Supplementary Materials for

Orientation-Driven Chirality Funnels in Chiral Low-Dimensional Lead-Halide Perovskite Heterostructures

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Singe Crystal X-ray Crystallography

Data for (R)-EAPbI₃ were collected on a Bruker D8 Venture single crystal x-ray diffractometer equipped with a CPAD detector (Bruker Photon II), an IMS microsource with Mo- K_{α} radiation ($\lambda = 0.71073$ Å) and a Helios optic using the APEX3 software package.¹ Data for (S)-EAPbI₃ were collected on an Agilent Technologies Supernova-E CCD diffractometer (Mo- K_{α} radiation, microfocus X-ray tube, multilayer mirror optics) and integrated using CrysAlisPro.² Measurements were performed on single crystals coated with perfluorinated ether. The crystals were fixed on top of a kapton micro sampler (for (R)-EAPbI₃) or on a MiTeGen microloop (for (S)-EAPbI₃) and frozen under a stream of cold nitrogen. A matrix scan was used to determine the initial lattice parameters. Reflections were corrected for Lorentz and polarisation effects, scan speed, and background using SAINT.³ Absorption correction, including odd and even ordered spherical harmonics was performed using SADABS.³ Space group assignments were based upon systematic absences, E statistics, and successful refinement of the structures. The structures were solved using SHELXT⁴ with the aid of successive difference Fourier maps, and were refined against all data using SHELXL⁵ in conjunction with SHELXLE⁶ (for (R)-EAPbI₃) or with OLEX2⁷ (for (S)-EAPbI₃). Hydrogen atoms were calculated in ideal positions as follows: Methyl hydrogen atoms were refined as part of rigid rotating groups, with a C-H distance of 0.98 Å and $U_{iso}(H) = 1.5 \cdot U_{eq}(C)$. Non-methyl H atoms were placed in calculated positions and refined using a riding model with methylene, aromatic, and other C-H distances of 0.99 Å, 0.95 Å, and 1.00 Å, respectively, and $U_{iso}(H) = 1.2 \cdot U_{eq}(C)$. Non-hydrogen atoms were refined with anisotropic displacement parameters. Full-matrix least-squares refinements were carried out by minimizing $\Sigma w(F_o^2 - F_c^2)^2$ with the SHELXL weighting scheme.⁵ Neutral atom scattering factors for all atoms and anomalous dispersion corrections for the non-hydrogen atoms were taken from International Tables for Crystallography.⁸ CCDC 2402411 and 2402238 contain the supplementary crystallographic data for this paper. These data are provided free of charge by the joint Cambridge Crystallographic Data Centre and Fachinformationszentrum Karlsruhe Access Structures service www.ccdc.cam.ac.uk/structures.

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	(R)-EAPbI ₃	(S)-EAPbI ₃	
Empirical Formula	C9H14I3NPb	C9H14I3NPb	
Temperature, K	100 K	120 K	
Formula Weight, g/mol	724.10	724.10	
Crystal System	Orthorhombic	Orthorhombic	

Table S1. Crystallographic data for (R)-EAPbI₃.

Space group	$P 2_1 2_1 2_1$	$P 2_1 2_1 2_1$
a, Å	8.1004(4)	8.1025(2)
b, Å	8.7167(5)	8.7237(2)
c, Å	21.6184(11)	21.6253(4)
α, °	90	90
β, °	90	90
γ, °	90	90
Volume, Å ³	1526.45(14)	1528.37(6)
Z	4	4
Density (calculated), g/cm ³	3.151	3.147
Absorption coefficient, mm ⁻¹	17.091	17.070
F(000)	1264	1264
Radiation	$MoK_{\alpha} (\lambda = 0.71073)$	$MoK_{\alpha} (\lambda = 0.71073)$
2θ range for data collection, °	2.25 to 28.75	5.036 to 68.28
Reflections collected	70309	38666
Independent reflections	3875 [R(int) = 0.0427]	6104 [R(int) = 0.0896]
Data/restraints/parameters	3875/3/129	6104/3/141
Goodness of fit on F ²	1.153	1.019
Final R indexes [I>2 σ (I)]	$R_1 = 0.0160, wR_2 = 0.0366$	$R_1 = 0.0306, wR_2 = 0.0528$
Final R indexes (all data)	$R_1 = 0.0161, wR_2 = 0.0367$	$R_1 = 0.0368, wR_2 = 0.0558$
CCDC number	2402411	2402238



Fig. S1. Photograph of (R)-EAPbI₃ single crystals on millimeter-grid paper.

	Dh I Dondlonath	Bond length	Bond angle	
	ro-i bolid leligui	distortion index	variance degrees ²	
(R)-EAPbI3	3.151		-	
	3.081		68.08	
	3.220	0.007		
	3.302	0.027		
	3.232			
	3.431			
(S)-EAPbI3	3.149		67.82	
	3.302			
	3.220	0.027		
	3.232	0.027 07.02		
	3.430			
	3.082			

Table S2. Octahedral distortion calculation for (R)-EAPbI₃ and (S)- EAPbI₃.

The bond length distortion index (D) and bond angle variance degrees (σ^2) are calculated by the following equations:

$$D = \frac{1}{6} \sum_{i=1}^{6} \frac{|d_i - d_0|}{d_0}$$
$$\sigma^2 = \frac{1}{11} \sum_{i=1}^{12} (\theta_i - 90)^2,$$

where d_i is the corresponding Pb-I bond distance, d_0 represents the average Pb-I bond length, and θ_i means I-Pb-I bond angle in octahedral structures. All these values can be obtained from single crystal XRD data.



Fig. S2. In-situ XRD maps obtained while monitoring the formation of three different perovskite thin films produced using DMF, mixed DMSO-DMF, and DMSO solvents. The XRD patterns cover a broad region from 9° to 30°.



Fig. S3. Comparison of XRD patterns between films prepared with different solvents. All the films were annealed at 100 °C for 10 mins.



Fig. S4. 2D GIWAXS data of chiral (R)-EAPbI₃ perovskites films fabricated using DMF solvent without thermal annealing (a), as well as films produced using mixed DMSO-DMF (b), DMF (c), and DMSO solvents (d) followed by annealing at 100 °C for 10 min.



Fig. S5. AFM images of chiral (R)-EAPbI₃ perovskite films. The data were collected from perovskite films fabricated using DMF solvent without thermal annealing (a), as well as films produced using mixed DMSO-DMF (b), DMF (c), and DMSO solvents (d) followed by annealing at 100 °C for 10 min.



Fig. S6. **a**, Anisotropy factor (g_{abs}) of chiral (R)-EAPbI₃ perovskite films. The perovskite films were prepared using three different solvent solutions, as indicated in the legend. **b**, CD spectra collected from chiral (R), (S), and Racemic-EAPbI₃ perovskite thin films (DMSO-DMF).

The absorption anisotropy factors (g_{abs}) are calculated by following equations:

$$g_{abs} = \frac{2 \times (Abs_{\sigma^-} - Abs_{\sigma^+})}{Abs_{\sigma^-} + Abs_{\sigma^+}}$$

where Abs_{σ^+} and Abs_{σ^-} are the absorption intensities of right-handed and left-handed circularly polarized light, respectively.



Fig. S7. Room temperature PL spectra of chiral (R)-EAPbI₃ perovskite films. The PL measurements were performed under 340 nm excitation and were collected from films produced using three different solvents, as indicated in the legend.



Fig. S8. Photographs of (R)-EAPbI₃ and PbI₂ spin-coated thin films.



Fig. S9. **a**, **b**, Steady-state absorption spectra (a) and XRD patterns (b), collected from (R)-EAPbI₃, pure PbI₂, and (R)-EAPbI₃ films with excess PbI₂ adding into spin-coating solution. (R)-EAPbI₃ spin-coating films were prepared by dissolving 40 mg (R)-EAPbI₃ single crystals in 200 μ L DMSO-DMF solvent. Pure PbI₂ films were prepared by dissolving 40 mg PbI₂ powder in 200 μ L DMSO-DMF solvent. (R)-EAPbI₃ + PbI₂ films were prepared by dissolving 40 mg (R)-EAPbI₃ single crystals and 20 mg PbI₂ powder in 200 μ L DMSO-DMF solvent.



Fig. S10. Comparison of steady-state absorption spectra, collected from (R)-EAPbI₃, (R)-EAPbI₃ films with excess (R)-EAI adding into spin-coating solution. (R)-EAPbI₃ spin-coating films were prepared by dissolving 40 mg (R)-EAPbI₃ single crystals in 200 μ L DMSO-DMF solvent. (R)-EAPbI₃ + (R)-EA films were prepared by dissolving 40 mg (R)-EAPbI₃ single crystals and 20 or 40 mg (R)-EAI powder in 200 μ L DMSO-DMF solvent.



Fig. S11. TA color maps of pure PbI₂ films using DMSO-DMF mixture solvent. The excitation wavelength is 340 nm with a fluence of $\sim 212 \mu J/cm^2$.



Fig. S12. CTA kinetics collected from GSB1 regions in Fig. 4a-c.



Fig. S13. TA color maps of chiral (R)-EAPbI₃ perovskite samples using DMF (a), DMSO-DMF mixture (b), and DMSO solvents (c). The excitation wavelength is 430 nm to avoid the excitation of GSB1 signals (~378 nm).



Fig. S14. Low-temperature TA measurements on chiral (R)-EAPbI₃ perovskite films. TA color map (a) and corresponding spectra (b) obtained from chiral perovskite films (DMSO-DMF mixture) at 5 K.



Fig. S15. Low-temperature CTA kinetics obtained from GSB2 regions in Fig. 5h.

References

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