Supporting Information

Surface-assisted synthesis of perovskite nanosheets with bivalent aromatic cation

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The preparation of reference sample.

The single crystals used as references for GIWAXS and SAED measurements were prepared according to previously published procedure.¹

Briefly, a 57% w/w aqueous HI solution was diluted to 47% w/w. A 0.3 M solution of PbI₂ in freshly prepared HI (47%) was kept at 120 °C. At the same time, a 0.03 M solution of 1,5-naphthaleneammonium iodide was prepared with stirring at 130 °C. Then 224 mL of amine precursor solution was rapidly added to the 448 μ L of PbI₂ solution. Single crystals were obtained by slowly cooling the mixture to room temperature. The crystals were isolated, gently washed with Et₂O, and dried under reduced pressure.



Figure S1. The schematic layered structure of (1,5-DAN)PbI₄ perovskite.



Figure S2. SEM images of perovskite nanosheets on Si wafer grown at room temperature (a) and at 50 °C (b) (Note: Fig. S1 is identical to Fig. 2d).



Figure S3. SEM images of perovskite nanosheets on Si wafer grown at room temperature from solutions of different precursor concentartions (a) 2.5 mM, (b) 5 mM, and (c) 10 mM.



Figure S4. Single crystal X-ray structure of bulk (1,5-DAN)PbI₄ (view along a axis). The Pb-I-Pb bond angle is 150.67°, which indicates a strong distortion. For clarity, all hydrogen atoms were removed. Violet balls, iodine atoms; dark gray balls, lead atoms; light grey balls, carbon atoms; blue balls, nitrogen atoms.



Figure S5. (a) STEM image of a perovskite particle grown directly on a TEM grid. (b) EDX spectrum of the area marked with red square in (a).



Figure S6. (A)-(C) represent the topography images recorded with the KPFM images of the particle prior to the illumination (A), during 405 nm illumination (B), and after the illumination (C). D-F represent the corresponding KPFM images: prior to the illumination (D), during 405 nm illumination (E), and after the illumination (F).



Figure S7. (A)-(C) represent the topography image recorded with the KPFM images prior to the illumination (A), during 458 nm illumination (B), and during 405 nm illumination (C). (D)-(F) represent the KPFM images prior to the illumination (D), during 458 nm illumination (E), and during 405 nm illumination (F).

References

(1) A. Mitrofanov, Y. Berencén, E. Sadrollahi, R. Boldt, D. Bodesheim, H. Weiske, F. Paulus, J. Geck, G. Cuniberti, A. Kuc and B. Voit, *J. Mater. Chem. C*, 2023, **11**, 5024–5031.