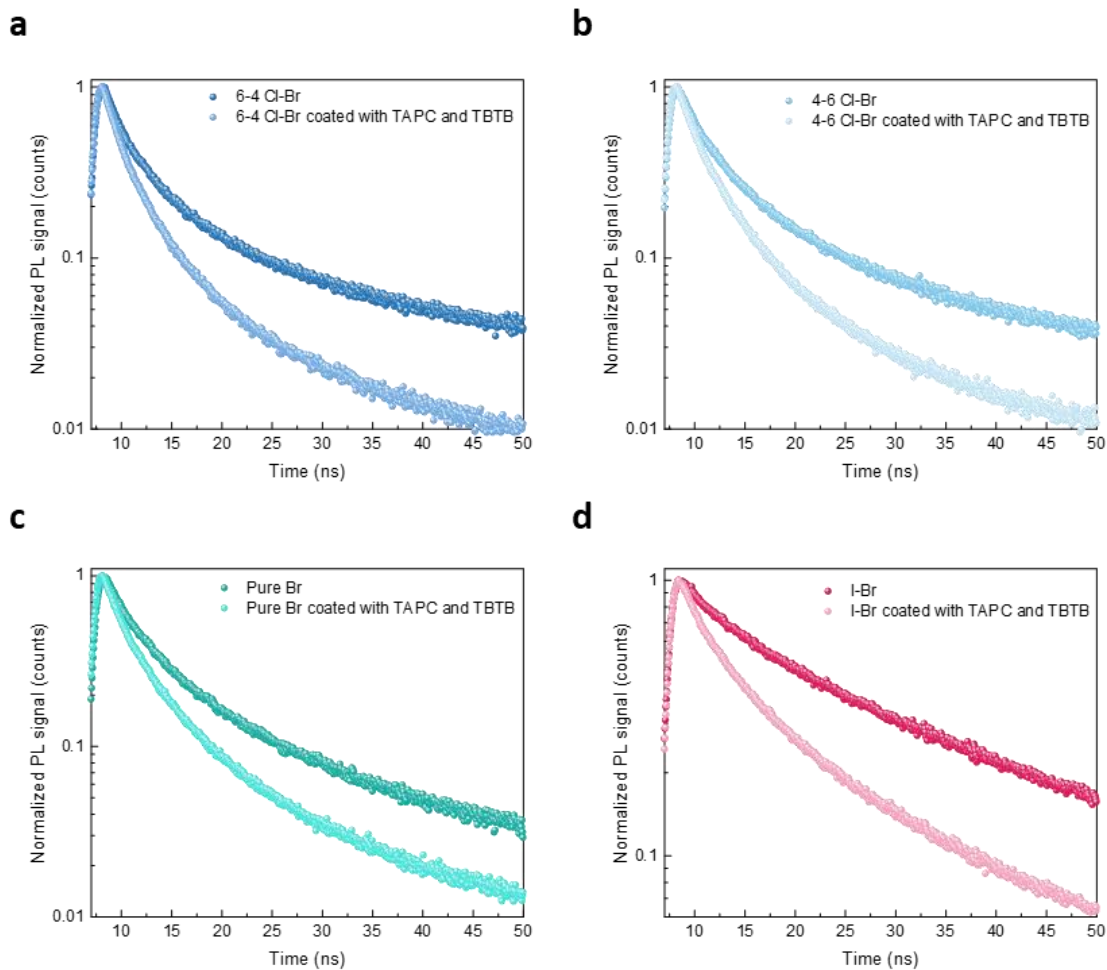
**Supplementary Figure 13. Ultraviolet photoelectron spectroscopy (UPS) characteristics.**

Photoemission cutoff energy of **a**, 6-4 Cl-Br PeQWs, **c**, 4-6 Cl-Br PeQWs, **e**, CsPbBr<sub>3</sub> QWs, **h**, I-Br PeQWs and the valence-band region of **b**, CsPbBr<sub>3</sub> QWs, **d**, 4-6 Cl-Br PeQWs, **f**, 6-4 Cl-Br PeQWs, **i**, I-Br PeQWs from ultraviolet photoemission spectra.

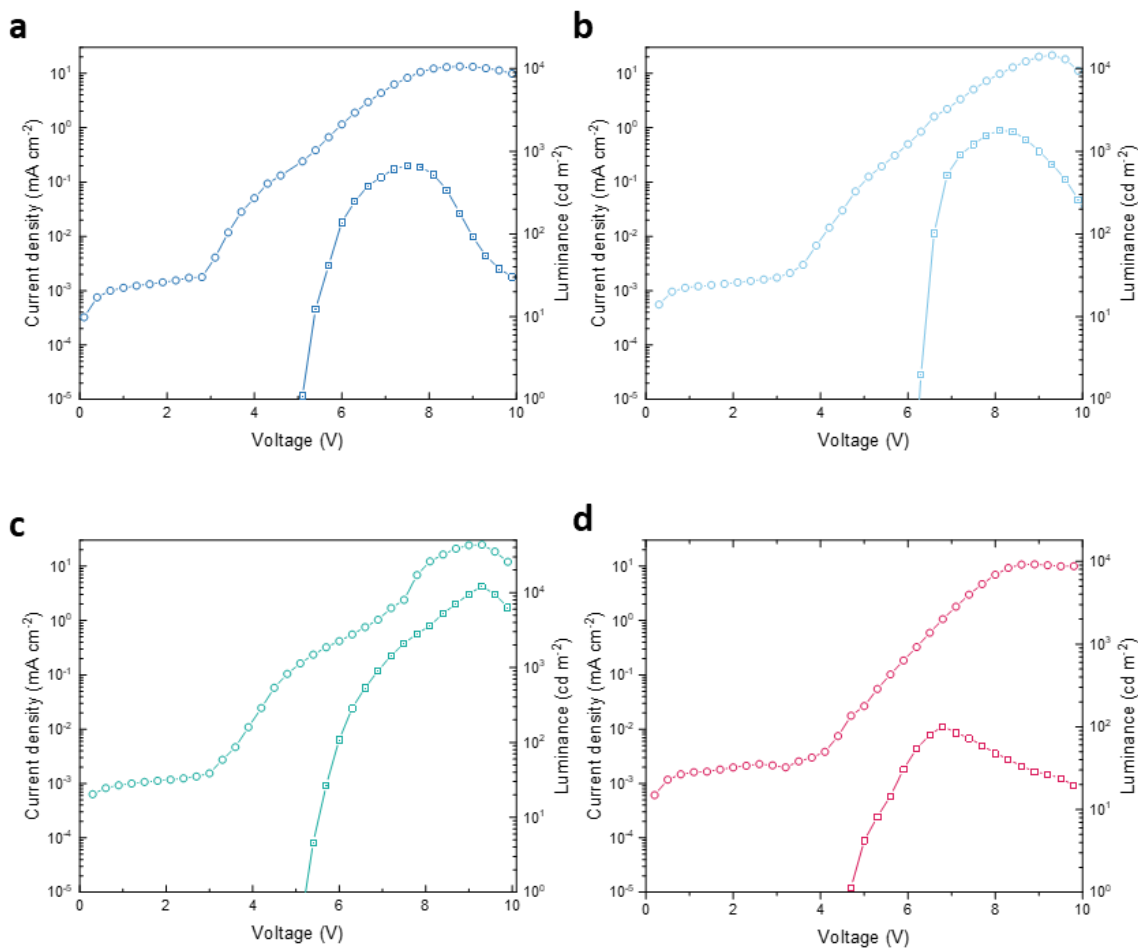




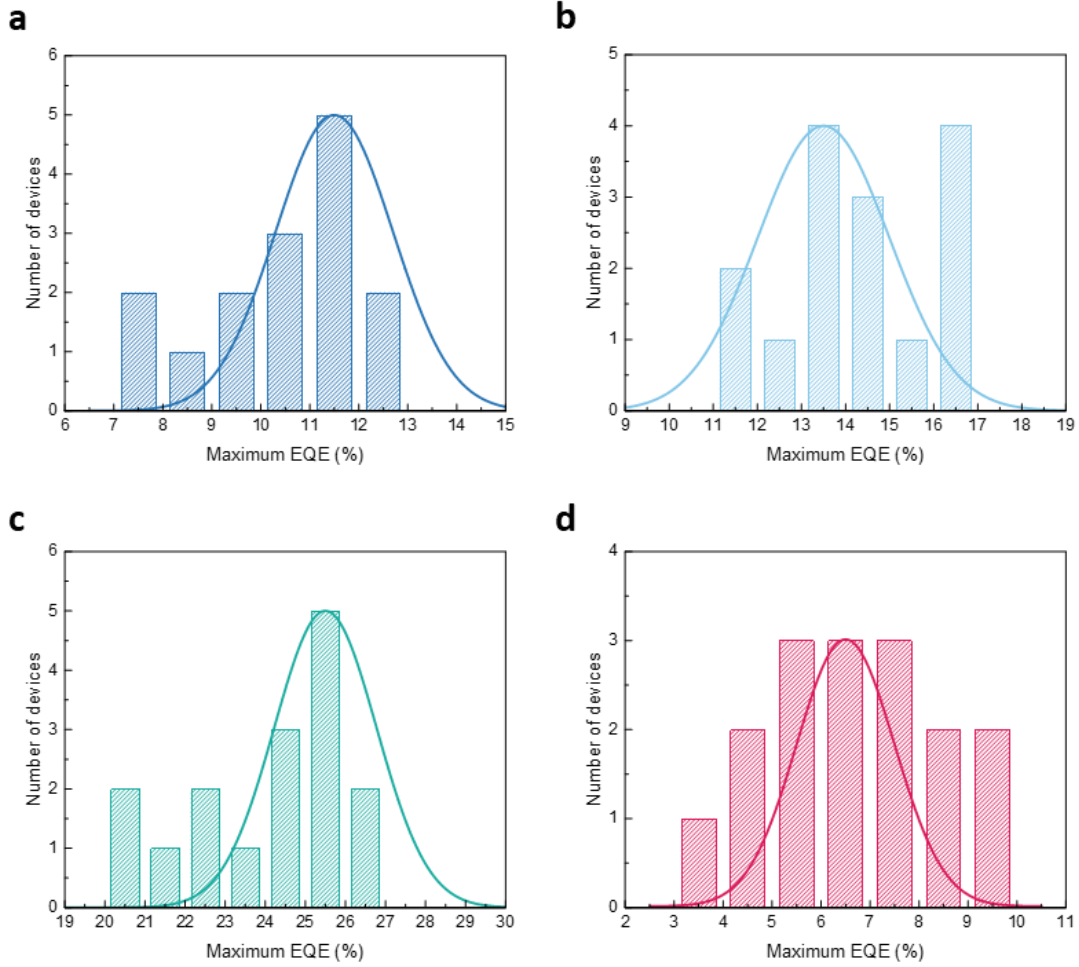
**Supplementary Figure 14. Cross-sectional HRTEM image of perovskite QWs with ultra-thin Al<sub>2</sub>O<sub>3</sub> layer.** **a** and **b**, Cross-sectional HRTEM image showing perovskite QWs and ultra-thin Al<sub>2</sub>O<sub>3</sub> layer. From the image, we can tell that the thickness of Al<sub>2</sub>O<sub>3</sub> layer is around 5 nm.



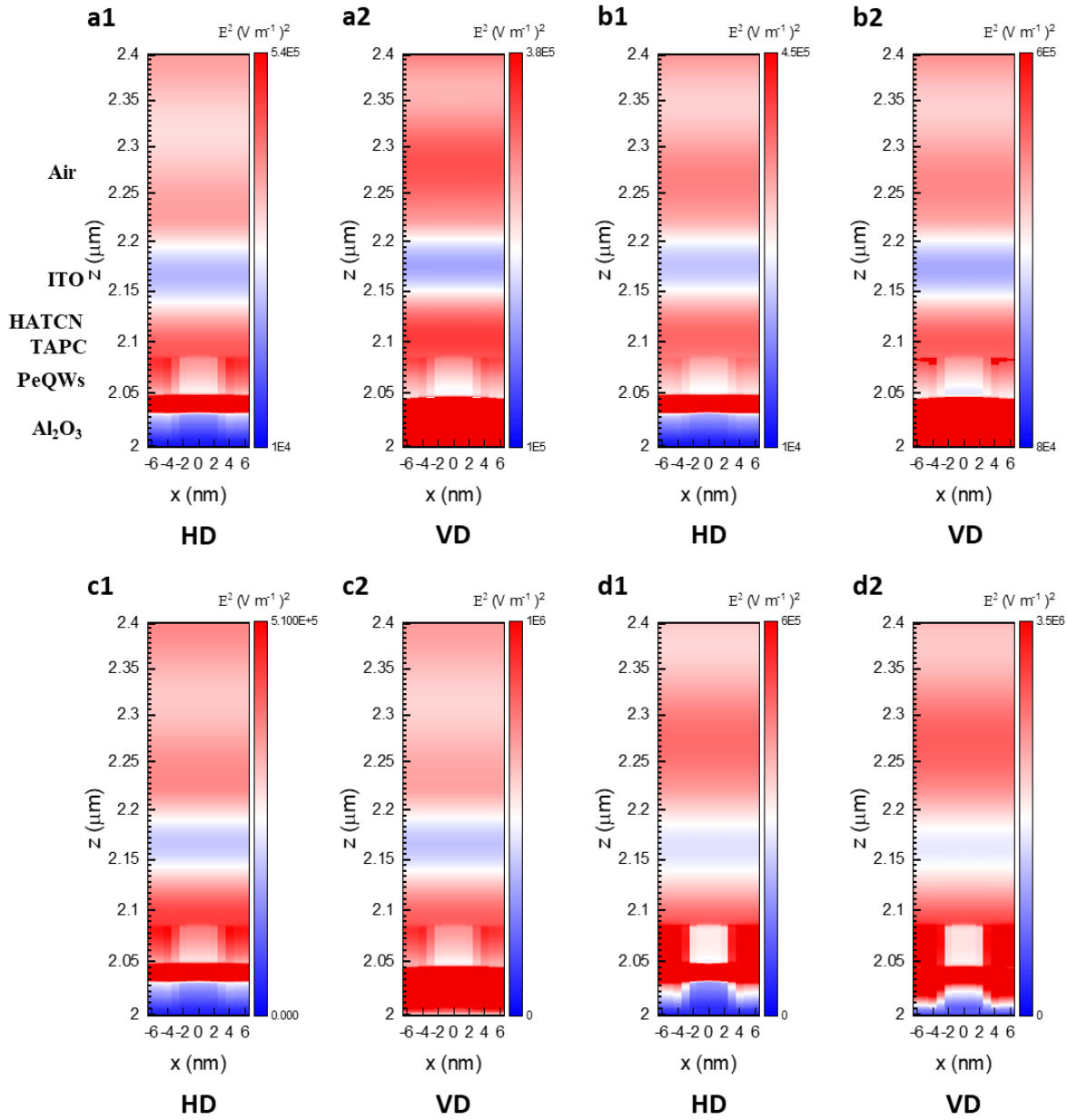
**Supplementary Figure 15. Photoluminescence lifetime characteristics.** Normalized time-resolved photoluminescence decay curves for **a**, 6-4 Cl-Br PeQWs, **b**, 4-6 Cl-Br PeQWs, **c**, CsPbBr<sub>3</sub> QWs and **d**, I-Br PeQWs with and without TAPC and TBTB.



**Supplementary Figure 16. The champion luminance of PeQWs-based LED device.** Current density ( $J$ )-voltage ( $V$ ) and luminance ( $L$ )-voltage ( $V$ ) curves of **a**, 6-4 Cl-Br PeQWs. **b**, 4-6 Cl-Br PeQWs. **c**, CsPbBr<sub>3</sub> QWs. **d**, I-Br PeQWs.



**Supplementary Figure 17. Statistical data for device performance.** Histograms of maximum EQEs for **a**, 6-4 Cl-Br PeQWs, **b**, 4-6 Cl-Br PeQWs, **c**, CsPbBr<sub>3</sub> QWs and **d**, I-Br PeQWs based LED devices.

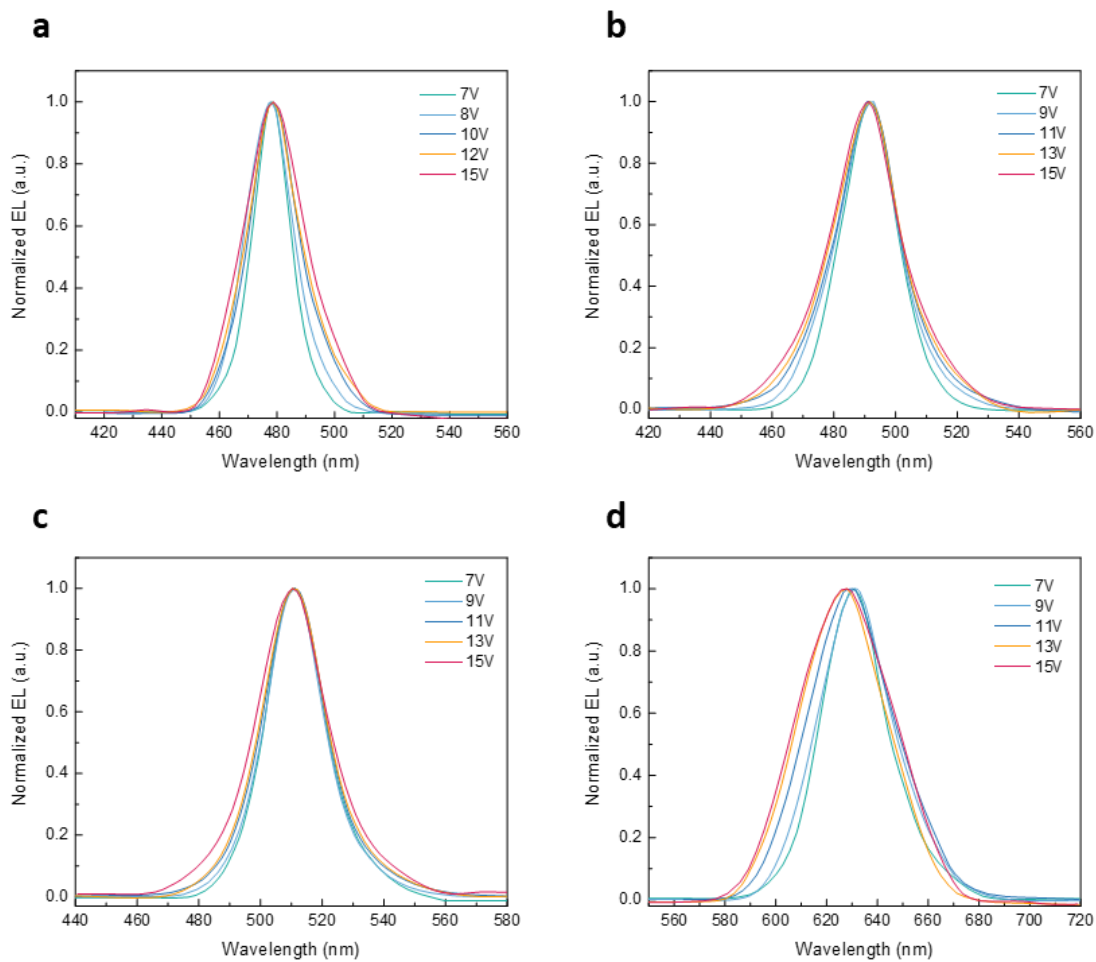


**Supplementary Figure 18. Optical simulation.** Cross-sectional  $E^2$  intensity profiles of horizontal dipole (HD) and vertical dipole (VD) for **a1-a2**, 6-4 Cl-Br PeQWs, **b1-b2**, 4-6 Cl-Br PeQWs, **c1-c2**, CsPbBr<sub>3</sub> QWs and **d1-d2**, I-Br PeQWs based LED devices.

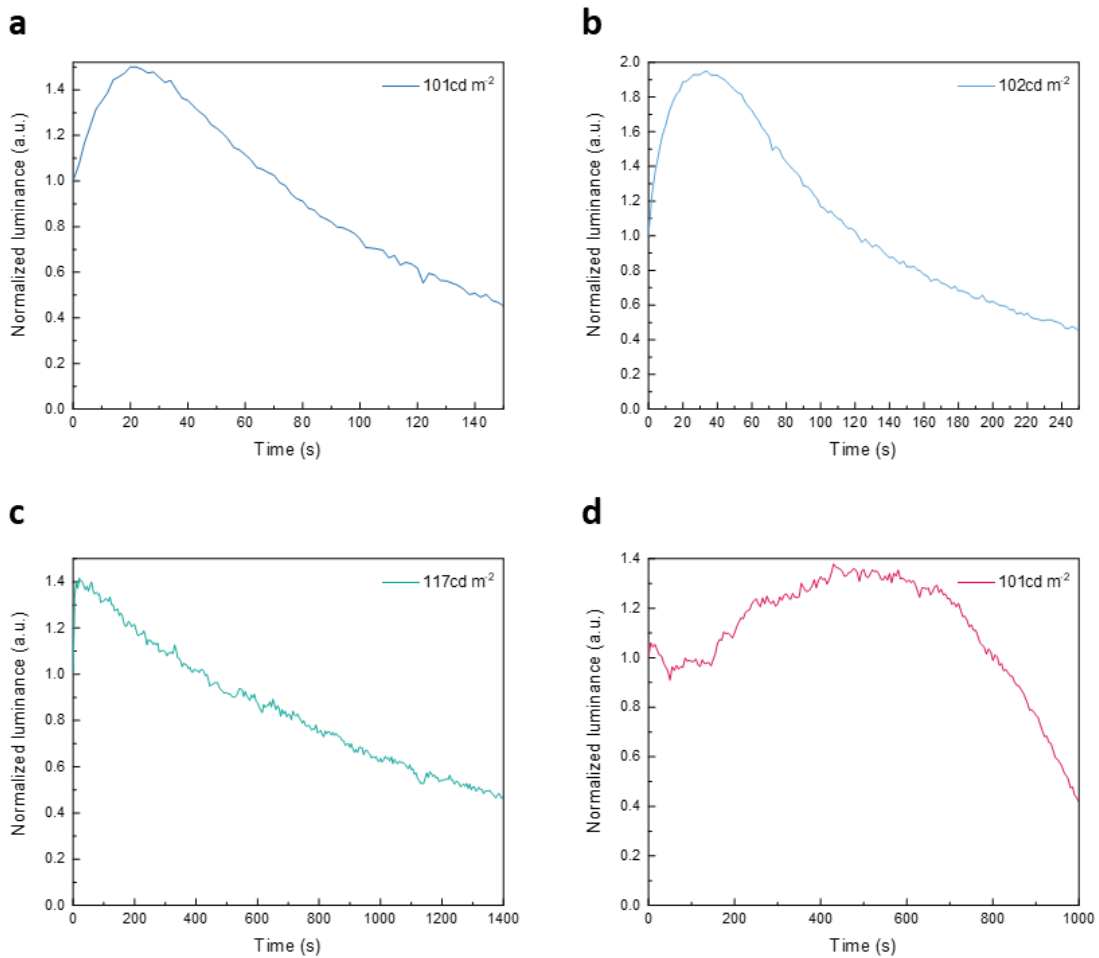
Perovskite QWs	Dipole polarization	Out-coupling efficiency (%)	Average out-coupling efficiency (%)
6-4 Cl-Br QWs	HD	87.15	77.67
	VD	58.10	
4-6 Cl-Br QWs	HD	87.22	78.75
	VD	61.82	
Pure Br QWs	HD	86.89	79.52
	VD	64.80	
I-Br QWs	HD	83.91	82.06
	VD	78.46	

**Supplementary Table 1. Summary of simulated outcoupling efficiency for PeQWs based LED devices.**

The optical model we used is similar to our previous report and it can be found in Figure S4<sup>1</sup>. According to the supplementary note 2 of our previous report<sup>2</sup>, the effective refractive index of perovskite QWs and PAM template is 1.79 (478 nm), 1.80 (491 nm), 1.81 (511 nm) and 1.92 (630 nm) for 6-4 Cl-Br PeQWs, 4-6 Cl-Br PeQWs, CsPbBr<sub>3</sub> QWs and I-Br PeQWs, respectively. Compared with perovskite itself, embedding perovskite in our PAM template reduces the effective refractive index of the emitting layer, greatly relieving the non-desired light-trapping effect usually defined by  $1/n^2$  where  $n$  is the index of the emitting layer.

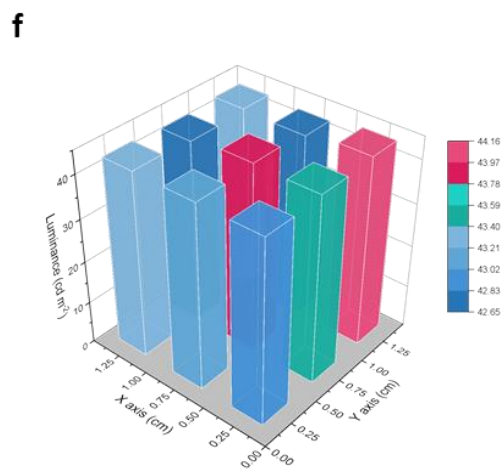
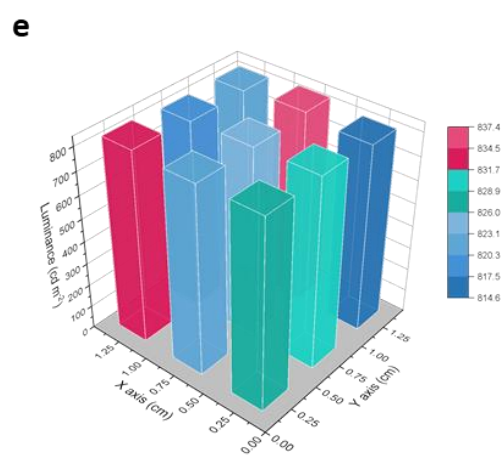
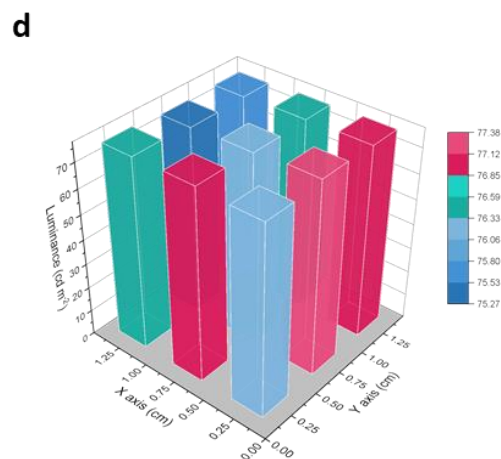
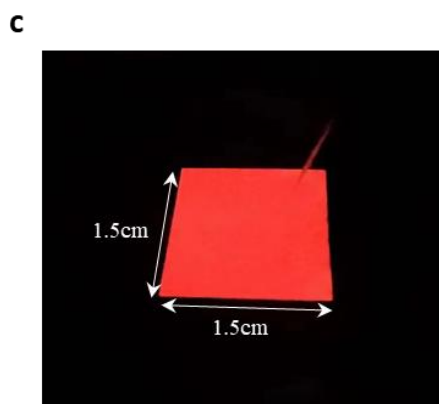
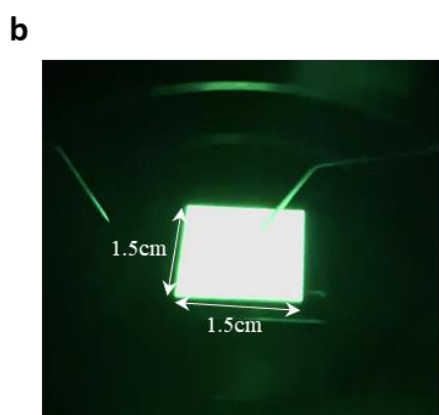
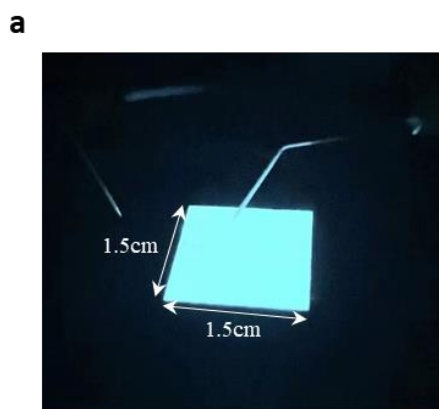


**Supplementary Figure 19. Stability of the electroluminescence spectra of PeQWs-based LEDs.** Electroluminescence spectra as a function of electric bias ( $>10$  V) for **a**, 6-4 Cl-Br PeQWs, **b**, 4-6 Cl-Br PeQWs, **c**, CsPbBr<sub>3</sub> QWs and **d**, I-Br PeQWs based LED devices.

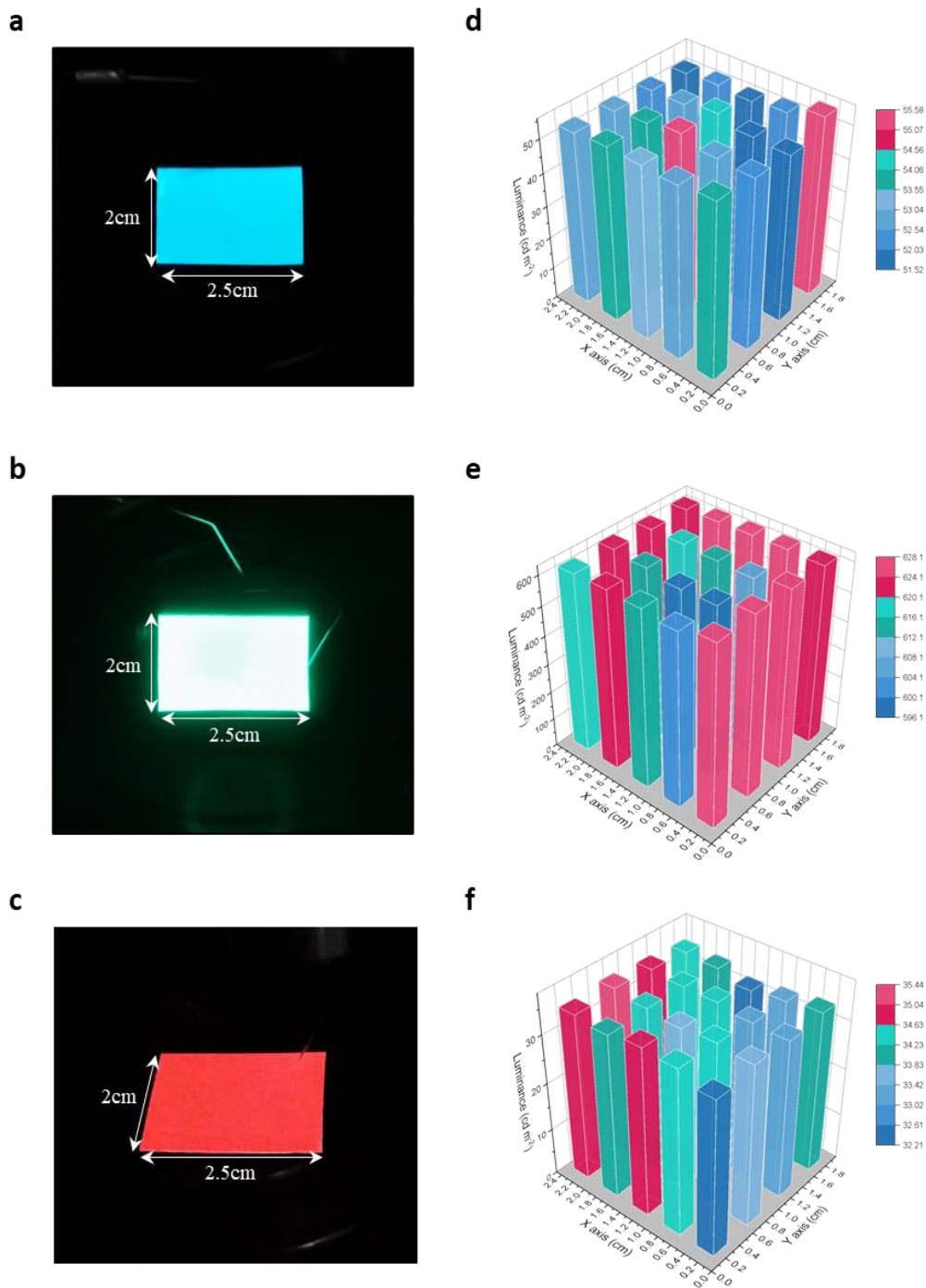


**Supplementary Figure 20. Operation stability of PeQWs-based LEDs.**  $T_{50}$  lifetime of **a**, 6-4 Cl-Br PeQWs, **b**, 4-6 Cl-Br PeQWs, **c**, CsPbBr<sub>3</sub> QWs and **d**, I-Br PeQWs based devices under different luminance.





**Supplementary Figure 21. 1.5×1.5 cm<sup>2</sup> PeQWs-based LED device.** **a-c**, The photo image of large-area PeQWs-based LEDs with an emission area of 1.5×1.5 cm<sup>2</sup>. **d-f**, The corresponding luminance distribution under working condition. Luminance is presented by a 3×3-pixel 3D column chart.



**Supplementary Figure 22.  $2 \times 2.5 \text{ cm}^2$  PeQWs-based LED device. a-c,** The photo image of large-area PeQWs-based LEDs with an emission area of  $2 \times 2.5 \text{ cm}^2$ . **d-f,** The corresponding luminance distribution under working condition. Luminance is presented by a  $4 \times 5$ -pixel 3D column chart.

**Supplementary Table 2.** Time-resolved photoluminescence lifetimes of PeQWs with and without TAPC.

	$\tau_{avr}$ (ns)	$\tau_1$ (ns)	$\tau_2$ (ns)	$\chi^2$
6-4 Cl-Br QWs	5.45	1.36 (32.23%)	7.39 (67.77%)	3.281
4-6 Cl-Br QWs	6.87	1.78 (30.99%)	9.15 (69.01%)	2.487
Pure Br QWs	7.40	1.90 (26.94%)	9.43 (73.06%)	2.282
I-Br QWs	12.94	1.20 (2.94%)	13.30 (97.06%)	4.313
6-4 Cl-Br QWs coated with TAPC and TBTB	4.36	1.63 (50.48%)	7.38 (47.52%)	3.285
4-6 Cl-Br QWs coated with TAPC and TBTB	4.78	1.87 (48.30%)	7.50 (51.40%)	2.847
Pure Br QWs coated with TAPC and TBTB	5.45	1.93 (40.55%)	7.85 (59.45%)	3.755
I-Br QWs coated with TAPC and TBTB	9.22	1.62 (14.38%)	10.50 (85.62%)	3.867

**Supplementary Table 3.** Performance of reported top-emitting perovskite LEDs

Perovskites	$EQE_{max}$ (%)	CIE (x, y)	$L_{max}$ ( $cd\ m^{-2}$ )	$\lambda_{EL}$ (nm)	Reference
6-4 Cl-Br QWs	12.41	(0.11, 0.13)	670	478	This work
4-6 Cl-Br QWs	16.49	(0.08, 0.33)	1788	491	
Pure Br QWs	26.09	(0.06, 0.71)	12147	512	
I-Br QWs	9.97	(0.70, 0.30)	101	630	
QDs: CsPbI <sub>3</sub>	12.6	(0.72, 0.28)	10171	682	<b>Ref. 3</b>
RDP: PEABr:CsPbBr <sub>3</sub>	13.6	(NA	~11000	512	<b>Ref. 4</b>
QDs: Zr <sup>2+</sup> :CsPbI <sub>3</sub>	13.7	(0.71, 0.28)	14725	686	<b>Ref. 5</b>
3D perovskites: FAPbI <sub>3</sub>	20.2	NA	114.9 $mW\ sr^{-1}\ m^{-2}$	~800	<b>Ref. 6</b>

$EQE_{max}$ , maximum external quantum efficiency; CIE, Commission Internationale de l'Eclairage;  $L_{max}$ , maximum luminance;  $R_{max}$ , maximum radiance;  $\lambda_{EL}$ , EL wavelength; RDP, reduced dimensional perovskite; QDs, quantum dots.

**Supplementary Table 4.** Performance of reported all-inorganic perovskite LEDs

Perovskites	$EQE_{max}$ (%)	CIE (x, y)	$L_{max}$ ( $cd\ m^{-2}$ )	$\lambda_{EL}$ (nm)	Reference
6-4 Cl-Br QWs	12.41	(0.11, 0.13)	670	478	This work
4-6 Cl-Br QWs	16.49	(0.08, 0.33)	1788	491	
Pure Br QWs	26.09	(0.06, 0.71)	12147	512	
I-Br QWs	9.97	(0.70, 0.30)	101	630	
QDs: K <sup>+</sup> :CsPb(Br/Cl) <sub>3</sub>	1.96	(0.11, 0.12)	212.9	476	<b>Ref. 7</b>
3D perovskites: Rb <sup>+</sup> :CsPb(Br/Cl) <sub>3</sub>	11.0	(0.107, 0.115)	2180	477	<b>Ref. 8</b>
QDs: CsPbBr <sub>3</sub>	12.3	(NA, 0.13)	~400	479	<b>Ref. 9</b>
RDP: YCl <sub>3</sub> :CsPbBr <sub>3</sub>	13.5	(0.07, 0.25)	2224	488	<b>Ref. 10</b>
QDs: CsPb(Br/Cl) <sub>3</sub>	1.9	NA	35	490	<b>Ref. 11</b>
RDP: CsPbBr <sub>3</sub>	13.8	(0.06, 0.36).	2825	496	<b>Ref. 12</b>
QDs: CsPbBr <sub>3</sub>	22.0	NA	~1000	505	<b>Ref. 9</b>
3D perovskites: CsPbBr <sub>3</sub>	16.45	NA	112824	522	<b>Ref. 13</b>
QDs: K <sup>+</sup> :CsPbI <sub>3</sub>	23	(0.69, 0.30)	~1000	640	<b>Ref. 14</b>

$EQE_{max}$ , maximum external quantum efficiency; CIE, Commission Internationale de l'Eclairage;  $L_{max}$ , maximum luminance;  $\lambda_{EL}$ , EL wavelength; RDP, reduced dimensional perovskite; QDs, quantum dots.

## Supplementary References

- 1 Zhang, Q. *et al.* Three-Dimensional Perovskite Nanophotonic Wire Array-Based Light-Emitting Diodes with Significantly Improved Efficiency and Stability. *ACS Nano* **14**, 1577-1585 (2020).
- 2 Zhang, Q. *et al.* Efficient metal halide perovskite light-emitting diodes with significantly improved light extraction on nanophotonic substrates. *Nature Communications* **10**, 727 (2019).
- 3 Lu, M. *et al.* Surface ligand engineering-assisted CsPbI<sub>3</sub> quantum dots enable bright and efficient red light-emitting diodes with a top-emitting structure. *Chemical Engineering Journal* **404**, 126563 (2021).
- 4 Cai, L. *et al.* High-Efficiency Top-Emitting Green Perovskite Light Emitting Diode with Quasi Lambertian Emission. *Advanced Optical Materials* **n/a**, 2101137.
- 5 Lu, M. *et al.* Bright CsPbI<sub>3</sub> Perovskite Quantum Dot Light-Emitting Diodes with Top-Emitting Structure and a Low Efficiency Roll-Off Realized by Applying Zirconium Acetylacetonate Surface Modification. *Nano Letters* **20**, 2829-2836 (2020).
- 6 Miao, Y. *et al.* Microcavity top-emission perovskite light-emitting diodes. *Light: Science & Applications* **9**, 89 (2020).
- 7 Yang, F. *et al.* Efficient and Spectrally Stable Blue Perovskite Light-Emitting Diodes Based on Potassium Passivated Nanocrystals. *Advanced Functional Materials* **30**, 1908760 (2020).
- 8 Karlsson, M. *et al.* Mixed halide perovskites for spectrally stable and high-efficiency blue light-emitting diodes. *Nature Communications* **12**, 361 (2021).
- 9 Dong, Y. *et al.* Bipolar-shell resurfacing for blue LEDs based on strongly confined perovskite quantum dots. *Nature Nanotechnology* **15**, 668-674 (2020).
- 10 Liu, Y. *et al.* Water-Soluble Conjugated Polyelectrolyte Hole Transporting Layer for Efficient Sky-Blue Perovskite Light-Emitting Diodes. *Small* **17**, 2101477 (2021).
- 11 Pan, J. *et al.* Highly Efficient Perovskite-Quantum-Dot Light-Emitting Diodes by Surface Engineering. *Advanced Materials* **28**, 8718-8725 (2016).
- 12 Zhu, Z. *et al.* Highly Efficient Sky-Blue Perovskite Light-Emitting Diode Via Suppressing Nonradiative Energy Loss. *Chemistry of Materials* **33**, 4154-4162 (2021).
- 13 Feng, W. *et al.* Efficient all-inorganic perovskite light-emitting diodes enabled by manipulating the crystal orientation. *Journal of Materials Chemistry A* **9**, 11064-11072 (2021).
- 14 Wang, Y.-K. *et al.* All-Inorganic Quantum-Dot LEDs Based on a Phase-Stabilized  $\alpha$ -CsPbI<sub>3</sub> Perovskite. *Angewandte Chemie International Edition* **60**, 16164-16170 (2021).