Supporting Information

Polymeric Lewis-Base-Assisted Dual-Phase 3D $CsPbBr_3$–Cs_4PbBr_6 Perovskite Stability Based on Molecular Engineering and Self-Passivation

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Prof. Y. Zhou (E-mail: yezhou@szu.edu.cn)

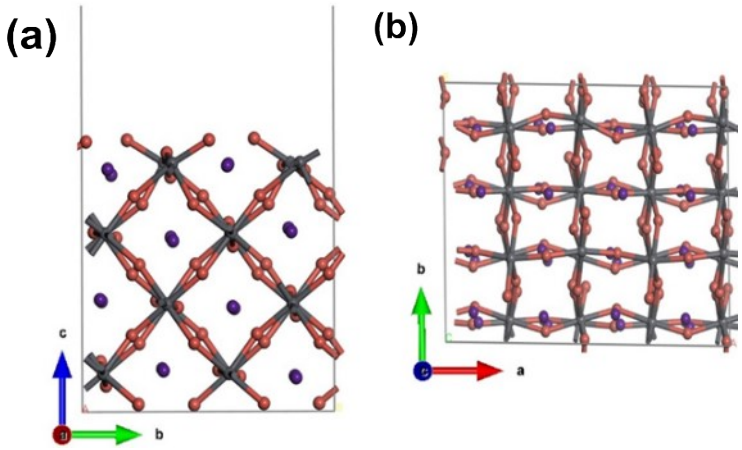

Fig. S1. (a) Side and (b) top views of optimized most stable slab model of $CsPbBr_3$ (121) surface (Color coding of the atoms: purple, Cs; brown, Br; gray, Pb).

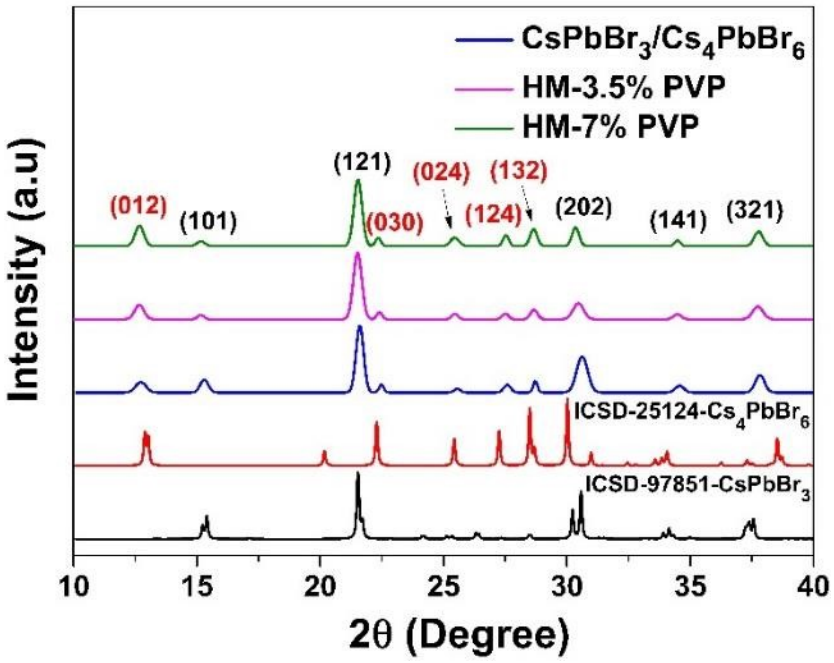

Fig. S2. XRD patterns of dual-phase $CsPbBr_3$/ Cs_4PbBr_6 composites with different concentrations of HM-PVP perovskite films.

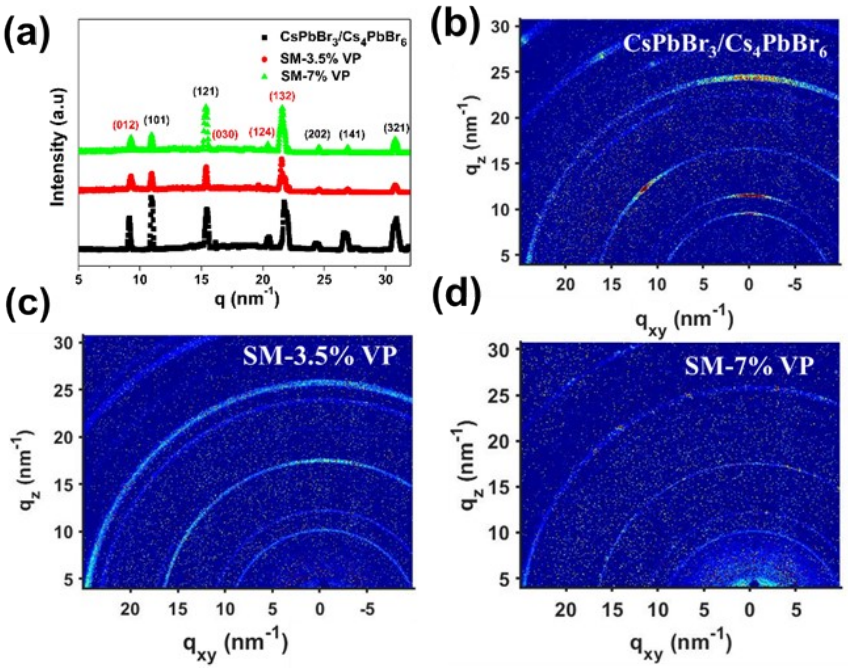

Fig. S3. (a) 1D GIWAXS profile of dual-phase $CsPbBr_3$/ Cs_4PbBr_6 composites at the different concentrations of SM-VP perovskite films. (b-d) Synchrotron 2D GIWAXS pattern of the different concentrations of SM-VP perovskite films.

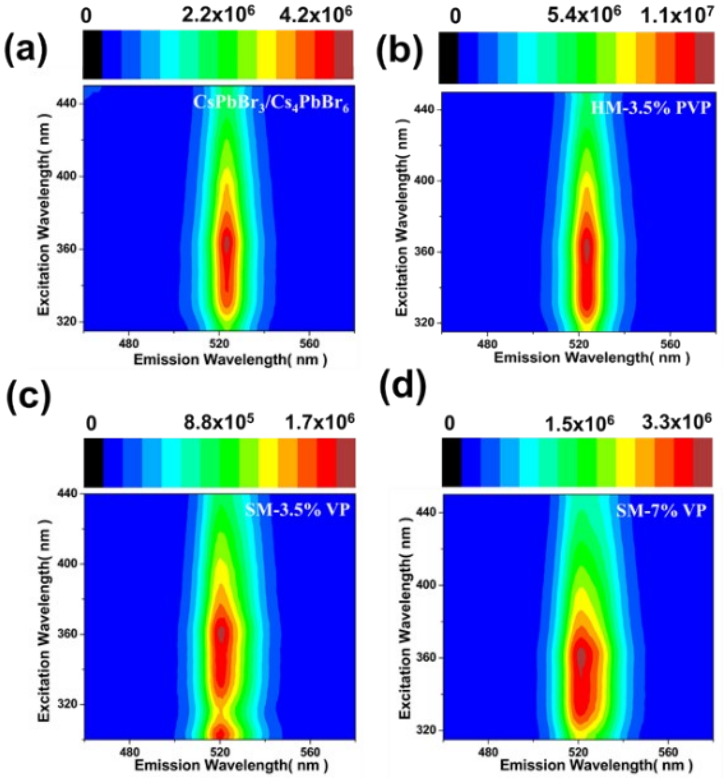

Fig. S4. 2D pattern PLE spectra of (a) $CsPbBr_3$/ Cs_4PbBr_6 (b) HM-3.5% PVP perovskite films, (c) SM-3.5% VP perovskite films, and (d) SM-7% VP perovskite films, respectively.

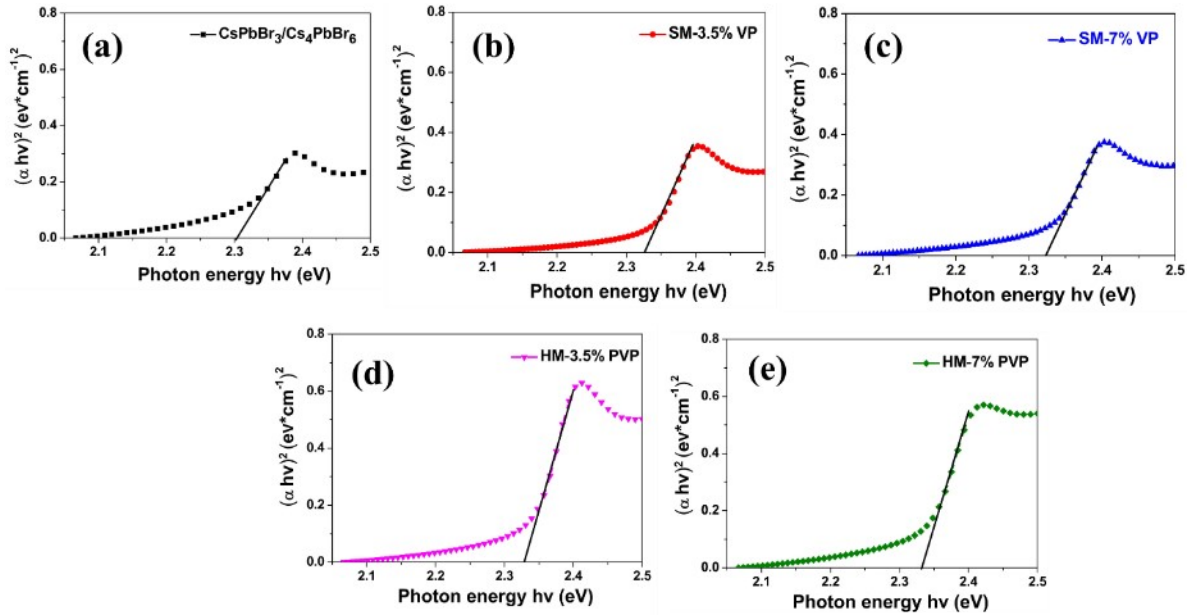

Fig. S5. Tauc plot of absorption spectra of $CsPbBr_3$/ Cs_4PbBr_6 at different concentrations of SM-VP perovskite films and HM-PVP perovskite films.

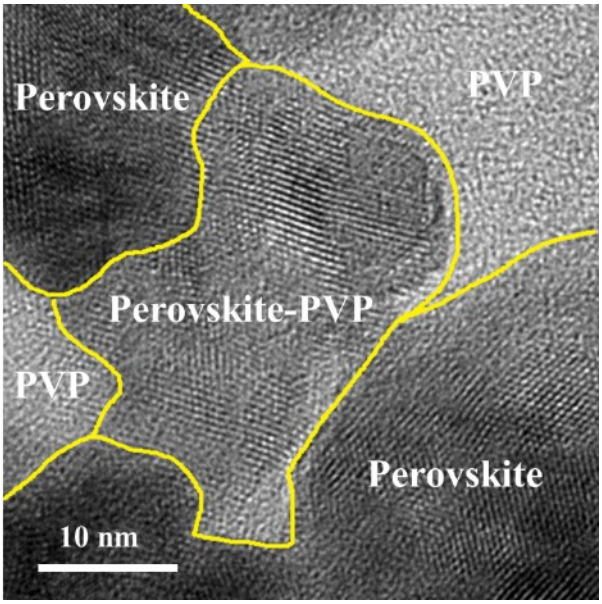

Fig. S6. HRTEM images intermediate phase between the HM-7% PVP (orange color) and perovskite grains (blue color). The yellow lines depicts the regions.

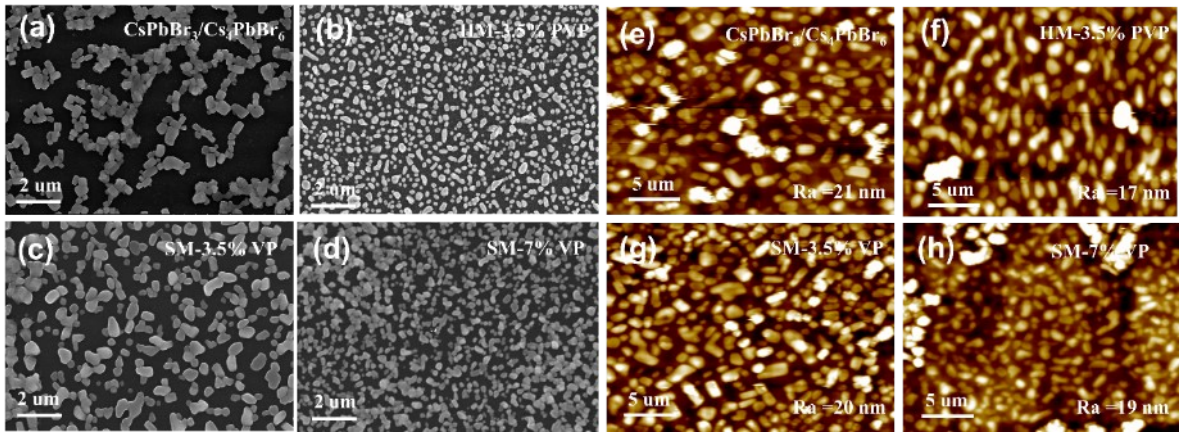

Fig. S7. SEM morphologies of (a) $CsPbBr_3$/ Cs_4PbBr_6, (b)HM-3.5% PVP, (c) SM-3.5% VP, and (d) SM-7% VP, respectively. AFM images of (e) $CsPbBr_3$/ Cs_4PbBr_6, (f) HM-3.5% PVP, (g) SM-3.5% VP, and (h) SM-7% VP, respectively.

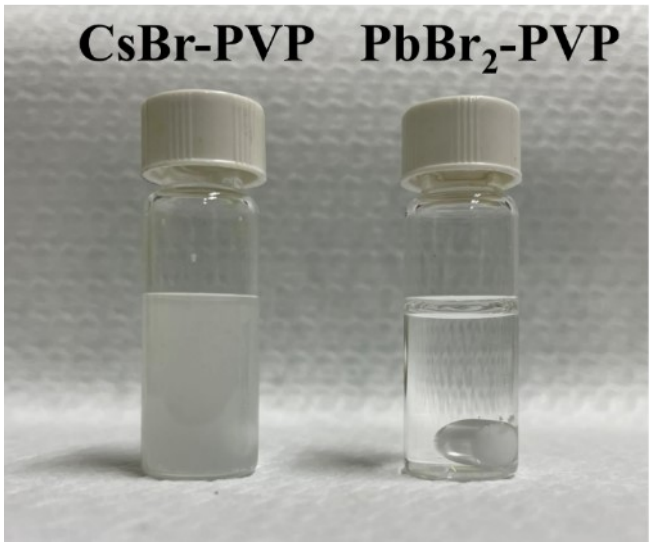

Fig. S8. Photographs of saturated CsBr−PVP, and $PbBr_2$−PVP mixed solution in DMSO.

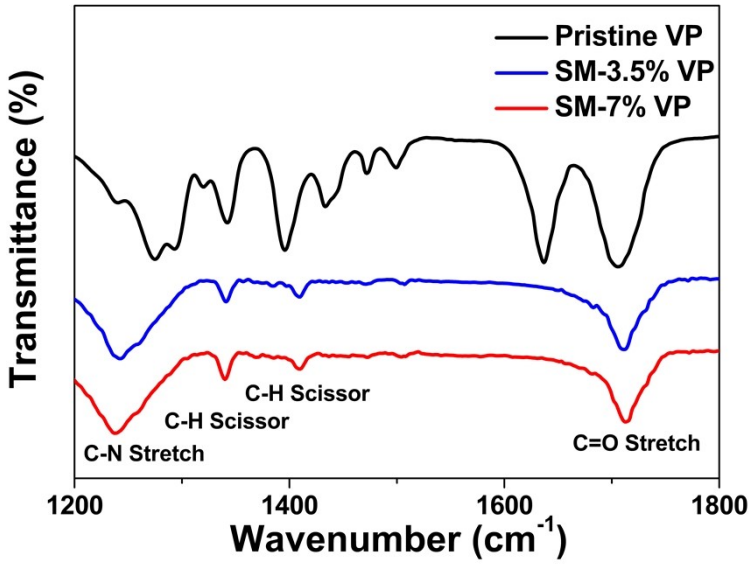

Fig. S9. FTIR spectra of VP at different concentrations of SM-VP perovskite films.

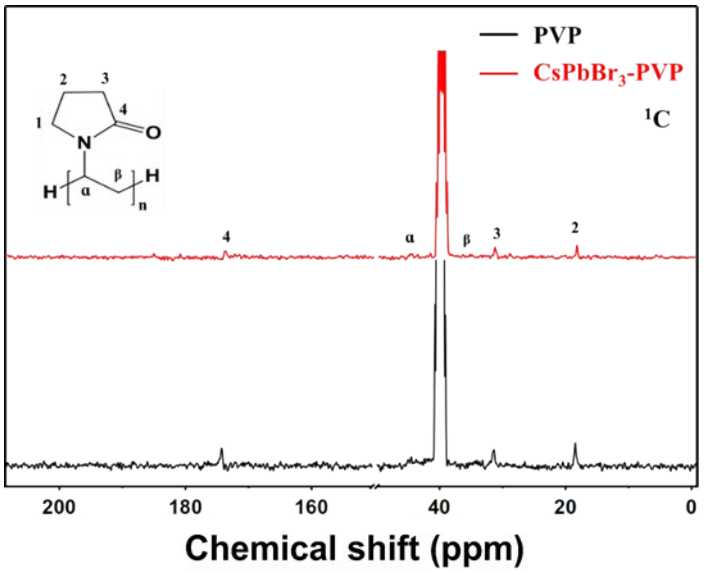

Fig. 10. ^{13}C liquid-state nuclear magnetic resonance (NMR) spectra of PVP solution and PVP-perovskite solution in deuterated DMSO-d6.

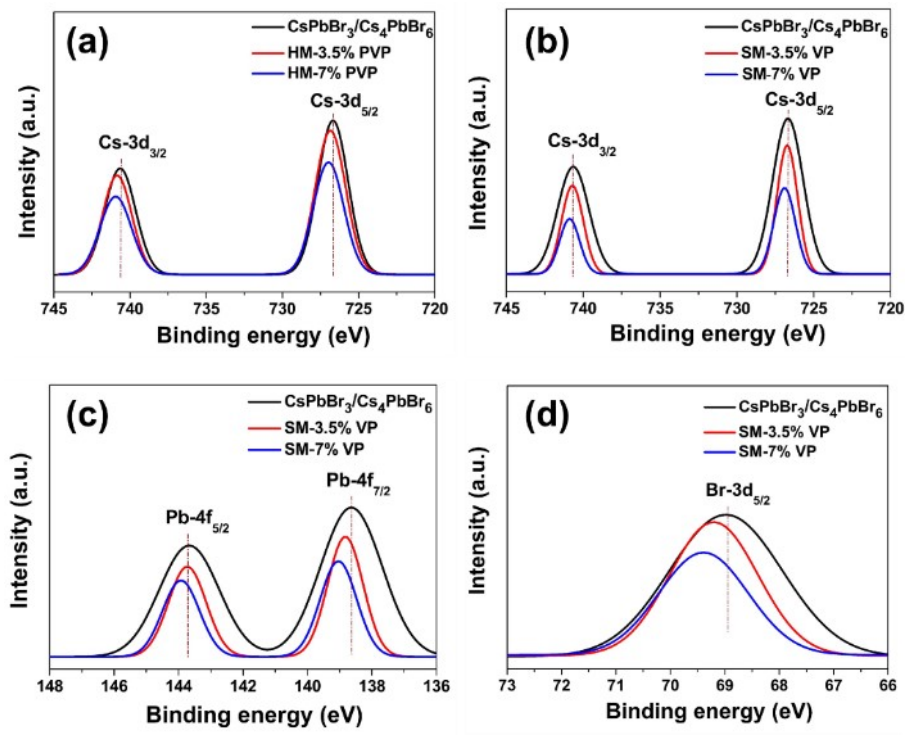

Fig. S11. (a) XPS spectra of the different concentrations of HM-PVP perovskite films for Cs 3d regions (b-d) XPS spectra of the different concentrations of SM-VP perovskite films (b) Cs 3d, (c) Pb 4f, and (d) Br 3d regions.

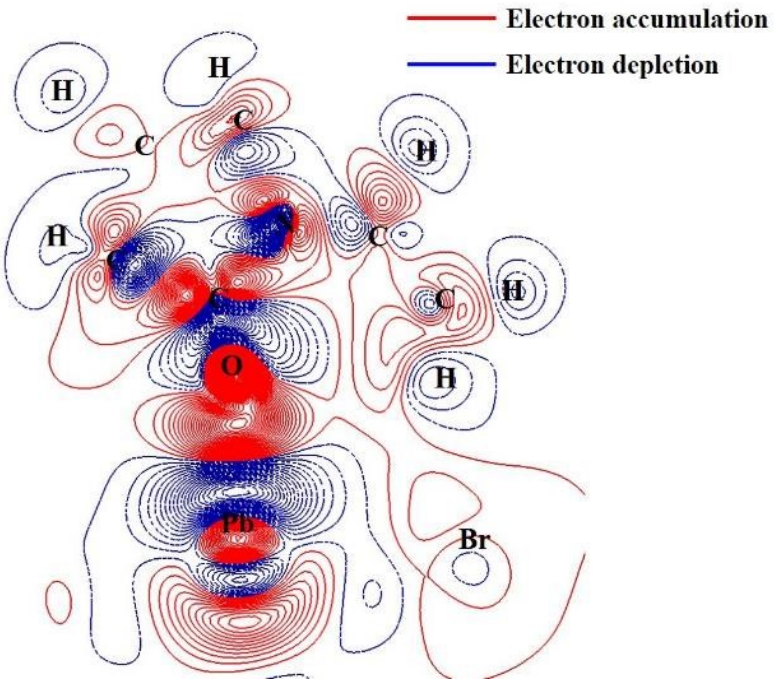

Fig. S12. 2D electron density difference plots of a VP monomer adsorbed on the $CsPbBr_3$ (121) surface. The isosurface value is 0.0005 e a.u. $^{-1}$.

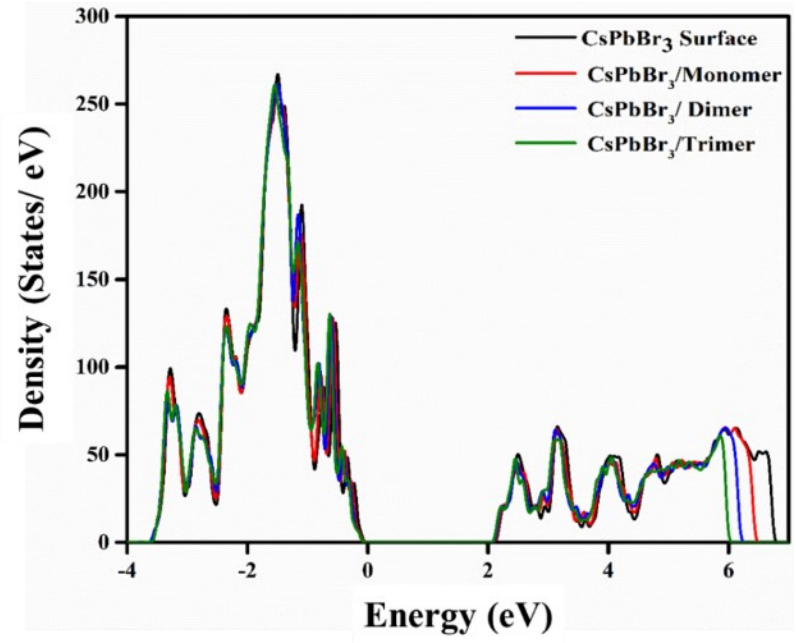

Fig. S13. Calculated density of states (DOS) of pristine $CsPbBr_3$ (121) perovskite surface (black line), and its stable adsorption of the different number of VP units on $CsPbBr_3$ (121) perovskite surface (Red: Monomer; Blue: Dimer; Green: Trimer).

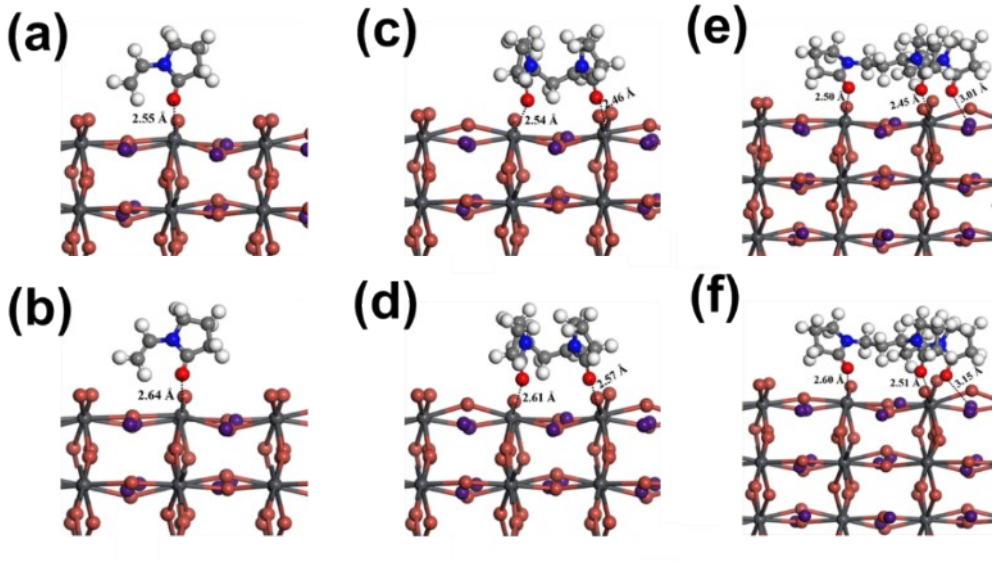

Fig. S14. Optimized structures of VP (a, b) monomer, (c, d) dimer, and (e, f) trimer adsorbed on the $CsPbBr_3$ (121) perovskite surface with hole and electron polaron, respectively. Color coding of the atoms: red, O; white, H; blue, N; light gray, C; purple, Cs; brown, Br; gray, Pb.

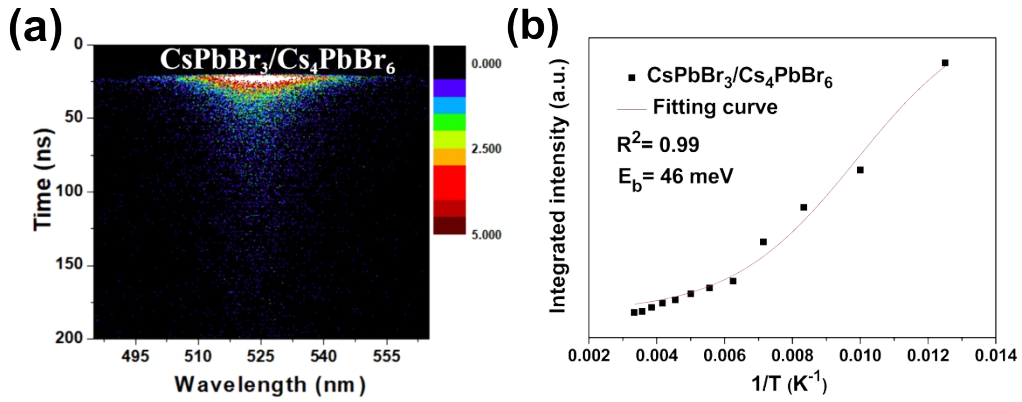

Fig. S15. 2D pseudo-color plot (a) and (b) temperature-dependent PL spectra of $CsPbBr_3/Cs_4PbBr_6$.

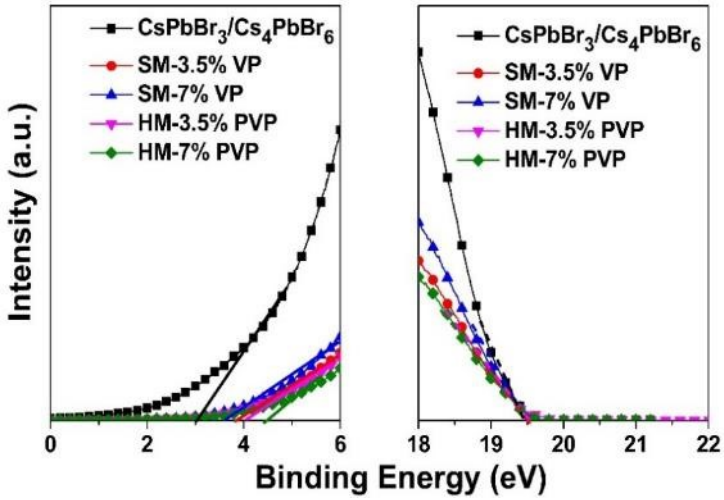

Fig. S16. UPS spectra of $CsPbBr_3$/ Cs_4PbBr_6 at different concentrations of SM-VP perovskite films and HM-PVP perovskite films.

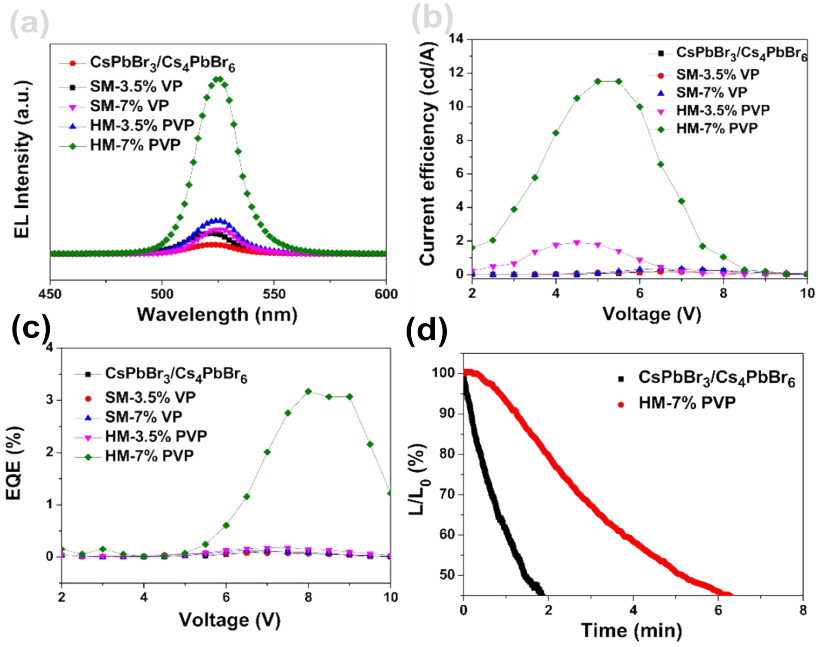

Fig. S17. (a) EL spectra of $CsPbBr_3/Cs_4PbBr_6$ at different concentrations of SM-VP, and HM-PVP. (b) Current efficiency of HM-7% PVP. (b) Current efficiency−voltage and (c) EQE−voltage of $CsPbBr_3/Cs_4PbBr_6$, SM-VP, and HM-PVP. (d) The operational lifetime (T_{50}) with L_0 of 100 cd m^{-2} of $CsPbBr_3/Cs_4PbBr_6$ and HM-7% PVP.

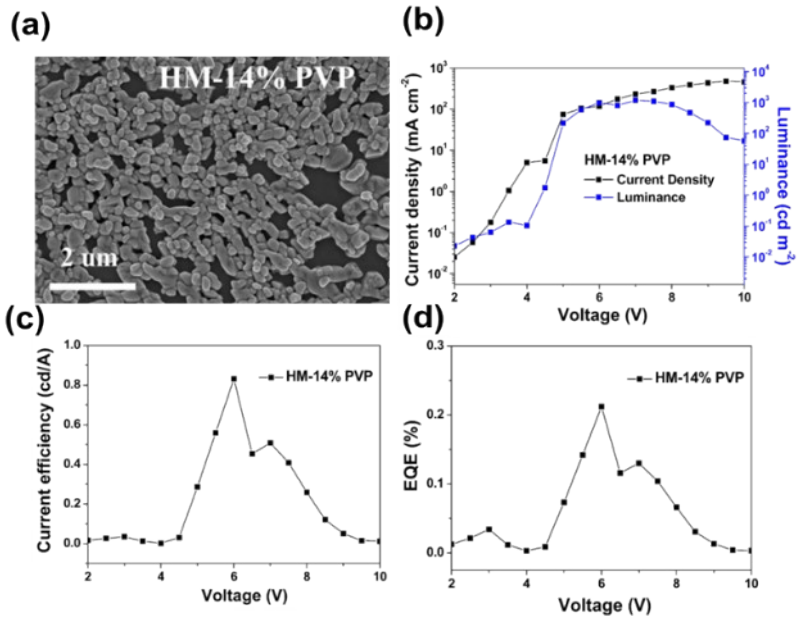

Fig. S18. (a) SEM morphology of HM-14% PVP perovskite composites (b) Current density−voltage and luminance−voltage of HM-14% PVP. (c,d) Current efficiency and EQE of HM-14% PVP perovskite.

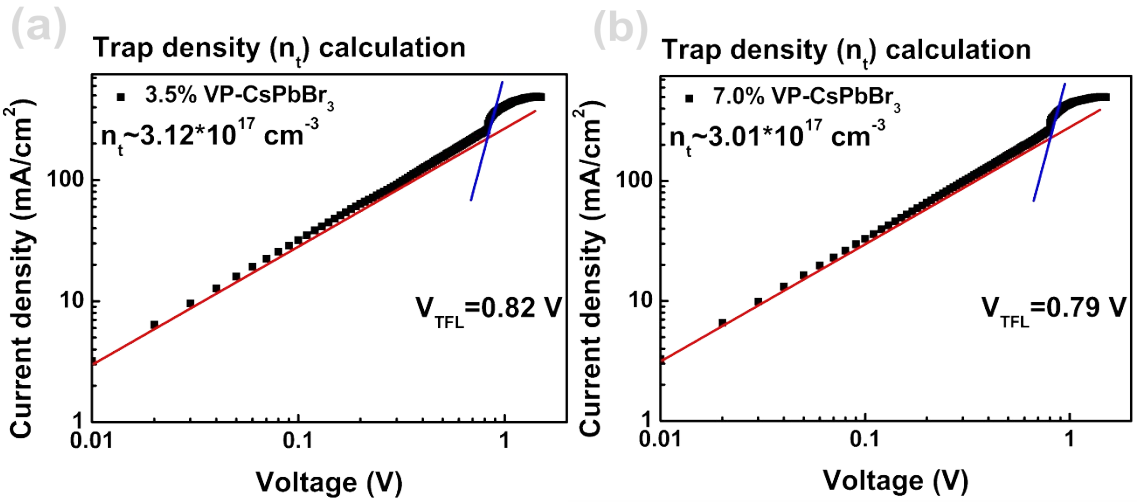

Fig. S19. (a,b) J-V characteristics of hole-only device. (ITO/PEDOT:PSS/3.5% SM-VP and 7% SM-VP / Au)

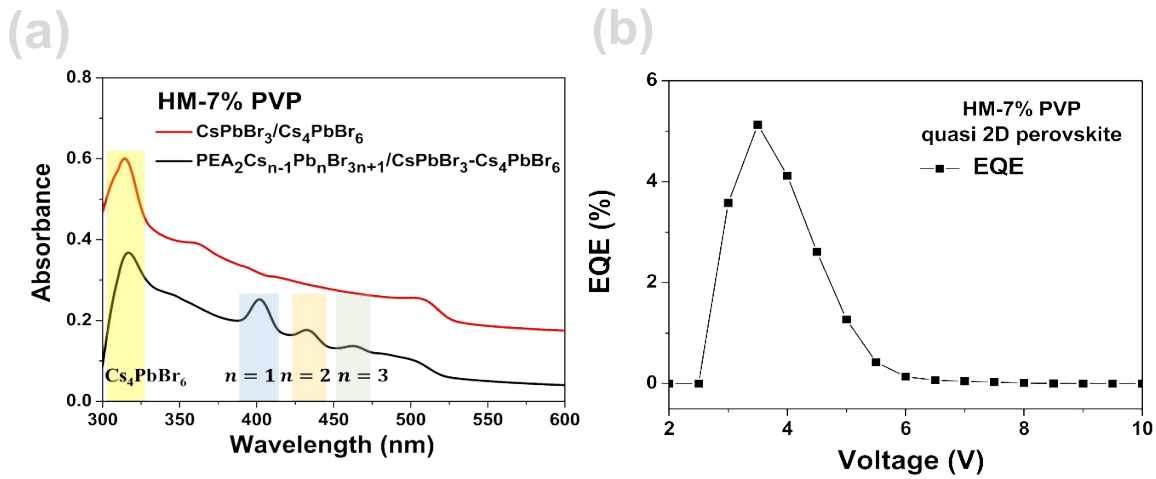

Fig. S20. (a)UV absorption of HM-7% PVP bulk 3D $CsPbBr_3/Cs_4PbBr_6$ composites and HM-7% PVP-Quasi 2D $CsPbBr_3/Cs_4PbBr_6$ composites. (b) EQE−voltage of HM-7% PVP-Quasi 2D $CsPbBr_3/Cs_4PbBr_6$ composites.

Table S1. The calculated surface energies for the different terminations of the $CsPbBr_3$ (121) surface.

Surface termination No.	Surface Energy ($eV/Å^2$)	Termination atom
1	0.119	Cs, Pb, Br
2	0.132	Br
3	0.119	Cs, Pb, Br
4	0.132	Cs, Pb, Br
5	0.145	Cs, Pb, Br
6	0.132	Cs, Pb, Br
7	0.069	Cs, Pb, Br
8	0.115	Br
9	0.170	Br
10	0.081	Cs, Br
11	0.118	Cs, Pb, Br
12	0.130	Cs, Pb, Br
13	0.057	Cs, Pb, Br
14	0.097	Br
15	0.151	Br
16	0.072	Cs, Br
17	0.056	Cs, Pb, Br
18	0.058	Cs, Br
19	0.082	Cs, Pb, Br

Table S2. The geometrical parameters (interatomic distances in Å) of the different number of VP units adsorption on the $CsPbBr_3$ (121) surface at initial configuration, number in parentheses represents the total number of distances less than 4 Å.

	Distance*		
	$\mathbf{d_{O\text{-}Pb}}$	$\mathbf{d_{H\text{-}Br}}$	$\mathbf{d_{O\text{-}Cs}}$
Monomer	2.57	2.30~3.26(2)	-
Dimer	2.62~2.69(2)	2.77~3.36(6)	-
Trimer	2.21~2.91(2)	2.00~3.73(9)	2.53

Co-ordinates

VP monomer adsorption on the $CsPbBr_3$ (121) perovskite surface

23.206400000000 0.000000000000 0.000000000000
-0.400114655638 16.543062070619 0.000000000000
0.000000000000 0.000000000000 36.540600000000

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0.495690000000 0.222590000000 0.012260000000
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0.205650000000 0.939890000000 0.013100000000
0.345270000000 0.094480000000 0.042820000000
0.095810000000 0.625900000000 0.056800000000
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0.094950000000 0.071810000000 0.081710000000
0.234570000000 0.226400000000 0.111430000000
0.499530000000 0.445480000000 0.111440000000

0.444530000000	0.943700000000	0.112270000000
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VP dimer adsorption on the $CsPbBr_3$ (121) perovskite surface

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0.095810000000	0.625900000000	0.056800000000
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0.094950000000	0.071810000000	0.081710000000
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0.499530000000	0.445480000000	0.111440000000
0.444530000000	0.943700000000	0.112270000000
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0.460040000000	0.694200000000	0.126260000000

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0.199660000000	0.697700000000	0.224540000000
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0.208220000000	0.952690000000	0.239070000000
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0.448750000000	0.227840000000	0.239640000000
0.211060000000	0.450230000000	0.240020000000
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0.103850000000	0.135960000000	0.283100000000
0.352310000000	0.047710000000	0.292720000000
0.103770000000	0.581970000000	0.307070000000
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0.007210000000	0.945720000000	0.336630000000
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0.249200000000	0.240180000000	0.338420000000
0.466430000000	0.707890000000	0.351520000000
0.253050000000	0.480970000000	0.350980000000
0.207040000000	0.981850000000	0.353280000000
0.466510000000	0.206870000000	0.350840000000
0.107200000000	0.351970000000	0.382350000000
0.359880000000	0.884780000000	0.391660000000
0.108590000000	0.798760000000	0.400610000000
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0.253300000000	0.685000000000	0.444620000000
0.217990000000	0.180480000000	0.442560000000
0.989150000000	0.018150000000	0.440100000000
0.701750000000	0.443650000000	0.011360000000
0.995690000000	0.222590000000	0.012260000000
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0.595810000000	0.625900000000	0.056800000000

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0.608590000000	0.798760000000	0.400610000000
0.857200000000	0.338070000000	0.421440000000

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0.088700000000 0.603690000000 0.167950000000
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0.360280000000 0.071230000000 0.182910000000
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0.083470000000 0.352530000000 0.283870000000
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0.588700000000 0.603690000000 0.167950000000
0.832790000000 0.597780000000 0.170680000000

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0.474490000000 0.837460000000 0.175430000000

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0.224490000000	0.337460000000	0.175430000000
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0.720110000000	0.083140000000	0.062270000000
0.720110000000	0.583140000000	0.062270000000
0.970110000000	0.583140000000	0.062270000000
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0.974490000000	0.837460000000	0.175430000000
0.974490000000	0.337460000000	0.175430000000
0.724490000000	0.337460000000	0.175430000000
0.979060000000	0.595280000000	0.290440000000
0.729500000000	0.589670000000	0.289330000000
0.727730000000	0.088230000000	0.290030000000
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0.732720000000	0.327010000000	0.404910000000
0.982390000000	0.359340000000	0.404520000000
0.983630000000	0.868090000000	0.405100000000
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VP trimer adsorption on the $CsPbBr_3$ (121) perovskite surface

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0.345270000000	0.094480000000	0.042820000000

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0.448750000000	0.227840000000	0.239640000000
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0.234110000000 0.827990000000 0.404950000000

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0.720110000000	0.083140000000	0.062270000000
0.720110000000	0.583140000000	0.062270000000
0.970110000000	0.583140000000	0.062270000000
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0.974490000000	0.837460000000	0.175430000000
0.974490000000	0.337460000000	0.175430000000
0.724490000000	0.337460000000	0.175430000000
0.979060000000	0.595280000000	0.290440000000
0.729500000000	0.589670000000	0.289330000000
0.727730000000	0.088230000000	0.290030000000
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0.732720000000	0.327010000000	0.404910000000
0.982390000000	0.359340000000	0.404520000000
0.983630000000	0.868090000000	0.405100000000
0.734110000000	0.827990000000	0.404950000000

Table S3. Calculated adsorption energies (E_{ads}) and change in Bader charges (ΔQ) upon adsorption of the different number of VP units on the $CsPbBr_3$ (121) perovskite surface.

VP units	**E_{ads} (eV)**	**ΔQ (e)**
Monomer	-0.83	0.04
Dimer	-1.69	0.06
Trimer	-3.02	0.04

Table S4. Calculated adsorption energies (E_{ads}) and change in Bader charges (ΔQ) upon adsorption of the different number of VP units on the $CsPbBr_3$ (121) perovskite surface in the presence of an electron and hole polaron.

VP units	E_{ads} (eV)	ΔQ (e)
Monomer (h^+)	-0.94	0.05
Monomer (e^-)	-0.58	0.02
Dimer (h^+)	-2.23	0.08
Dimer (e^-)	-1.55	0.04
Trimer (h^+)	-3.09	0.10
Trimer (e^-)	-2.30	0.02

h^+ and e^- denote the surface with hole and electron polaron, respectively.

Table S5. A summary of TRPL and PLQY decay curves of $CsPbBr_3/Cs_4PbBr_6$, SM-VP, and HM-PVP.

Materials	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)	τ_{av} (ns)	*PLQY* (Film) (%)
$CsPbBr_3/Cs_4PbBr_6$	3.87	0.87	26.05	0.17	16.47	10
SM-3.5% VP	6.67	14.37	20.79	1.66	10.42	15
SM-7% VP	7.53	10.32	23.75	1.35	12.27	23
HM-3.5% PVP	2.73	0.85	24.36	0.2	17.37	31
HM-7% PVP	6.21	0.49	33.17	0.48	28.84	45

Table S6. The characteristic of $CsPbBr_3/Cs_4PbBr_6$, SM-VP, HM-PVP PeLEDs performance and HM-PVP 7% quasi 2D PeLEDs performance.

Material	V_{on} [V]	Max. luminance [cd m^{-2}]	Max. CE [cd A^{-1}]	Max. EQE [%]
PEDOT:PSS-AgNPs /$CsPbBr_3$-Cs_4PbBr_6	3.5	2075	0.28	0.108
PEDOT:PSS-AgNPs /SM-3.5% VP	3.5	1265	0.16	0.07
PEDOT:PSS-AgNPs /SM-7% VP	3.5	2560	0.37	0.125
PEDOT:PSS-AgNPs /HM-3.5% PVP	3.5	3645	0.62	0.18
PEDOT:PSS-AgNPs /HM-7% PVP	3	12000	11.5	3.17
PEDOT:PSS-AgNPs /Poly-TPD/PVK/HM-7% PVP quasi 2D	3	9500	15.4	5.13