Supplementary Information

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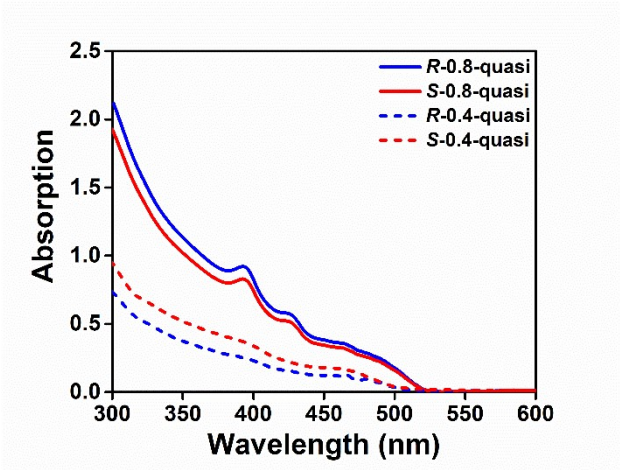

Fig. S1. UV-Vis spectra of *R*-/*S*-0.8-quasi films and *R*-/*S*-0.4-quasi films.

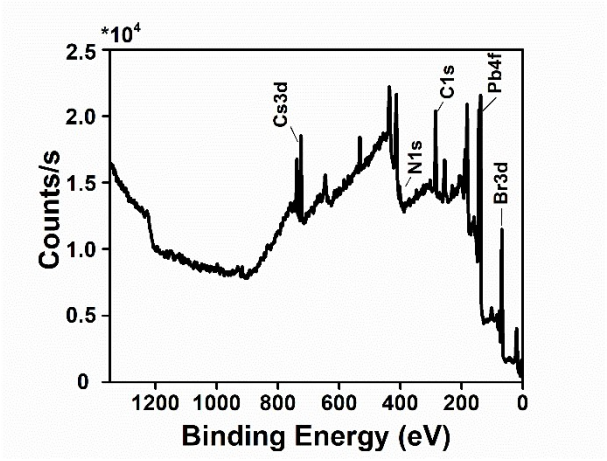

Fig. S2 X-ray photoelectron spectroscopy spectra (XPS) of a *R*-0.4-quasi film**.**

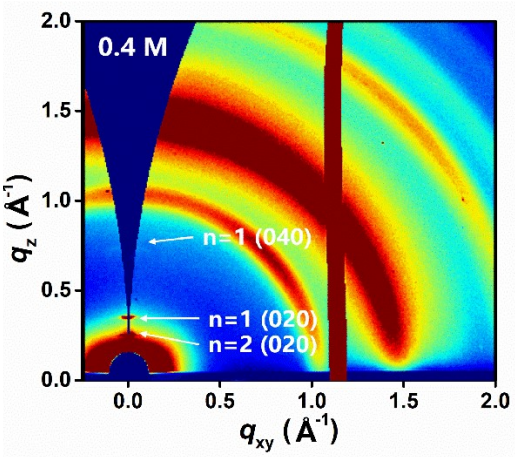

Fig. S3 Enhanced GIWAXS pattern of *R*-0.4-quasi film.

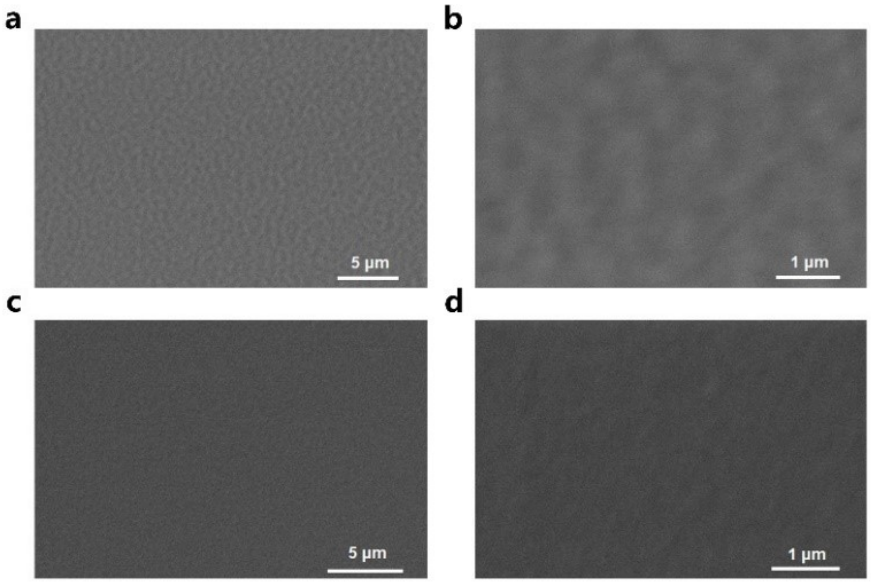

Fig. S4 SEM images of quasi-2D films: **a**, **b** *R*-0.8-quasi film. **c**, **d** *R*-0.4-quasi film.

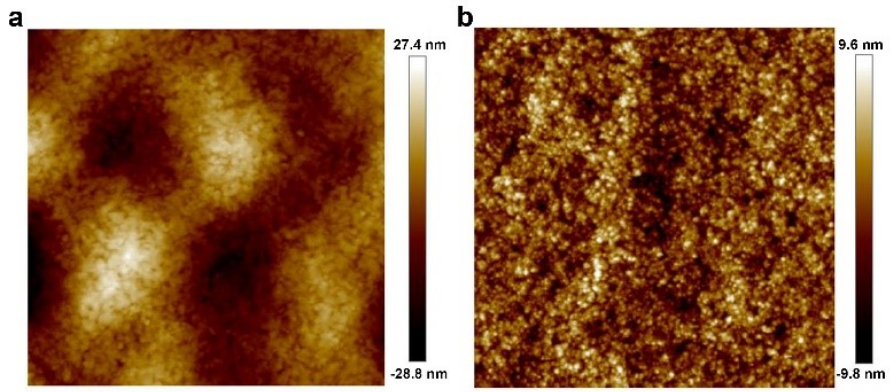

Fig. S5 AFM image area of quasi-2D films (2*2 μm^2): **a** *R*-0.8-quasi film. **b** *R*-0.4-quasi film.

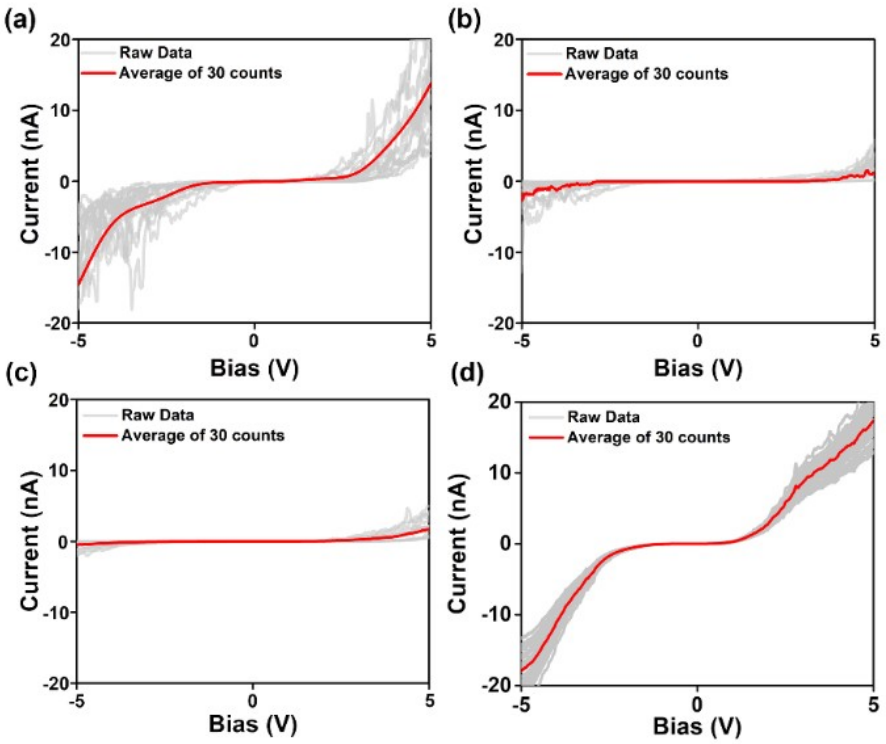

Fig. S6 Raw and averaged *I*−*V* curves obtained using the mCP-AFM for *R*-0.2-quasi film **a** Tip_{down}, **b** Tip_{up} and *S*-0.2-quasi film **c** Tip_{down}, **d** Tip_{up}.

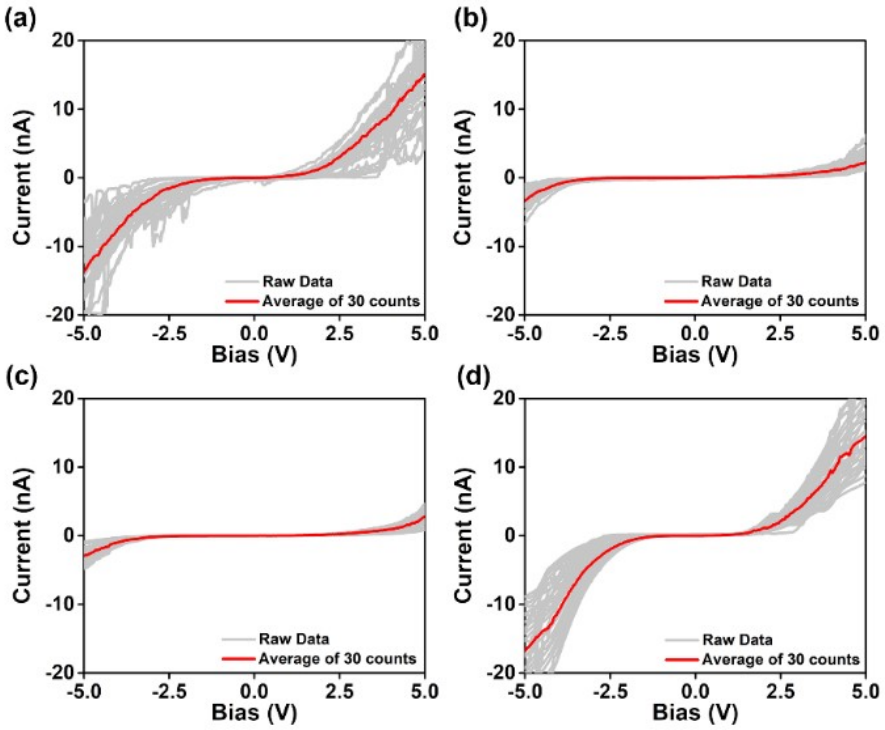

Fig. S7 Raw and averaged *I*−*V* curves obtained using the mCP-AFM for *R*-2D film **a** Tip_{down}, **b** Tip_{up} and *S*-2D film **c** Tip_{down}, **d** Tip_{up}.

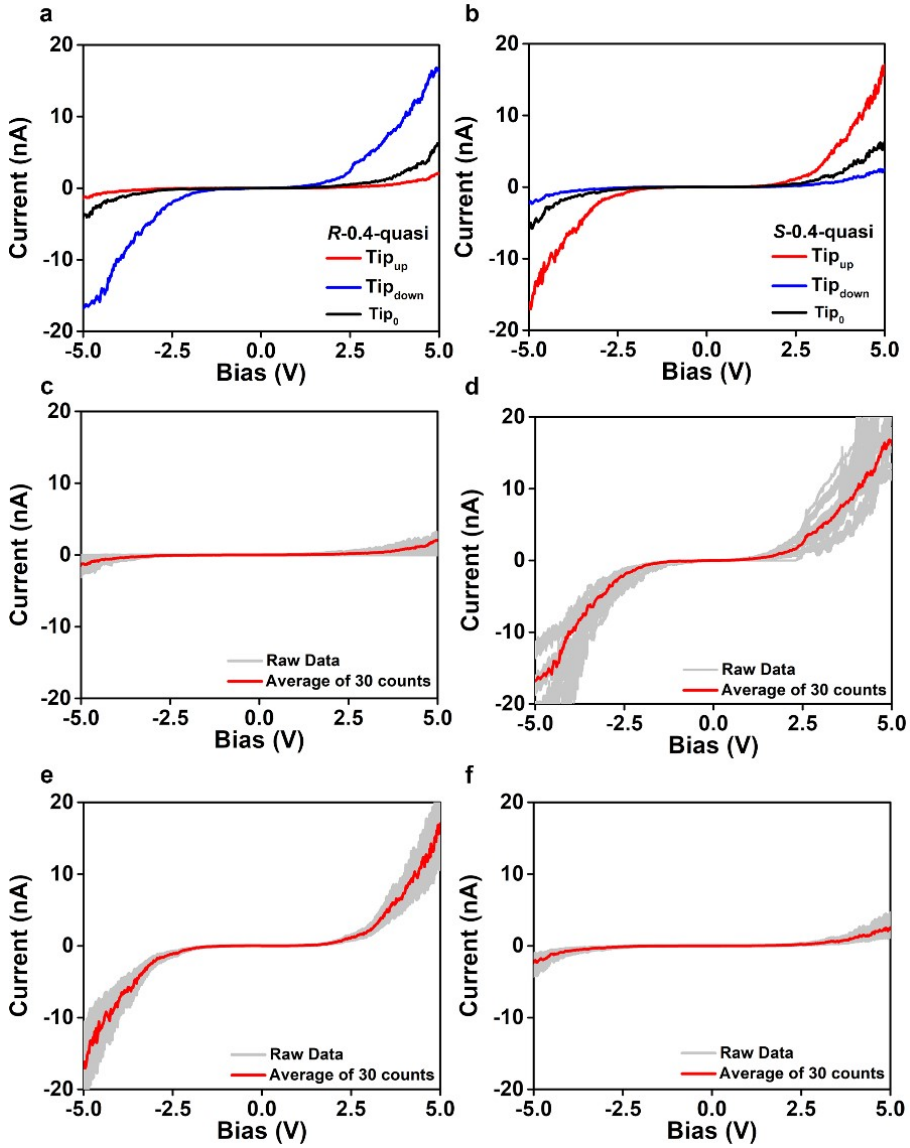

Fig. S8 Raw and averaged *I*−*V* curves obtained using the mCP-AFM for *R*-0.4-quasi film **c** Tip_{down}, **d** Tip_{up} and *S*-0.4-quasi film **e** Tip_{down}, **f** Tip_{up}.

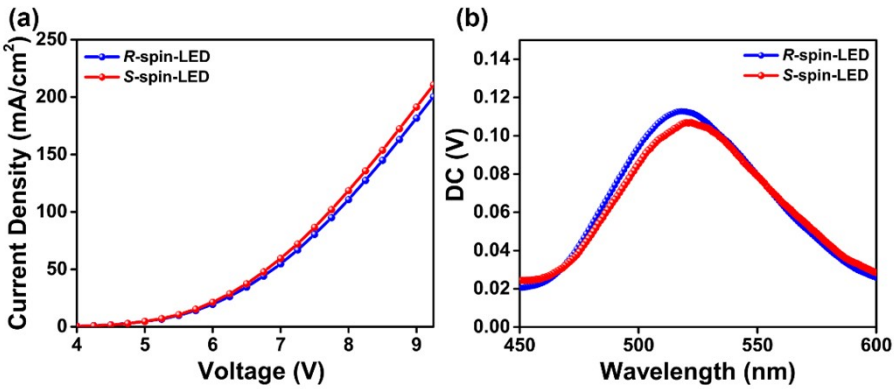

Fig. S9 Electroluminescence performance of spin-LEDs: **a** Current density-voltage curves of Spin-LEDs. **b** DC spectrum measured at the same time with CP-EL spectrum.

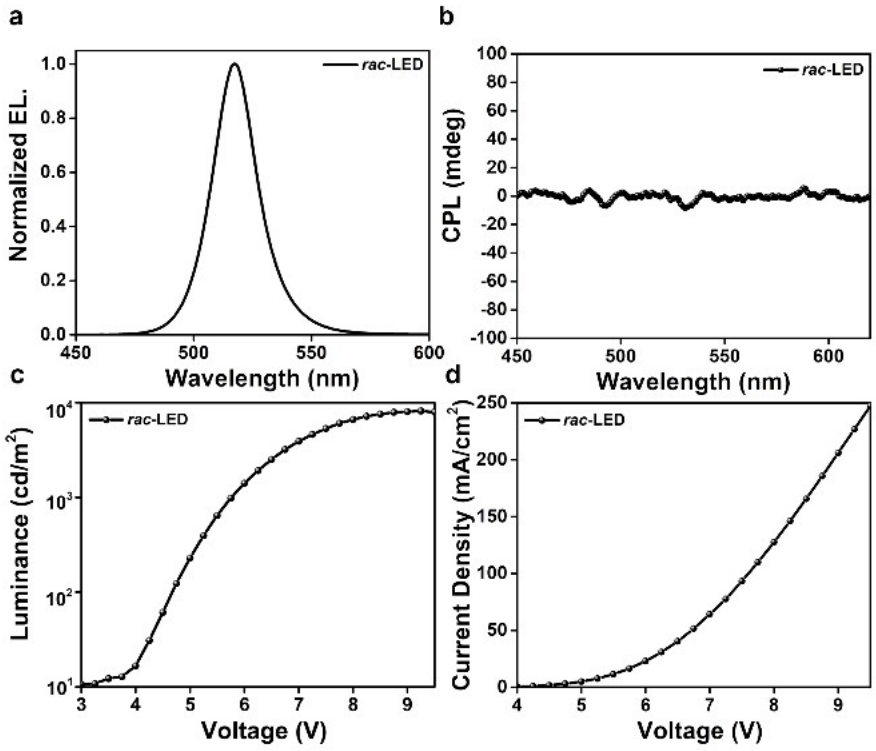

Fig. S10 Electroluminescence properties of the *rac*-LED: **a** EL spectrum of the *rac*-LED. **b** CP-EL spectrum of the *rac*-LED. **c** Luminance versus voltage curves of the *rac*-LED. **d** Current Density-Voltage curves of *rac*-LED.

Table S1 Elemental analysis of quasi-2D film

Atomic ratio (%)	C1s	N1s	Br3d	Cs3d5	Pb4f7	N/Pb	Cs/Pb
R-0.4-quasi film	59.47	7.54	23.88	1.28	7.82	0.96	0.16
Precursor	N/A	N/A	N/A	N/A	N/A	1.27	0.33

Table S2 Summary of CP-EL Emitting LEDs Based on CISS

Spin-LEDs	$\lvert g_{CP\text{-}EL} \rvert$	Luminance (cd/m^2)	EQE (%)	ref
Chiral 2D - achiral 3D	$5.0*10^{-3}$	4200	10.05	1
Chiral core - achiral shell	$6.0*10^{-3}$	1500	3.60	2
Chiral 2D perovskite- achiral CdSe	$1.6*10^{-2}$	18000	2.70	3
Core–Shell Perovskite QD	0.24	1962	5.47	4
This work	0.05	9300	3.80	-

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