

**Vertically oriented two-dimensional Ruddlesden–Popper phase perovskite passivation layer
for efficient and stable inverted perovskite solar cells**

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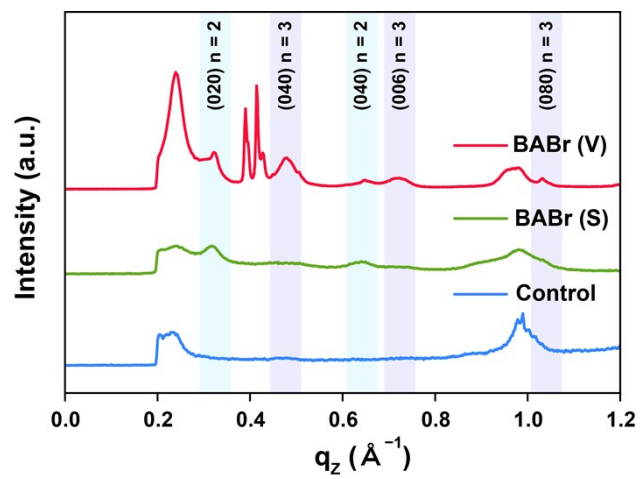


Fig. S1 Signal intensity profiles of GIWAXS patterns along q_z axis.

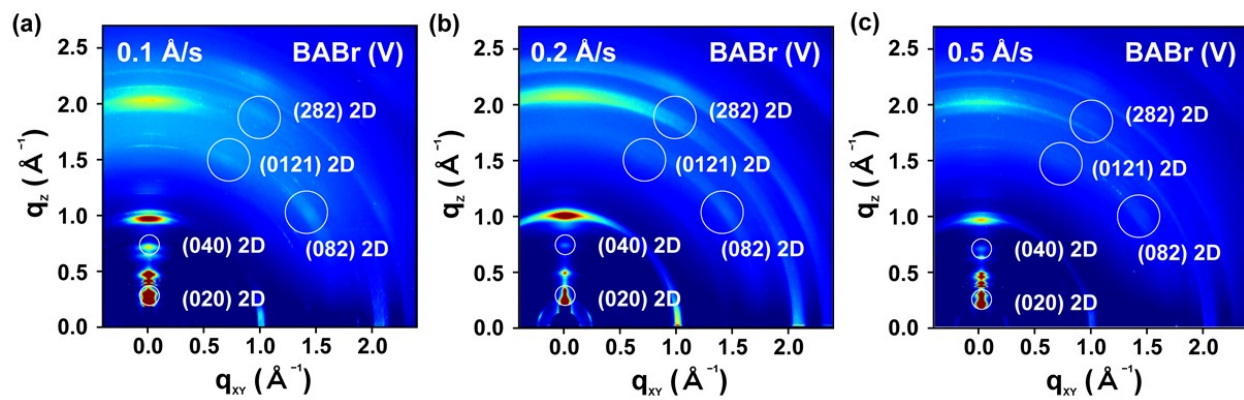


Fig. S2 GIWAXS patterns of BABr (V) films with RP phase perovskite deposition rates of (a) 0.1, (b) 0.2, and (c) 0.5 $\text{\AA}/\text{s}$.

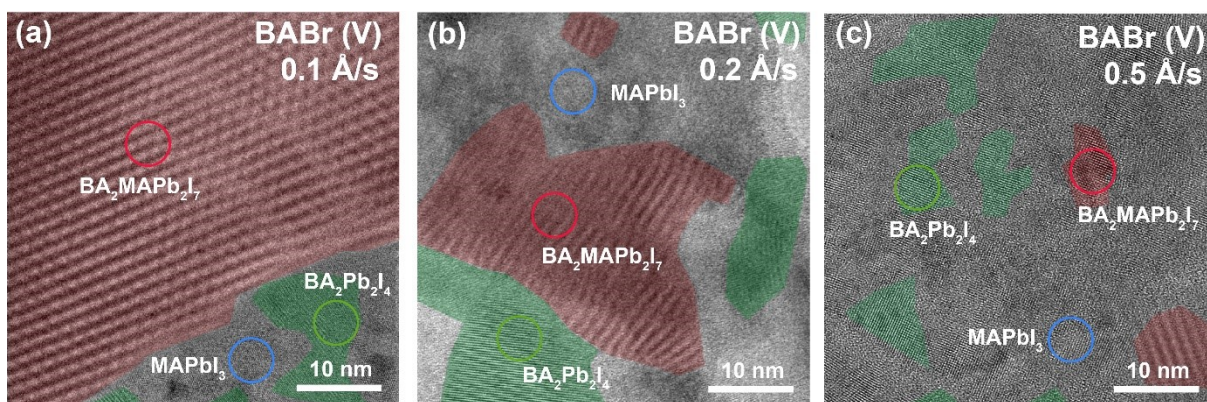


Fig. S3 HRTEM images of BABr (V) films with RP phase perovskite deposition rates of (a) 0.1, (b) 0.2, and (c) 0.5 $\text{\AA}/\text{s}$.

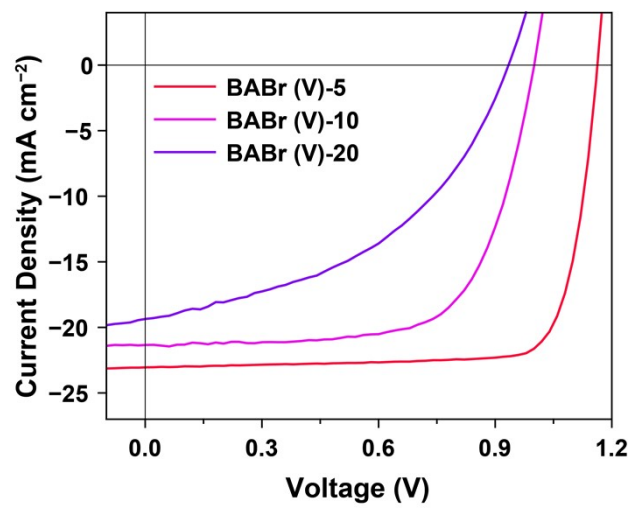


Fig. S4 J – V characteristics of BABr (V) PSCs with different RP perovskite deposition times.

Table S1 Photovoltaic parameters of BABr (V) PSCs with different RP perovskite deposition times.

Perovskite	J_{SC} [mA cm ⁻²]	V_{OC} [V]	FF	PCE [%]
BABr (V)-5	23.0	1.16	0.80	21.4
BABr (V)-10	21.1	1.00	0.66	13.9
BABr (V)-20	18.8	0.94	0.44	7.8

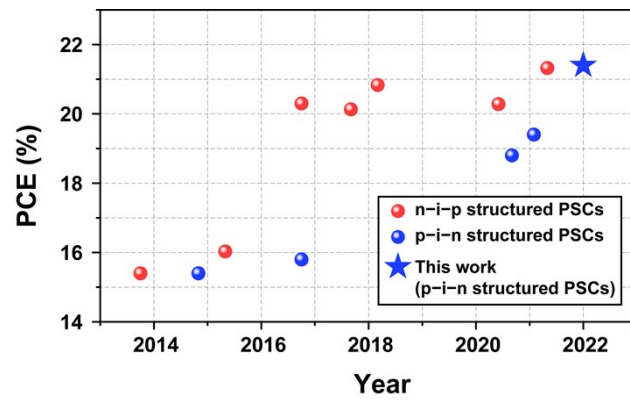


Fig. S5 Comparison of device performance for PSCs with vacuum-deposited perovskite active layer reported in literature.

Table S2 Photovoltaic parameters of PSCs with vacuum-deposited perovskite active layer reported in literature.

Year	Structure of PSCs	J_{sc} [mA cm ⁻²]	V_{oc} [V]	FF	PCE [%]	Ref
2013	n-i-p	21.5	1.07	0.67	15.4	[1]
2014	p-i-n	20.9	1.02	0.72	15.4	[2]
2015	n-i-p	22.3	1.00	0.72	16.0	[3]
2016	n-i-p	22.1	1.14	0.81	20.3	[4]
2016	p-i-n	20.0	1.08	0.73	15.8	[5]
2017	n-i-p	23.2	1.10	0.79	20.1	[6]
2018	n-i-p	21.9	1.16	0.82	20.8	[7]
2020	p-i-n	22.6	1.09	0.76	18.8	[7]
2020	n-i-p	23.3	1.12	0.78	20.3	[8]
2021	p-i-n	23.1	1.09	0.77	19.4	[9]
2021	n-i-p	24.9	1.11	0.77	21.3	[10]
2022	p-i-n	23.0	1.16	0.80	21.4	This work

Table S3 Fitted TRPL parameters for perovskite films with passivation layer.

Perovskite	τ_1 [ns]	Fraction 1 [%]	τ_2 [ns]	Fraction 2 [%]
Control	5.2	62	70.5	38
BABr (S)	3.1	71	33.8	29
BABr (V)	2.7	77	25.7	23

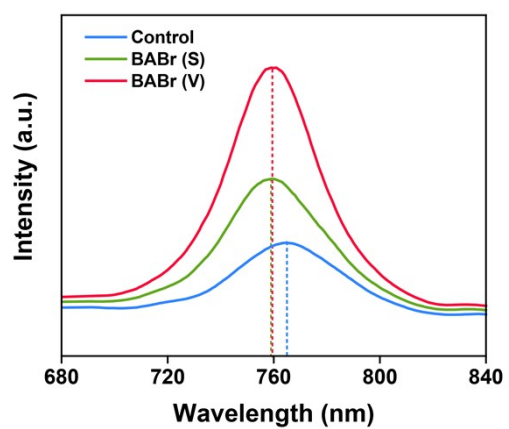


Fig. S6 Steady-state photoluminescence spectra of perovskite films with RP phase perovskite passivation layer.

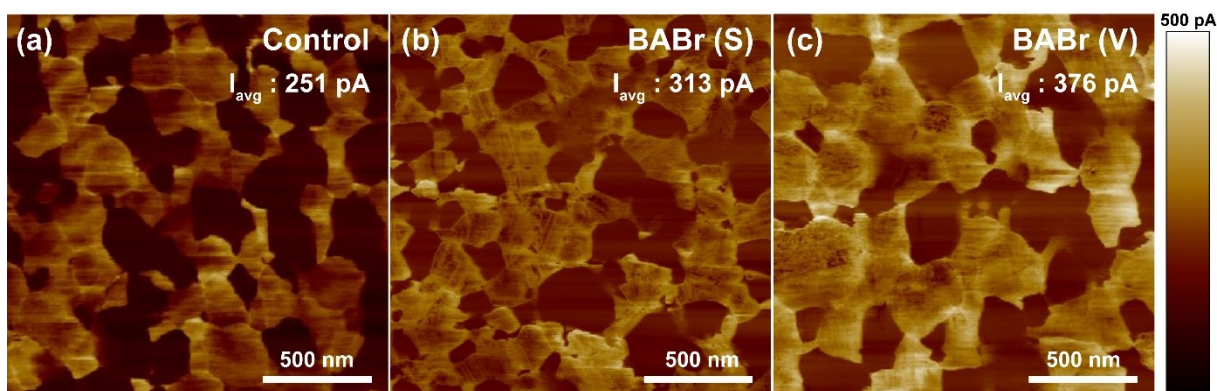


Fig. S7 Conductive AFM images of (a) control, (b) BABr (S), and (c) BABr (V) films.

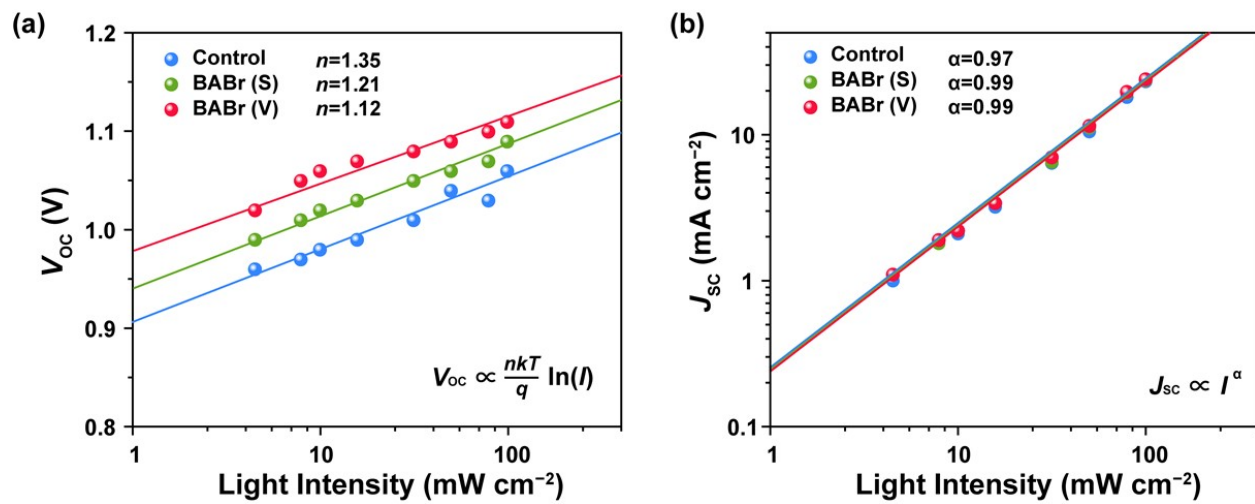


Fig. S8 Light-intensity-dependent (a) V_{OC} and (b) J_{SC} profiles for PSCs with RP phase perovskite passivation layer.

Table S4 Hole and electron trap density and carrier mobility values obtained by space-charge limited current analysis.

Perovskite	N_{th} [cm ⁻³]	N_{te} [cm ⁻³]	μ_{h} [cm ² V ⁻¹ s ⁻¹]	μ_{e} [cm ² V ⁻¹ s ⁻¹]
Control	1.86×10^{16}	5.44×10^{15}	1.2×10^{-4}	8.2×10^{-4}
BABr (S)	1.31×10^{16}	3.74×10^{15}	8.6×10^{-4}	1.2×10^{-3}
BABr (V)	1.11×10^{16}	3.33×10^{15}	1.0×10^{-3}	1.4×10^{-3}

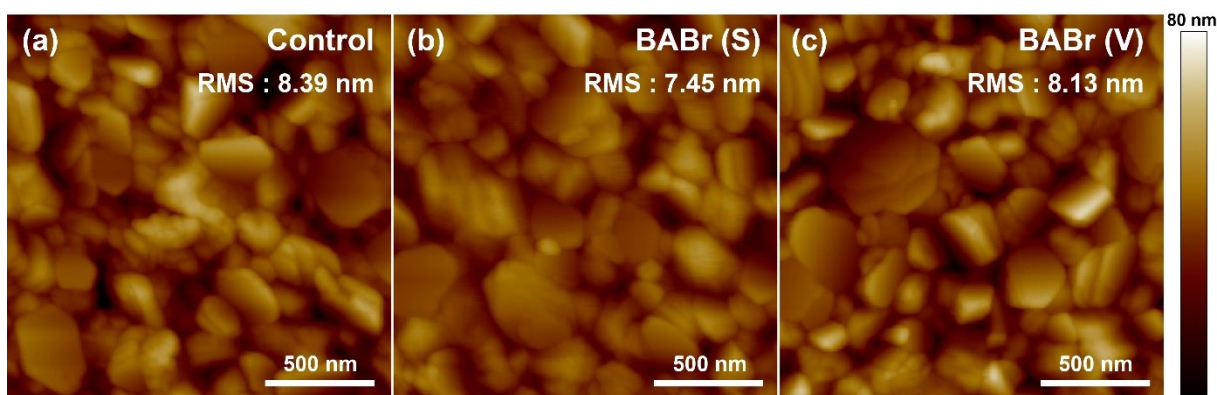


Fig. S9 AFM topography images of (a) control, (b) BABr (S), and (c) BABr (V) films.

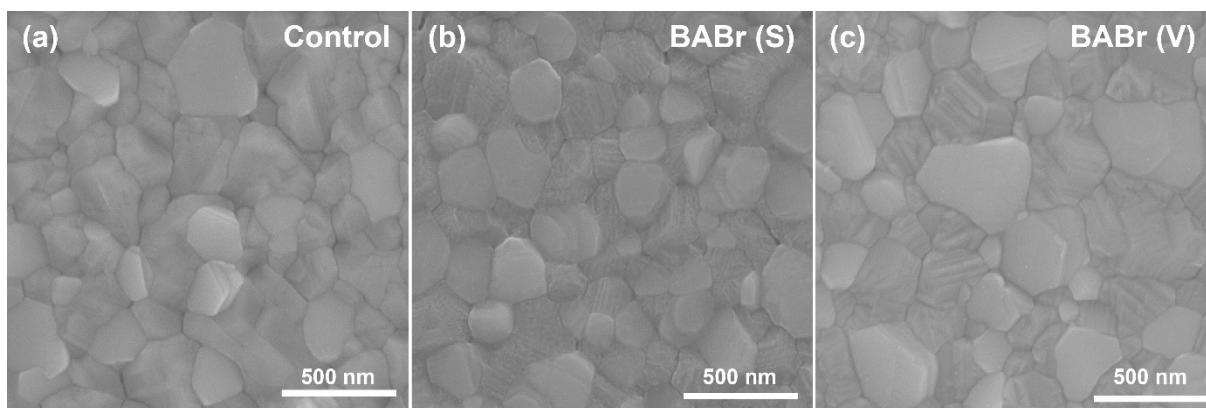


Fig. S10 Top-view SEM images of (a) control, (b) BABr (S), and (c) BABr (V) films.

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