Supplementary Information

Situ growth of 2D/3D mixture perovskite interface layer by seed-mediated and solvent-assisted Ostwald ripening for stable and efficient photovoltaics

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1 the authors made same contribution in this paper.

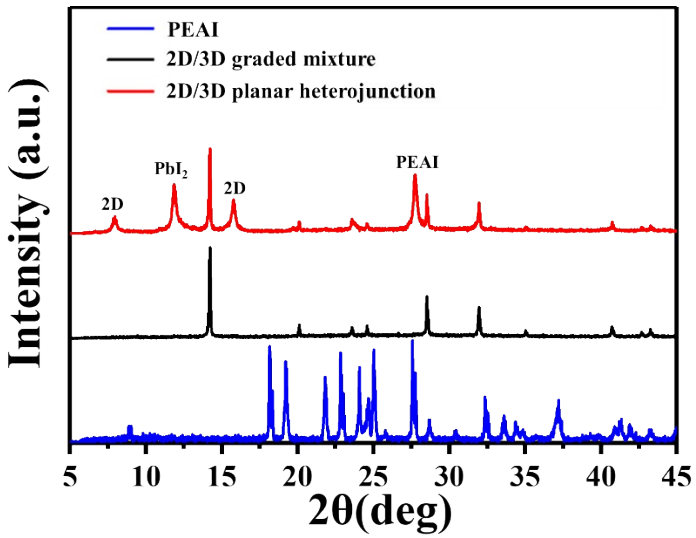

Figure S1 XRD patterns of PEAI and 2D/3D perovskite films based on higher concentrations of PEAI in isopropanol (4mg/ml).

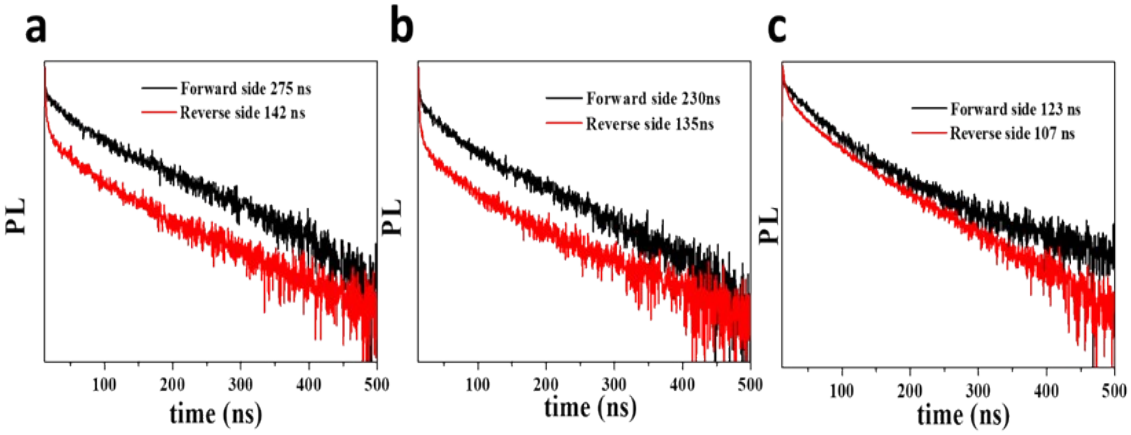

Figure S2 Time-resolved PL decay curves of (**a)** 2D/3D gradient mixture perovskite film, (**b)** 2D/3D planar heterojunction perovskite film and (**c)** regular 3D perovskite film measured from frond and back side, respectively

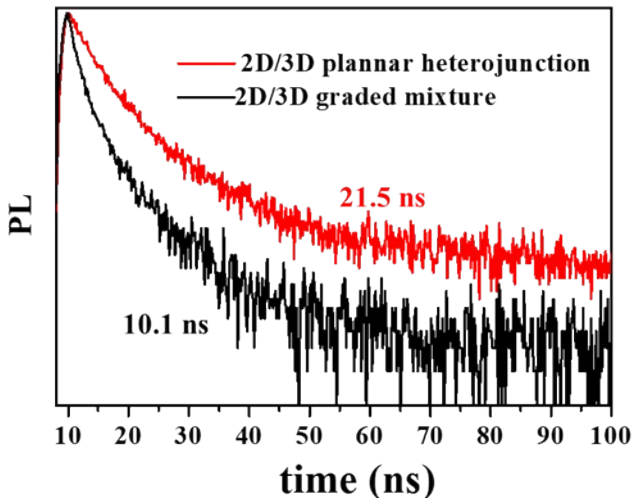

Figure S3. Time-resolved PL decay curves for 2D/3D gradient and 2D/3D planar heterojunction films with [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) as electron extraction layer.

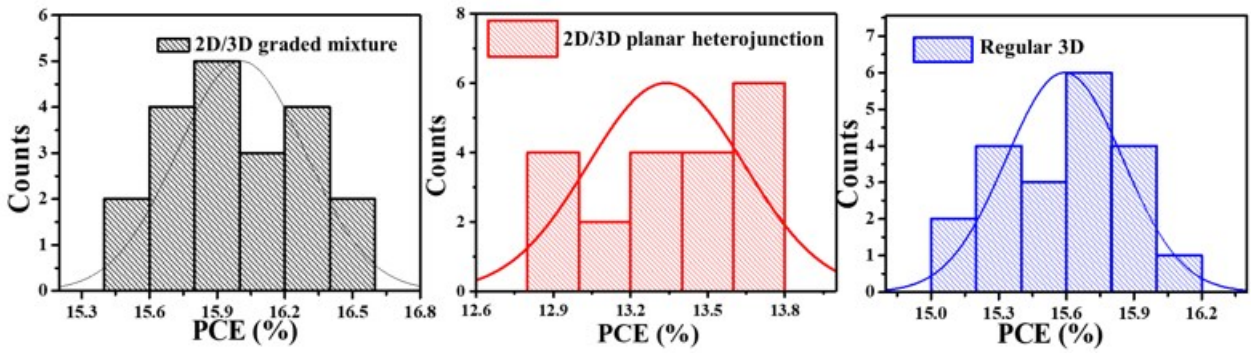

Figure S4 The PCE histogram of 2D/3D gradient mixture, 2D/3D planner

heterojunction, and regular 3D PSCs.

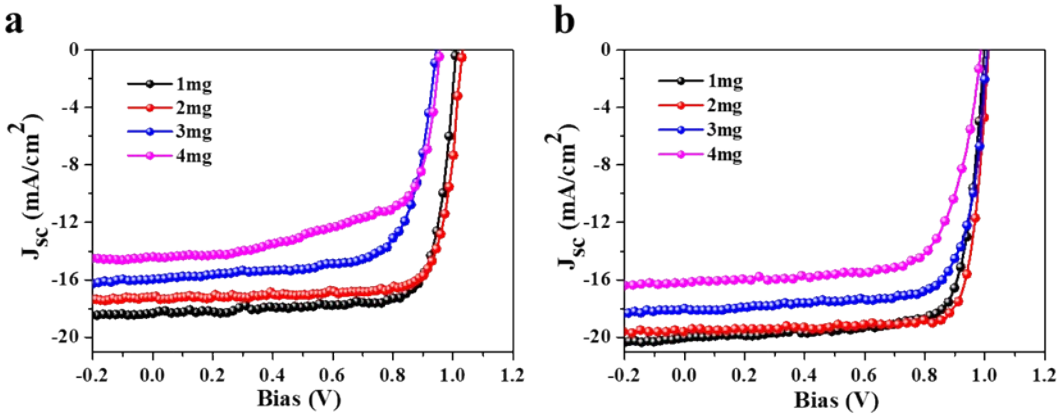

Figure S5. J-V characteristics of (**a**) 2D/3D planar heterojunction cells with different PEAI concentration, (**b**) 2D/3D gradient mixture cells with different PEAI concentration.

Table S1 Performance parameters of the PSCs based on 2D/3D mixture perovskite film made by PEAI-treatment with different concentration DMF additive. The data were obtained based on 20 devices for each kind.

DMF	V_{oc}（V）	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	BEST PCE (%)
without	1.04±0.01	16.88±0.26	76.3±0.7	13.34±0.29	13.72
0.5%	1.03±0.01	18.27±0.28	79.9±0.6	15.04±0.33	15.52
1%	1.02±0.01	19.52±0.36	80.2±0.4	16.01±0.28	16.46
1.5%	0.97±0.01	19.55±0.33	75.8±0.6	14.39±0.42	14.91

Table S2 Performance parameters of the PSCs based on 2D/3D mixture perovskite film made by PEAI-treatment (without DMF) with different PEAI concentration. The data were obtained based on 20 devices for each kind.

concentration	V_{oc}（V）	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	BEST PCE (%)
1mg	1.01±0.01	18.25±0.52	77.1±0.4	14.21±0.30	14.68
2mg	1.04±0.01	16.88±0.26	76.3±0.7	13.34±0.29	13.72
3mg	1.00±0.01	15.54±0.59	71.1±0.5	11.04±0.37	11.63
4mg	0.96±0.01	14.23±0.33	65.4±0.8	8.93±0.44	9.54

Table S3 Performance parameters of the PSCs based on 2D/3D mixture perovskite film made by PEAI-treatment (with 1% DMF) with different PEAI concentration. The data were obtained based on 20 devices for each kind.

concentration	V_{oc}（V）	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	BEST PCE (%)
1mg	1.00±0.01	19.98±0.43	78.9±0.4	15.77±0.35	16.29
2mg	1.02±0.01	19.52±0.36	80.2±0.4	16.01±0.28	16.46
3mg	1.01±0.01	18.14±0.61	75.8±0.5	13.89±0.33	14.54
4mg	0.98±0.01	16.39±0.56	70.2±0.6	11.28±0.46	11.90

Table S4 Performance parameters of the PSCs based on 2D/3D mixture perovskite film made by BAI-treatment with different concentration DMF additive. The data were obtained based on 20 devices for each kind.

DMF	V_{oc}（V）	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	BEST PCE (%)
Without	1.05±0.02	16.54±0.61	77.1±0.8	13.40±0.29	13.88
0.5%	1.02±0.01	18.06±0.44	79.5±0.7	14.64±0.33	15.15
1%	1.02±0.01	19.21±0.36	80.0±0.4	15.67±0.28	16.17
1.5%	0.99±0.02	19.06±0.39	75.1±0.6	14.17±0.39	14.71

Table S5 Performance parameters of the PSCs based on 2D/3D mixture perovskite film made by PAI-treatment with different concentration DMF additive. The data were obtained based on 20 devices for each kind.

DMF	V_{oc}（V）	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	BEST PCE (%)
Without	1.02±0.01	18.15±0.53	77.9±0.6	14.45±0.33	14.97
0.5%	0.99±0.01	19.12±0.69	79.1±0.4	15.02±0.37	15.54
1%	0.99±0.01	19.57±0.61	79.6±0.5	15.43±0.41	15.95
1.5%	0.97±0.01	18.43±0.55	74.4±0.7	13.37±0.53	13.99