## **Supporting Information**

## Enhanced and Stabilized Photoluminescence of Perovskite Cesium Lead Bromide Nanocubes through Ordered Assembly

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Fig. S1. Schematic illustrations of preparation methods for various arrays of CsPbBr<sub>3</sub> NCs using organic medium. The slow evaporation rate of the dispersion medium provides sufficient time for self-assembly of NCs by capillary forces at the air– liquid interface. Thus, we obtained large 2D arrays using the evaporation-driven assembly method.<sup>1</sup> On the other hand, the domain size of 2D arrays produced by the conventional spin coating is relatively small because the dispersion medium is quickly evaporated from the surface of the substrate. The ordered arrays are not formed by the evaporation of diluted dispersion. We obtained isolated NCs through the evaporation from a droplet of diluted dispersion. A collodion film was used to prevent aggregation of NCs through surface migration.



Fig. S2. Typical procedure for the PL measurement of NCs on a substrate after coverage with silica plates and resin.



Fig. S3. TEM image (a), SAED pattern (b), size distribution (c, d), absorption and PL spectra (e), and PLQY (f) of CsPbBr<sub>3</sub> NCs before (a-c) and after (d) the ligand exchange.



Fig. S4. FT-IR spectra of CsPbBr<sub>3</sub> NCs before ligand exchange (with OA and OLA) and after ligand exchange (with DDAB). In the FT-IR spectra, the disappearance of the =C-H stretching mode at 3075cm<sup>-1</sup> of olefine indicates desorption of OA and OLA from the NC surface after the ligand exchange.<sup>2,3</sup> On the other hand, the presence of CH<sub>2</sub> and CH<sub>3</sub> symmetric and asymmetric stretching vibrations in the range of 2840-2950 cm<sup>-1</sup>, and CH<sub>2</sub> bending vibration at 1458 cm<sup>-1</sup> suggests the absorption of DDAB. We added the following sentences to show the presence of DDAB on the NC surface.



Fig. S5. Schematic illustration of ordered 2D arrays of NCs covered with ligands on the CsPbBr<sub>3</sub> surface. The Br<sup>-</sup> and Cs<sup>+</sup> vacancies of the CsPbBr<sub>3</sub> surface are occupied with DDA<sup>+</sup> and Br<sup>-</sup>, respectively, which are formed with dissociation of DDAB. This illustration was prepared according to a previous work.<sup>4</sup>



Fig. S6. UV-visible absorption spectra and Tauc plots of CsPbBr<sub>3</sub> NCs in various states.



Fig. S7. Time courses of the PL intensity and the PLQY of CsPbBr<sub>3</sub> NCs in another set of 2D arrays and isolated NCs under illumination at 468 nm.



Fig. S8. Schematic illustrations for the structural difference of the large and small 2D arrays and isolated NCs. We compared the ratio of the total area attaching to other NCs to the total surface area of NCs. All the faces of the isolated NCs are not attached to other NCs. In the small and large 2D arrays, about 90% and 99% of faces of NCs are attached to other NCs respectively. The higher ratio of the total surface area contacting to other NCs improves the PLQY and the photostability, because the surface ligands between two faces are not easily desorbed.

## References

H. Imai, R. Matsumoto, M. Takasaki, K. Tsukiyama, K. Sawano and Y. Nakagawa, *CrystEngComm*, 2019, 21, 6905–6914.
J. H. Park, A. Lee, J. C. Yu, Y. S. Nam, Y. Choi, J. Park and M. H. Song, *ACS Appl. Mater. Interfaces*, 2019, 11, 8428–8435.
J. Pan, L. N. Quan, Y. Zhao, W. Peng, B. Murali, S. P. Sarmah, M. Yuan, L. Sinatra, N. M. Alyami, J. Liu, E. Yassitepe, Z. Yang, O. Voznyy, R. Comin, M. N. Hedhili, O. F. Mohammed, Z. H. Lu, D. H. Kim, E. H. Sargent and O. M. Bakr, *Adv. Mater.*, 2016, 28, 8718–8725.

4 M. Imran, P. Ijaz, L. Goldoni, D. Maggioni, U. Petralanda, M. Prato, G. Almeida, I. Infante and L. Manna, ACS Energy Lett., 2019, 4, 819–824