Supporting information for

Two-Dimensional Lead Halide Perovskites Templated by a Conjugated Asymmetric Diammonium

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Equations used to calculate Quadratic Elongation ($\langle\lambda\rangle$) and Bond angle variance (σ^2).[S1, S2]

Equation S1: Quadratic Elongation ($\langle\lambda\rangle$)

$$\langle\lambda\rangle = \frac{1}{6}\sum_{\{i=1\}}^{6}\left(\frac{d_i}{d_o}\right)^2$$

Where d_i is the Pb-X bond length, d_o is the Pb-X bond length from an ideal octahedron of the same volume, $\langle\lambda\rangle$ is dimensionless.

Equations S2: Bond angle variance (σ^2)

$$\sigma^2 = \frac{1}{11}\sum_{\{i=1\}}^{12}(\alpha_i - 90)^2$$

Where α_i is the Pb-X-Pb bond angles of the octahedra.

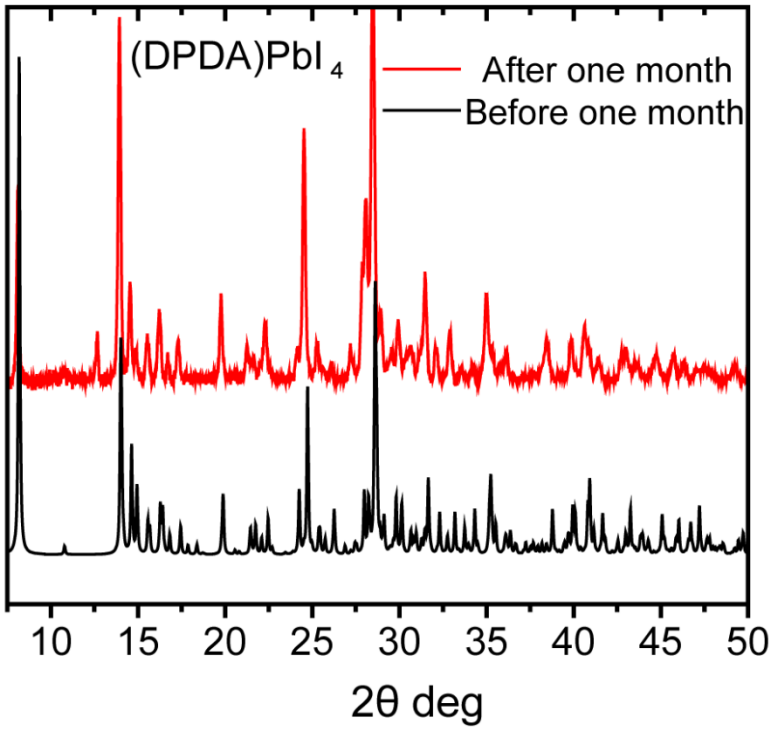

Figure S1. PXRD patterns of (DPDA)PbI_4 powders (crushed from single crystals) before and after one month of storage in ambient conditions.

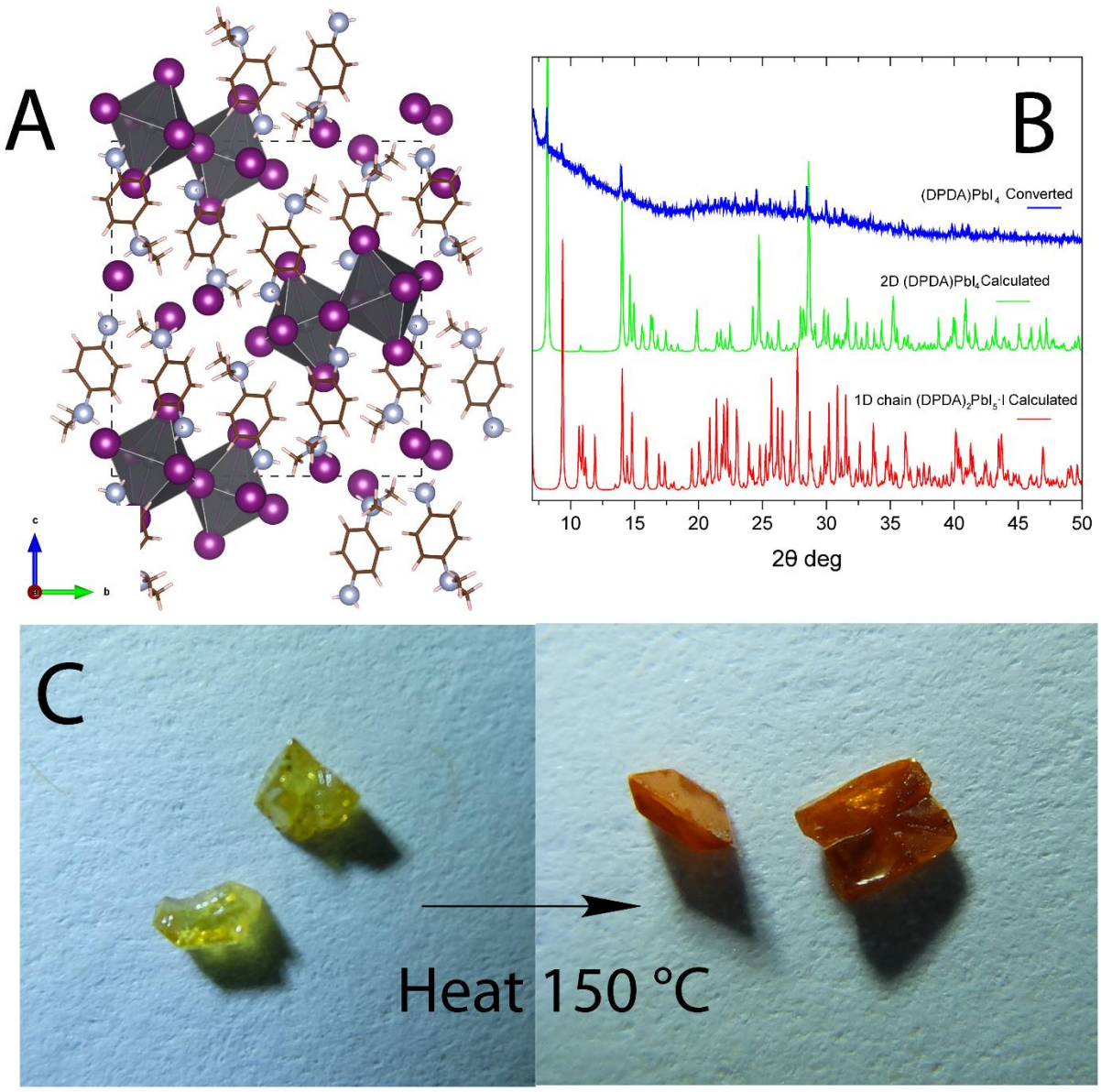

Figure S2. (A) Crystal structure of $(DPDA)_2PbI_5 \cdot I$ showing 1D corner sharing chains. Upon heating, these crystals turned from yellow to red (C) and showed peaks of the 2DN perovskite in the PXRD pattern (B) indicating partial degradation/conversion to the 2DN layered compound.

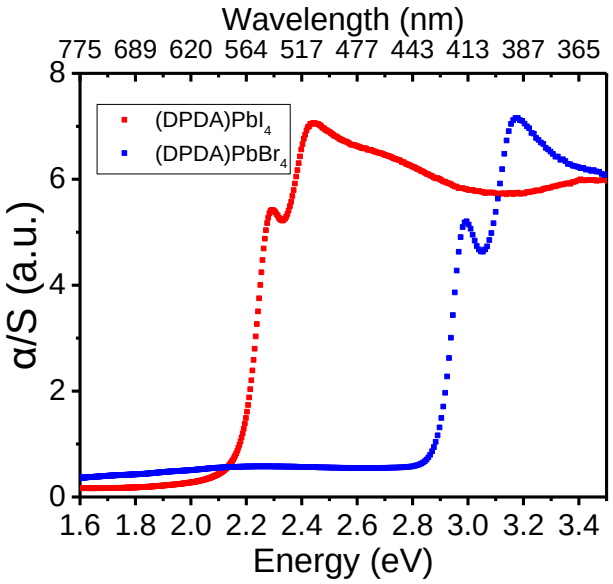

Figure S3. Reflectance spectroscopy of bulk powders of (DPDA)PbI_4 and (DPDA)$PbBr_4$.

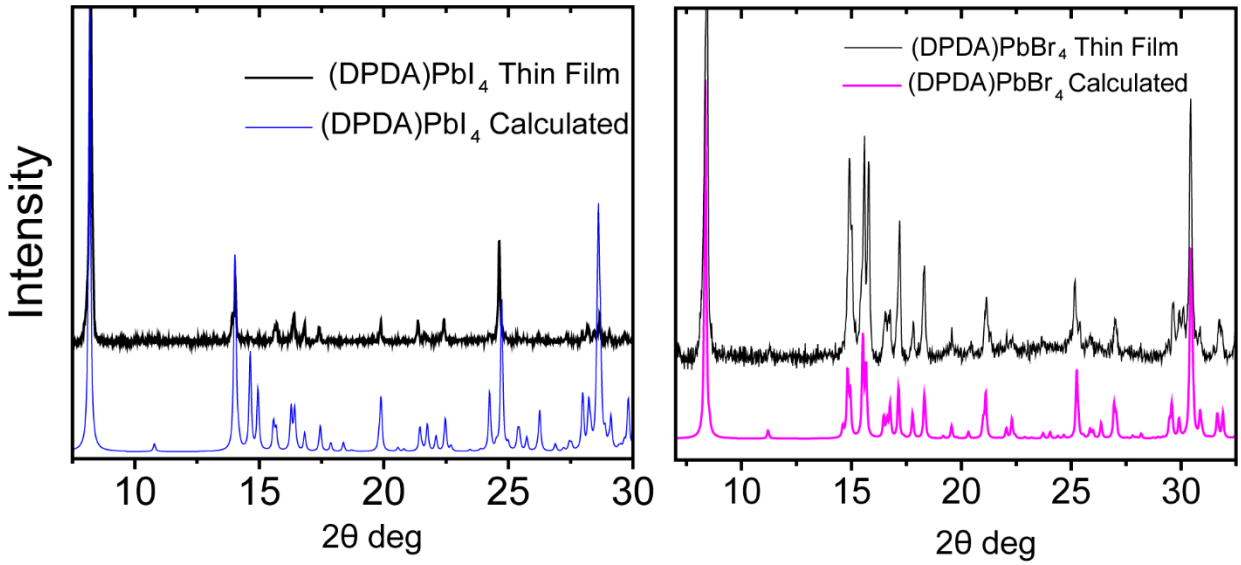

Figure S4. PXRD patterns of the spin coated thin films of (A) (DPDA)PbI_4 and (B) (DPDA)$PbBr_4$ in comparison with their respective calculated patterns from single crystal structures.

References

S1. Robinson, K.; Gibbs, G. V.; Ribbe, P. H., Quadratic elongation: a quantitative measure of distortion in coordination polyhedra. *Science* **1971,** *172*, 567.

S2. Brese, N. E.; O'Keeffe, M., Bond-valence parameters for solids. *Acta Crystallogr., Sect. B* **1991,** *47*, 192-197.