

## Supporting Information

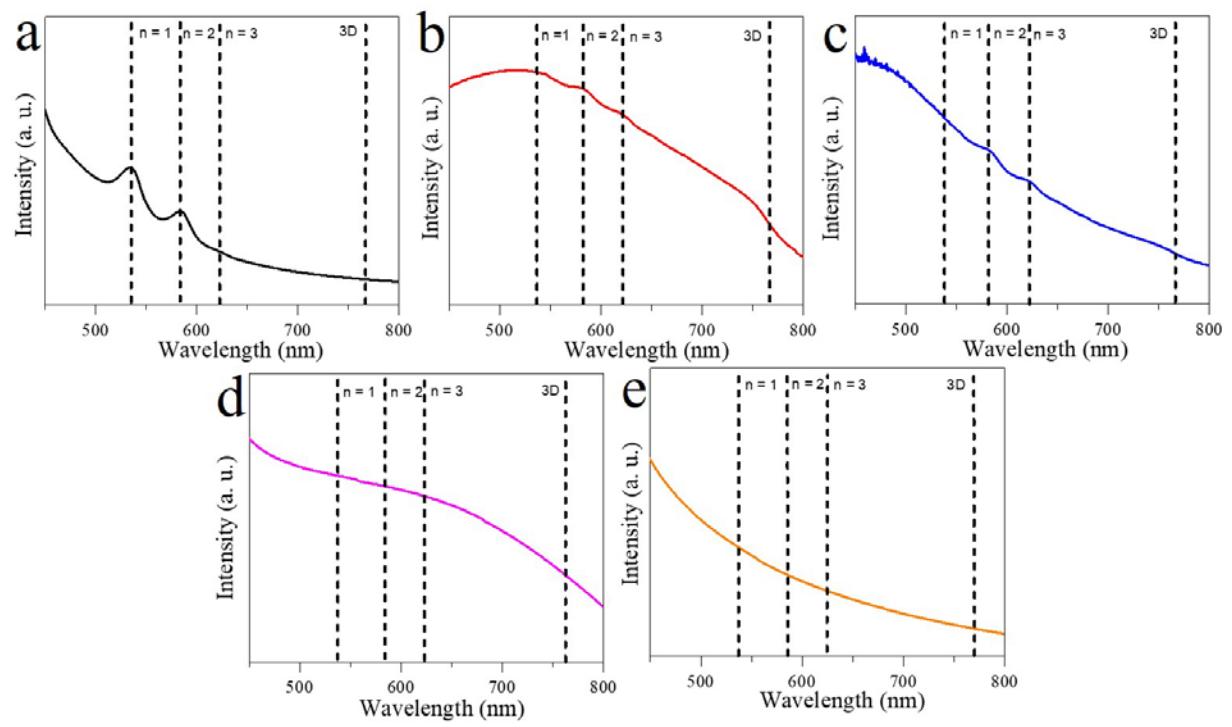
# A Simple Method for Phase control in Two-dimensional Perovskite Solar Cells

Chunqing Ma,<sup>ab</sup> Ming-Fai Lo<sup>\*ab</sup> and Chun-Sing Lee<sup>\*ab</sup>

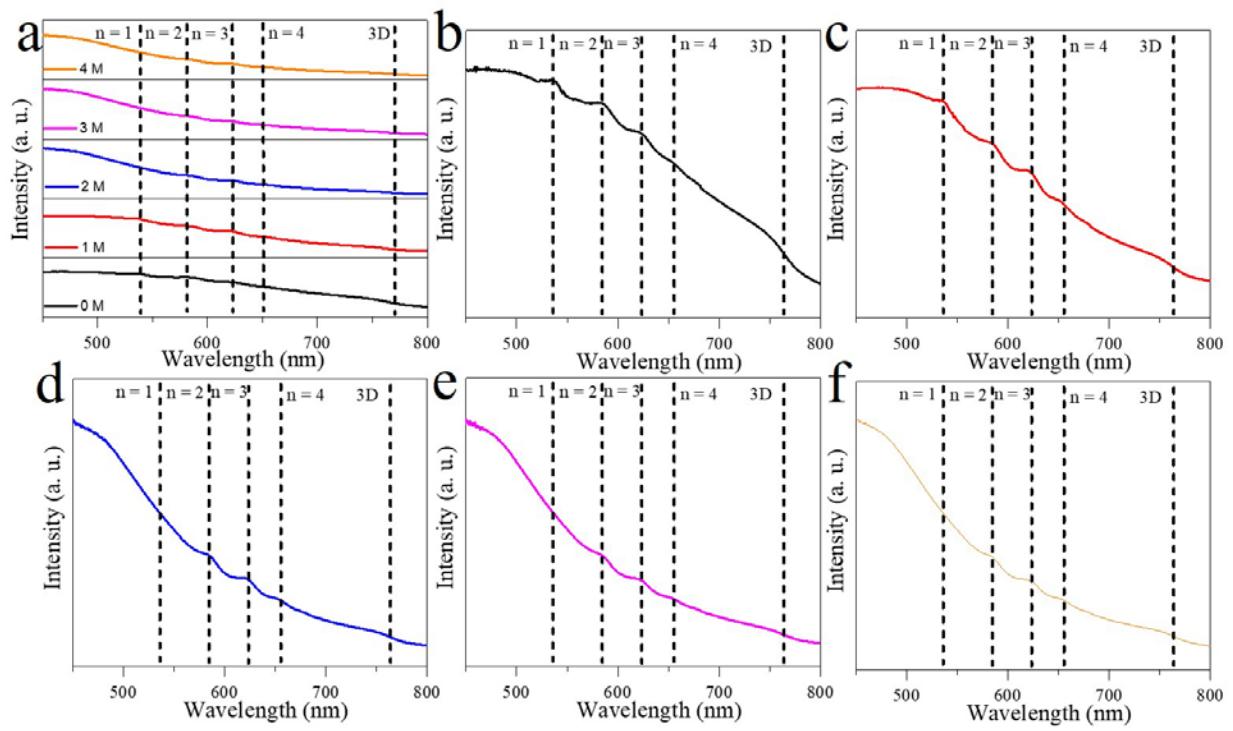
<sup>a</sup>Centre of Super-Diamond and Advanced Films (COSDAF) and Department of Chemistry, City University of Hong Kong, Hong Kong SAR, P. R. China

<sup>b</sup>City University of Hong Kong Shenzhen Research Institute, Shenzhen 518057, Guangdong, P.R. China

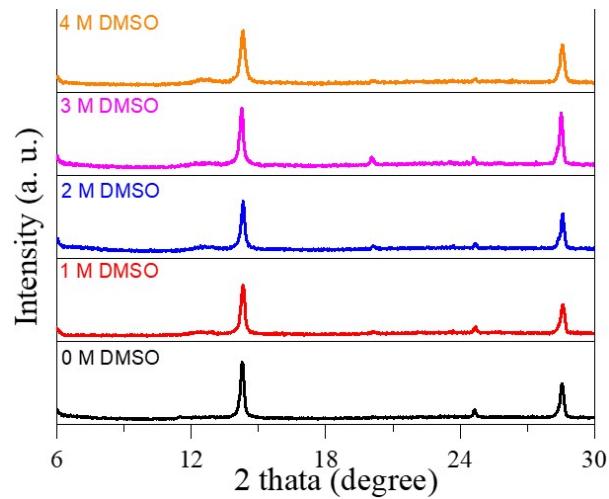
E-mail: mingflo@cityu.edu.hk and apcslee@cityu.edu.hk



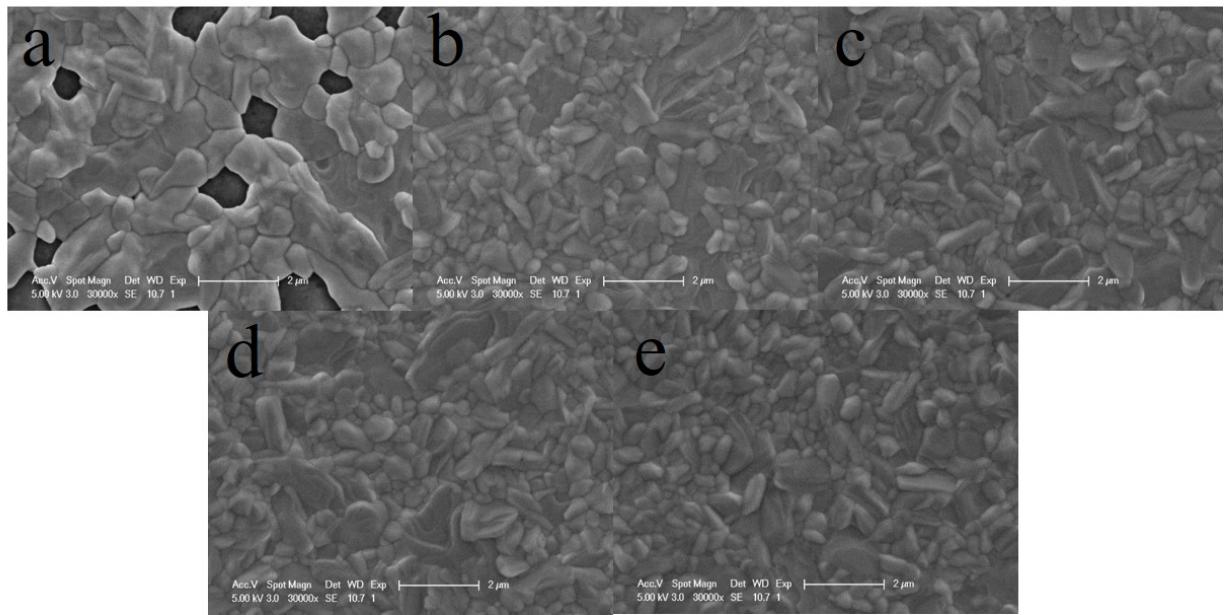
**Figure S1.** Magnified absorption spectra of (a) 0, (b) 1, (c) 2, (d) 3, (e) 4 M DMSO samples measured immediately after spin coating (i.e. no annealing).



**Figure S2.** (a) Absorption spectra of the 0 – 4 M DMSO samples measured after annealing, (b) 0, (c) 1, (d) 2, (e) 3, (f) 4 M DMSO samples.



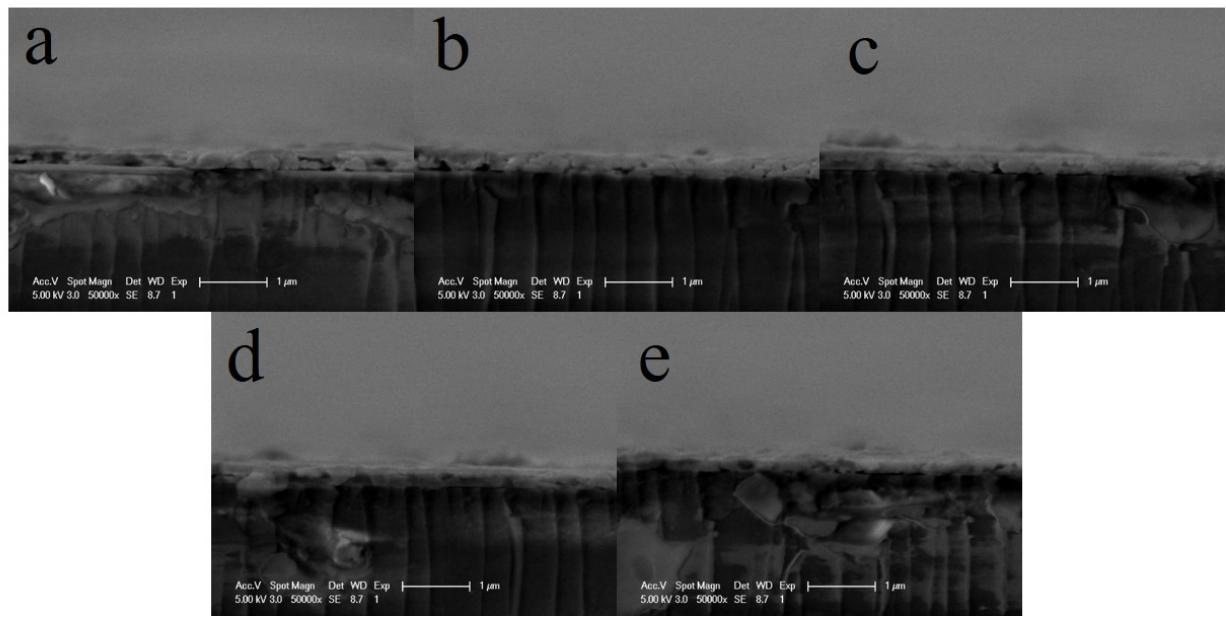
**Figure S3.** XRD patterns of the 0 – 4 M DMSO samples measured after annealing.



**Figure S4.** Top-view SEM images of the 0 – 4 M DMSO samples.

**Table S1.** Summary of the parameters from fits to the TRPL measurement. The fitting functions of treble exponential equation.  $y = y_0 + A_1 \exp(-\frac{t}{\tau_1}) + A_2 \exp(-\frac{t}{\tau_2}) + A_3 \exp(-\frac{t}{\tau_3})$

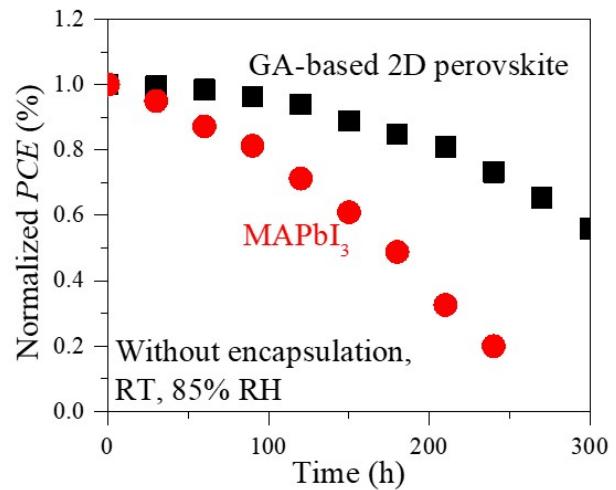
DMSO concentration (M)	A <sub>1</sub>	τ <sub>1</sub> (ns)	A <sub>2</sub>	τ <sub>2</sub> (ns)	A <sub>3</sub>	τ <sub>3</sub> (ns)
0	5434	0.31	102.7	2.55	6.56	66.7
1	4826	0.55	145.2	4.41	7.47	78.9
2	4031	1.01	286.3	5.86	21.1	124.44
3	3021	1.56	431.2	7.86	26.7	185.34
4	4548	0.7322	301.839	4.23	18.8	88.7



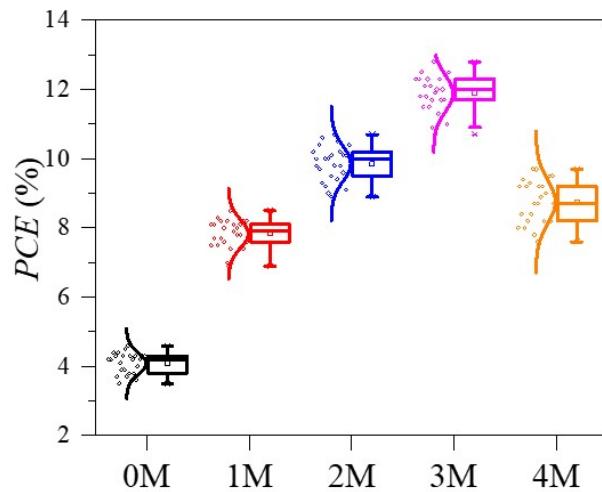
**Figure S5.** Cross-section SEM images of the (a) 0, (b) 1, (c) 2, (d) 3, (e) 4 M DMSO samples.

**Table S2.** Comparison of efficiency between this work and other 2D perovskite solar cells.

Reference	Large organic cations used	ACI perovskite	Efficiency (%)
1	PEA	No	4.7
2	BA	No	12.5
3	PEA	No	12.0
4	BA	No	13.7
5	PDA	No	13.0
6	BA	No	7.9
7	BA	No	12.8
8	i-BA	No	10.6
9	BA	No	8.2
10	BA	No	12.2
11	GA	Yes	7.3
Our work	GA	Yes	12.8



**Figure S6.** Stabilities of the PSCs based on ACI and 3D perovskites.



**Figure S7.** Statistics of *PCE* distribution of the ACI-based 2D PSCs with the 0 – 4 M DMSO samples.

## Reference

1. I. C. Smith, E. T. Hoke, D. Solis-Ibarra, M. D. McGehee and H. I. Karunadasa, *Angew. Chem. Int. Ed.*, 2014, **53**, 11232-11235.
2. H. Tsai, W. Nie, J. C. Blancon, C. C. Stoumpos, R. Asadpour, B. Harutyunyan, A. J. Neukirch, R. Verduzco, J. J. Crochet, S. Tretiak, L. Pedesseau, J. Even, M. A. Alam, G. Gupta, J. Lou, P. M. Ajayan, M. J. Bedzyk and M. G. Kanatzidis, *Nature*, 2016, **536**, 312-316.
3. J. Qing, X.-K. Liu, M. Li, F. Liu, Z. Yuan, E. Tiukalova, Z. Yan, M. Duchamp, S. Chen, Y. Wang, S. Bai, J.-M. Liu, H. J. Snaith, C.-S. Lee, T. C. Sum and F. Gao, *Adv. Energy Mater.*, 2018, **8**, 1800185.
4. X. Zhang, X. Ren, B. Liu, R. Munir, X. Zhu, D. Yang, J. Li, Y. Liu, D.-M. Smilgies, R. Li, Z. Yang, T. Niu, X. Wang, A. Amassian, K. Zhao and S. Liu, *Energy Environ. Sci.*, 2017, **10**, 2095-2102.
5. C. Ma, D. Shen, T. W. Ng, M. F. Lo and C. S. Lee, *Adv. Mater.*, 2018, **30**, 1800710.
6. A. Z. Chen, M. Shiu, J. H. Ma, M. R. Alpert, D. Zhang, B. J. Foley, D. M. Smilgies, S. H. Lee and J. J. Choi, *Nature communications*, 2018, **9**, 1336.
7. N. Zhou, Y. Shen, L. Li, S. Tan, N. Liu, G. Zheng, Q. Chen and H. Zhou, *J. Am. Chem. Soc.*, 2018, **140**, 459-465.
8. Y. Chen, Y. Sun, J. Peng, W. Zhang, X. Su, K. Zheng, T. Pullerits and Z. Liang, *Adv. Energy Mater.*, 2017, **7**, 1700162.
9. H. Li, J. Lu, T. Zhang, Y. Shen and M. Wang, *ACS Energy Lett.*, 2018, **3**, 1815-1823.
10. X. Zhang, R. Munir, Z. Xu, Y. Liu, H. Tsai, W. Nie, J. Li, T. Niu, D. M. Smilgies, M. G. Kanatzidis, A. D. Mohite, K. Zhao, A. Amassian and S. F. Liu, *Adv. Mater.*, 2018, **30**, 1707166.
11. C. M. M. Soe, C. C. Stoumpos, M. Kepenekian, B. Traore, H. Tsai, W. Nie, B. Wang, C. Katan, R. Seshadri, A. D. Mohite, J. Even, T. J. Marks and M. G. Kanatzidis, *J. Am. Chem. Soc.*, 2017, **139**, 16297-16309.