

Supporting Information

Chiral 0D hybrid lead-bromide perovskite with strong nonlinear chiroptical properties

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3. References

1. Experimental Section

Materials. *R*-3-Aminopiperidine dihydrochloride (*R*-APD·2HCl, Innochem, 98%), *S*-3-Aminopiperidine dihydrochloride (*S*-APD·2HCl, Innochem, 97%), 1*R*,2*R*-diaminocyclohexane (1*R*,2*R*-DACH, Innochem, 98%), 1*S*,2*S*-diaminocyclohexane (1*S*,2*S*-DACH, Innochem, 98%), Hydrobromic acid (HBr, ≥48% in H₂O, AR, KESHI), lead (II) acetate (Pb(OAc)₂·3H₂O, Innochem, 99.5%), lead (II) oxide (PbO, Alfa Aesar, 99.99%), lead (II) bromide (PbBr₂, Aladdin, 99.0%) all reagents and solvents were used without further purification.

Synthesis of (*R*-/*S*-APD)PbBr₄ single crystals. (*R*-/*S*-APD)PbBr₄ was prepared with stoichiometric quantities of (Pb(OAc)₂·3H₂O (1 mmol, 379 mg) and *R*-/*S*-APD·2HCl (1 mmol, 187.19 mg) in HBr solution (4 mL). After obtaining a clear solution by heating the mixture at 120 °C and stirring for 30 min. And the colorless platelike crystals of (*R*-/*S*-APD)PbBr₄ were obtained at room temperature overnight. The crystals were washed with cold diethyl ether.

Synthesis of (1*R*,2*R*-/1*S*,2*S*-DACH)₂PbBr₆·2H₂O single crystals. (1*R*,2*R*-/1*S*,2*S*-DACH)₂PbBr₆·2H₂O was prepared with an amount of PbO (2 mmol, 466 mg) powder dissolved in HBr solution (6.0 mL). After obtaining a clear solution by heating the mixture at 120 °C and stirring for 30 min. And then the 1*R*,2*R*-/1*S*,2*S*-DACH (2 mmol, 228 mg) was added to the solution. The needle-like white crystals of (1*R*,2*R*-/1*S*,2*S*-DACH)₂PbBr₆·2H₂O were obtained at room temperature overnight.

Synthesis of bulk single crystals of (*R*-/*S*-APD)PbBr₄ and (1*R*,2*R*-/1*S*,2*S*-DACH)₂PbBr₆·2H₂O. *R*-/*S*-APD·2HCl (4.15 mmol, 718 mg) and PbBr₂ (4.15 mmol, 1550 mg) were dissolved in 30.0 mL of HBr solution at 65 °C under constant stirring and formed a pale yellow transparent solution. The solution was filtered with a 0.2 μm Teflon syringe filter after cooling to room temperature. Block-shaped colorless (*R*-/*S*-APD)PbBr₄ single crystals were grown in an ambient atmosphere for about several weeks. The same method was used to prepare the bulk single crystals of (1*R*,2*R*-/1*S*,2*S*-

DACH)₂PbBr₆·2H₂O.

Characterization. Powder X-ray diffraction (PXRD) experiments were carried out using a Rigaku Ultima IV X-ray diffraction instrument in the 2θ between 5° and 50° with a step size of 0.02° under Cu K α radiation ($\lambda = 1.541871 \text{ \AA}$). Single crystal X-ray diffraction (SCXRD) measurements were performed using a XtaLAB Synergy R, DW system, HyPix diffractometer under Mo K α ($\lambda = 0.71073$). The single crystal structures were determined using the SHELXT methods with Olex2 program1. Thermogravimetric analyses (TGA) patterns were obtained using a Linseis STA PT1600 under N₂ atmosphere in the temperature range of 25 to 800 °C, with a heating rate of 10 °C/min. UV-vis absorption spectra were collected using a UV-vis spectrophotometer (UV 2600). Solid-state circular dichroism (CD) spectra were acquired using a JASCO J-1500 CD spectrometer. The perovskite sample was ground into powder. The samples for CD measurement were prepared though pressing the chiral perovskite powder and KBr powder (chiral perovskite: KBr = 1:100, w:w) into a pellet.

Second Harmonic Generation (SHG) Measurements. The SHG measurements of chiral perovskites were examined by using a home-built multiphoton nonlinear optical microscope system with a femtosecond pump (Mai Tai HP, 100 fs, 80 MHz, wavelength ranging from 690 to 1040 nm) in reflection geometry. The measurements were conducted under a reflection geometry at a 45° angle of both incidence and detection. The linearly polarized and circularly polarized pumps were altered with the $\lambda/2$ and $\lambda/4$ plates, respectively. For linear polarization-resolved SHG measurement, a $\lambda/2$ waveplate and an analyzer were respectively placed in the incident and output light path. For circular polarization-resolved SHG measurements, the $\lambda/2$ waveplate was replaced by a $\lambda/4$ waveplate and placed in the incident light path to modulate its polarization between LCP and RCP states. The power-dependent SHG measurements were completed by collecting the SHG signal intensity with increasing laser power upon laser excitation at specific wavelengths. When at a certain laser power, the SHG

intensity begins to deviate from the well-fitted quadratic relationship or drop. This power has been hence identified as the laser damage threshold (LDT) of the single crystal. The LDT was defined as $LDT = P/(F \times \pi r^2 t)$, where P , denoted incident power when the SHG signal is offset from the square dependence, F , r and t are frequency and radius of laser spot, pulse width, respectively. The laser spot was $\sim 20 \mu\text{m}$ in diameter. The pulse width was ~ 100 fs. The frequency was 80 MHz. The second-order NLO susceptibility of the chiral perovskites was determined by using Y-cut quartz with a d_{eff} of 0.3 pm/V as the reference.

Theoretical Calculations. The DFT calculations were performed using the program VASP (Vienna Ab Initio Simulation Package) 5.4.4, and the raw data was analyzed by vaspkit 1.4.1. We used the GGA-PBE function to describe the exchange correlation. A 400 eV kinetic energy cutoff and a $4 \times 3 \times 2$ for $(R-/S\text{-APD})\text{PbBr}_4$ / $3 \times 3 \times 1$ for $(1R,2R-/1S,2S\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$ gamma-centered K sampling grid were used for integration. The structures are firstly fully relaxed until the Hellmann–Feynmann force on each atom and the total energy difference between neighboring loops reach 10^{-6} eV and $0.02 \text{ eV} \cdot \text{Ang}^{-1}$ within 500 loops limitation. For the band structure, the K-Path was generated by vaspkit 1.4.1. Besides DOS, the PDOS (projected density of states) has been also calculated. PDOS is calculated as projected DOS, where the information about the different contributions of the different elements is computed.

2. Characterization

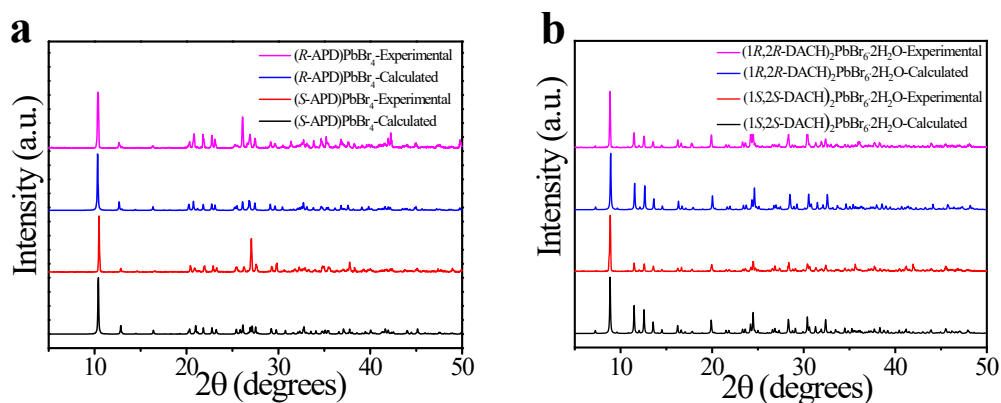


Figure S1. Experimental and calculated powder X-ray diffraction (PXRD) patterns of **a.** $(R-/S-APD)PbBr_4$ and **b.** $(1R,2R-/1S,2S-DACH)_2PbBr_6 \cdot 2H_2O$.

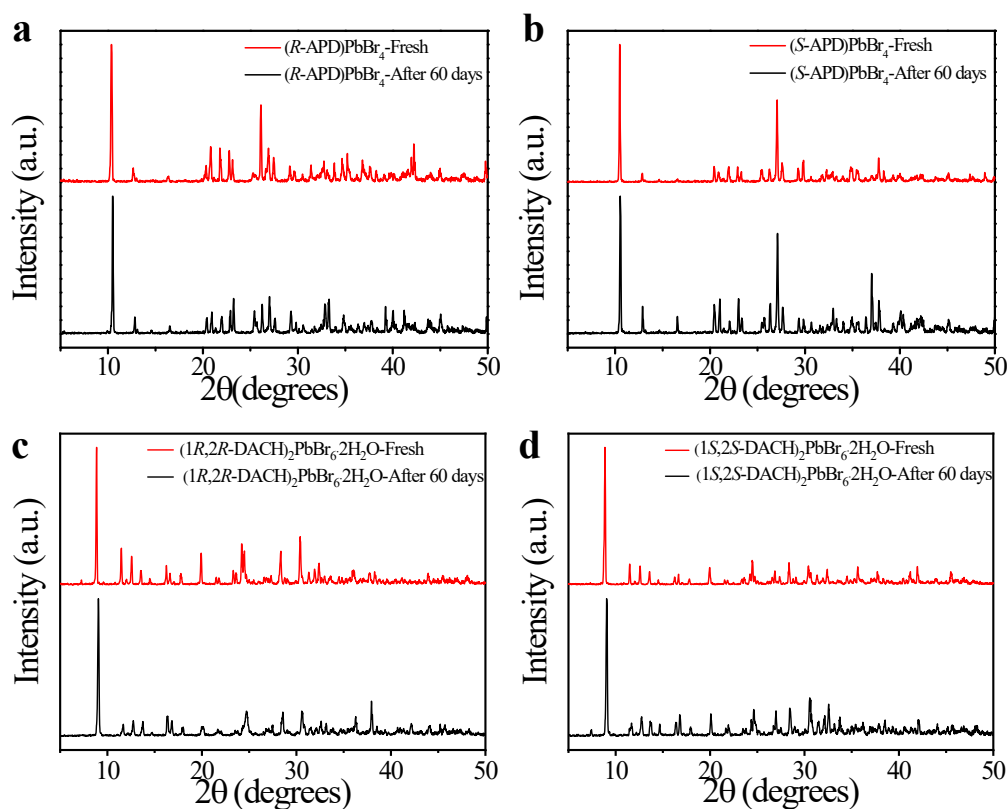


Figure S2. Powder X-ray diffraction (PXRD) patterns of the as-prepared and exposed in the air after 60 days. **a.** $(R-APD)PbBr_4$, **b.** $(S-APD)PbBr_4$, **c.** $(1R,2R-DACH)_2PbBr_6 \cdot 2H_2O$, **d.** $(1S,2S-DACH)_2PbBr_6 \cdot 2H_2O$.

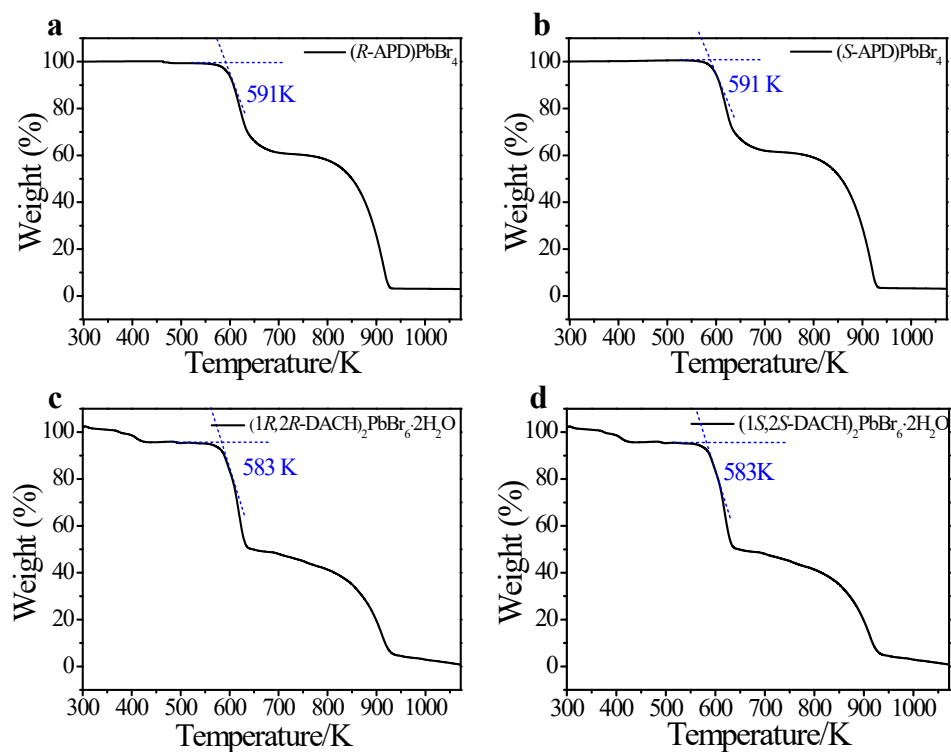


Figure S3. Thermal stability of **a.** $(R\text{-APD})\text{PbBr}_4$, **b.** $(S\text{-APD})\text{PbBr}_4$, **c.** $(1R,2R\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$ and **d.** $(1S,2S\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$, was measured by the thermogravimetric analysis (TGA) method.

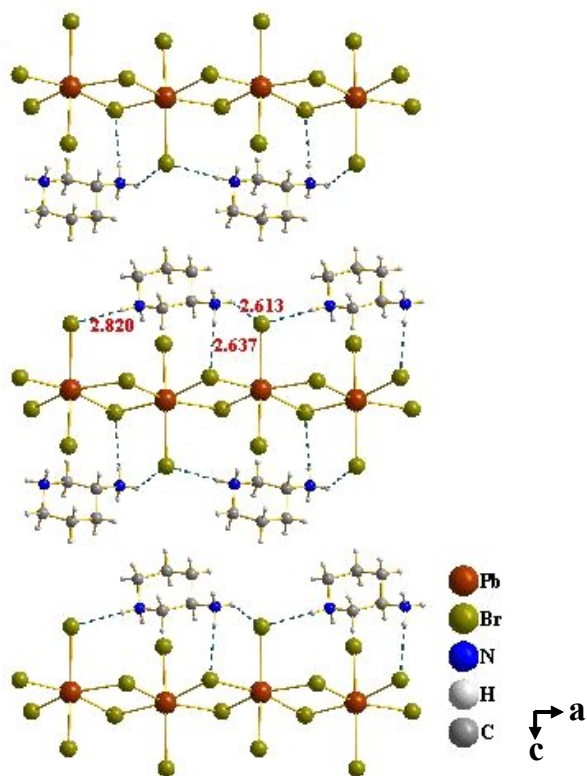


Figure S4. The hydrogen bonding $\text{N-H} \cdots \text{Br}$ of $(R\text{-APD})\text{PbBr}_4$, which are marked by the dotted blue lines.

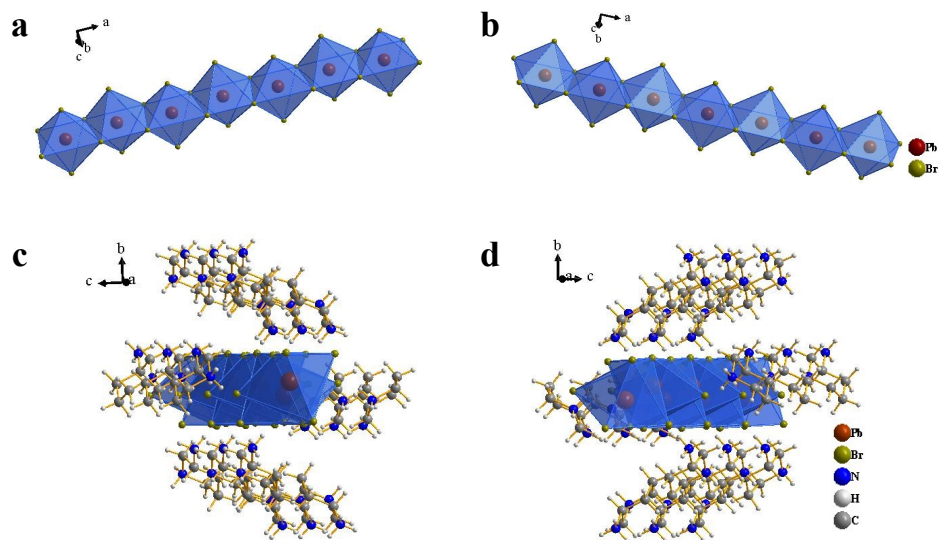


Figure S5. **a.** View of the individual inorganic chain based on edge-shared $[\text{PbBr}_6]^{4-}$ octahedrons of $(R\text{-APD})\text{PbBr}_4$. **b.** View of the individual inorganic chain based on edge-shared $[\text{PbBr}_6]^{4-}$ octahedrons of $(S\text{-APD})\text{PbBr}_4$. **c.** View of an individual one-dimensional inorganic chain wrapped by the organic cations of $(R\text{-APD})\text{PbBr}_4$. **d.** View of an individual one-dimensional inorganic chain wrapped by the organic cations of $(S\text{-APD})\text{PbBr}_4$.

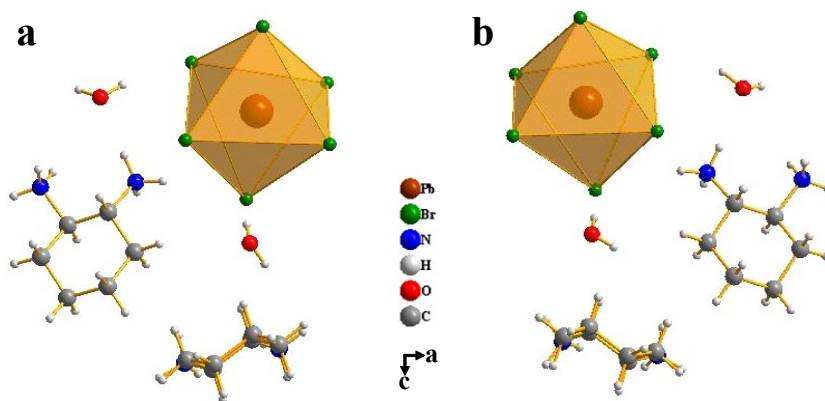


Figure S6. The asymmetry unit of **a.** $(1R,2R\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$ and **b.** $(1S,2S\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$ as viewed from the b -axis.

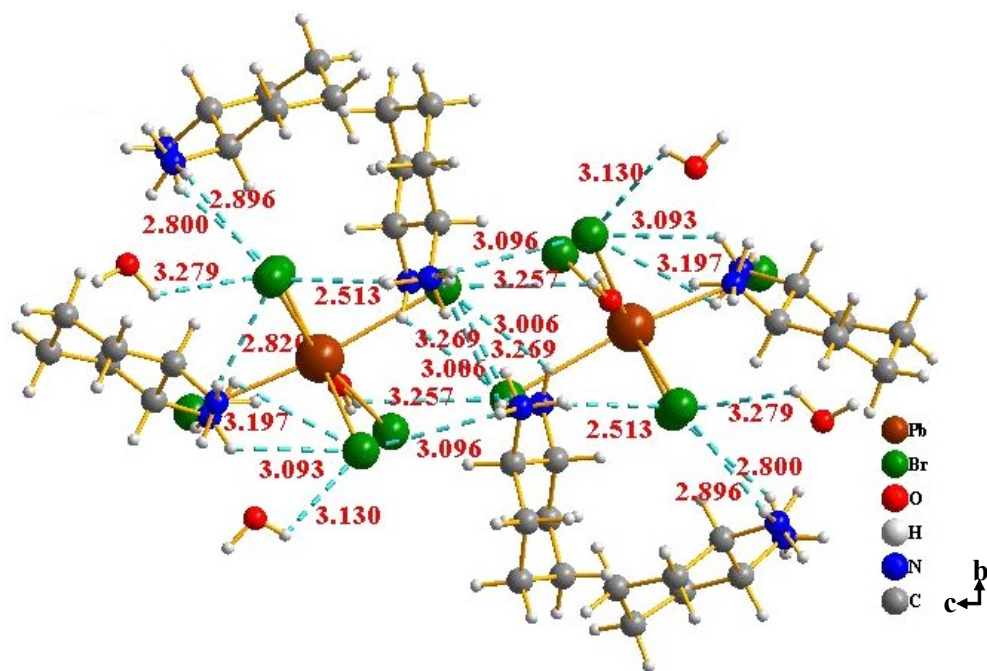


Figure S7. The hydrogen bonding N-H \cdots Br and O-H \cdots Br of (1*R*,2*R*-DACH)₂PbBr₆·2H₂O, which are marked by the dotted blue lines.

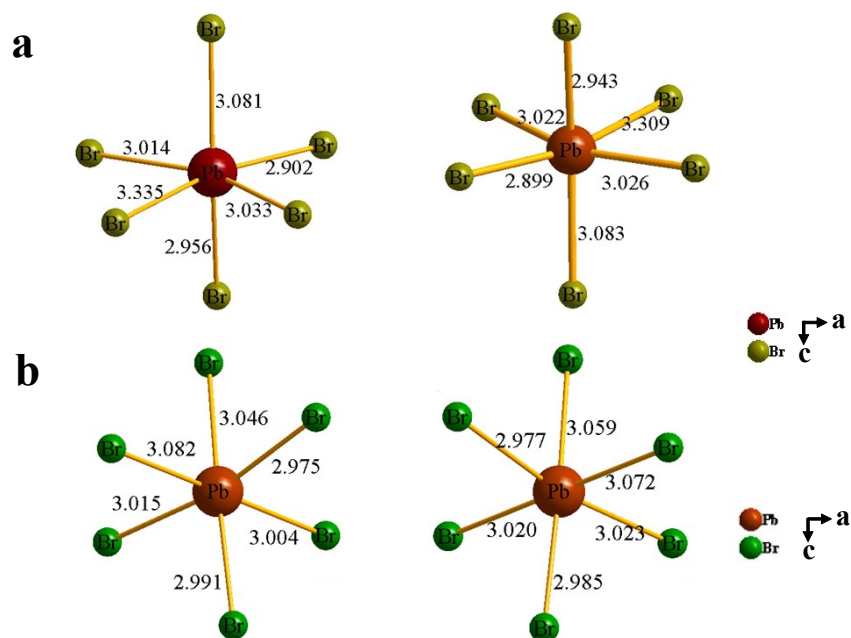


Figure S8. a. The distorted [PbBr₆]⁴⁻ structure of (*R*-APD)PbBr₄ (left) and (*S*-APD)PbBr₄ (right). **b.** The distorted [PbBr₆]⁴⁻ structure of (1*R*,2*R*-DACH)₂PbBr₆·2H₂O (left) and (1*S*,2*S*-DACH)₂PbBr₆·2H₂O (right).

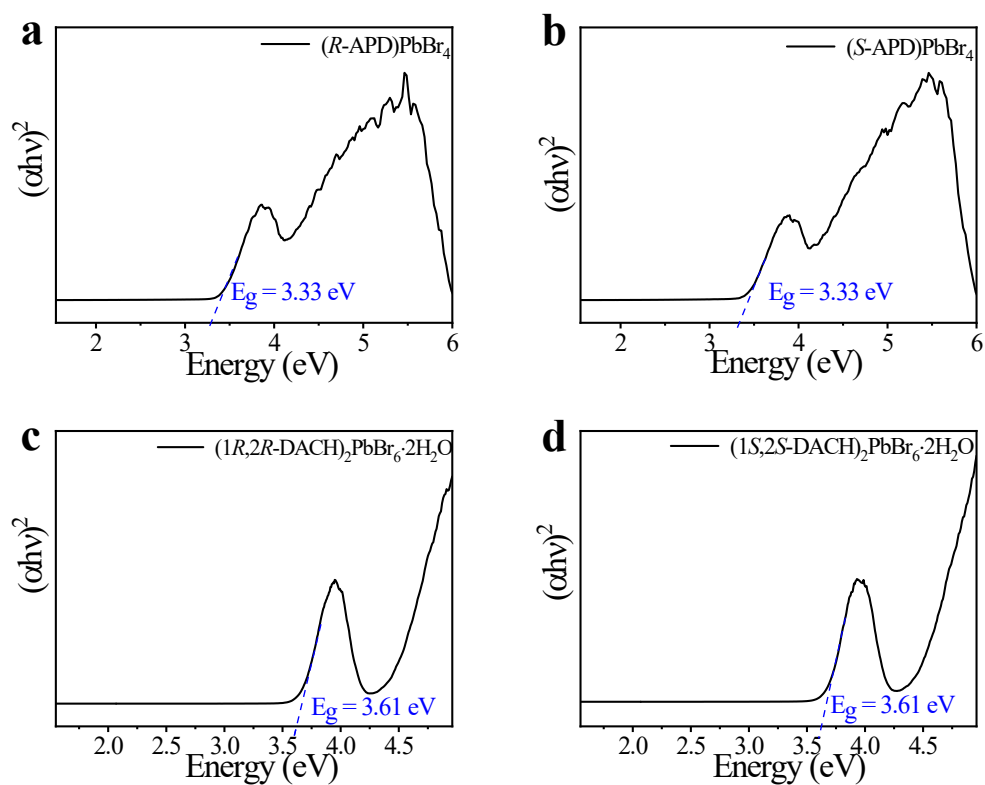


Figure S9. The corresponding Tauc plots of **a.** $(R\text{-APD})\text{PbBr}_4$, **b.** $(S\text{-APD})\text{PbBr}_4$, **c.** $(1R,2R\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$ and **d.** $(1S,2S\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$, respectively.

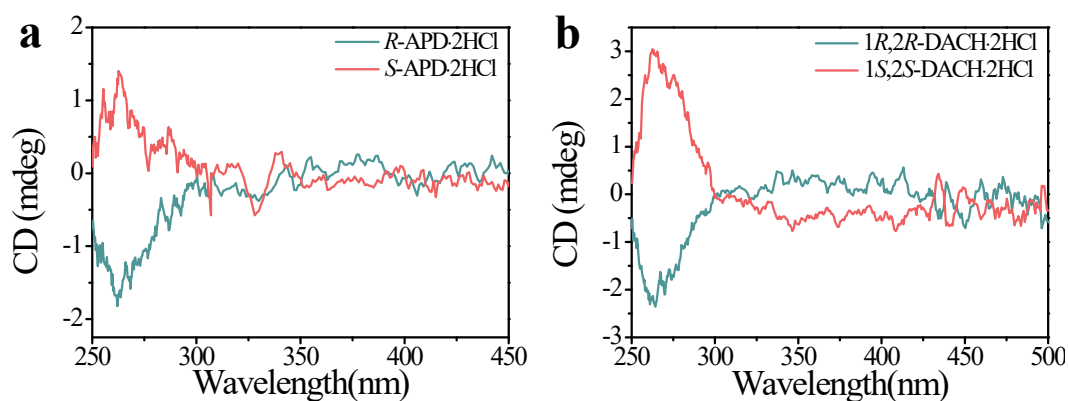


Figure S10. a. Circular dichroism spectra of $R\text{-APD} \cdot 2\text{HCl}$ and $S\text{-APD} \cdot 2\text{HCl}$. **b.** Circular dichroism spectra of $1R,2R\text{-DACH} \cdot 2\text{HCl}$ and $1S,2S\text{-DACH} \cdot 2\text{HCl}$.

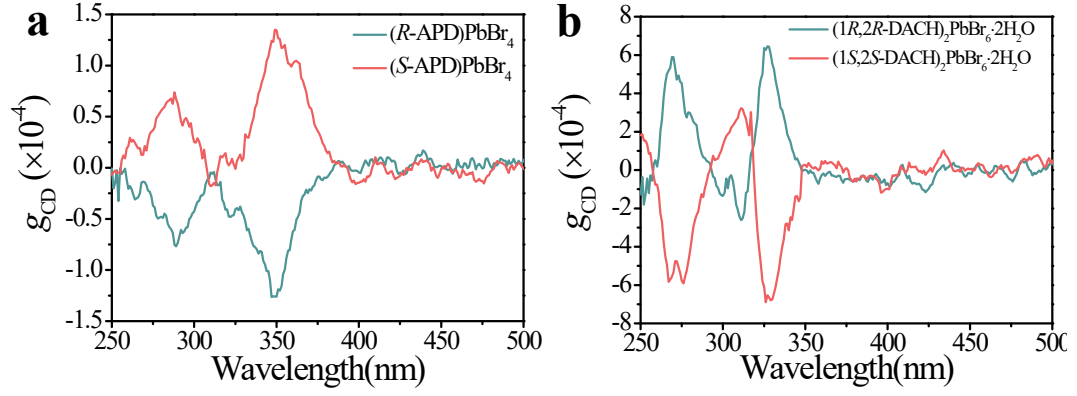


Figure S11. The g_{CD} spectra of **a.** (*R/S*-APD) $PbBr_4$ and **b.** (*1R,2R/1S,2S*-DACH) $_2PbBr_6 \cdot 2H_2O$, respectively.

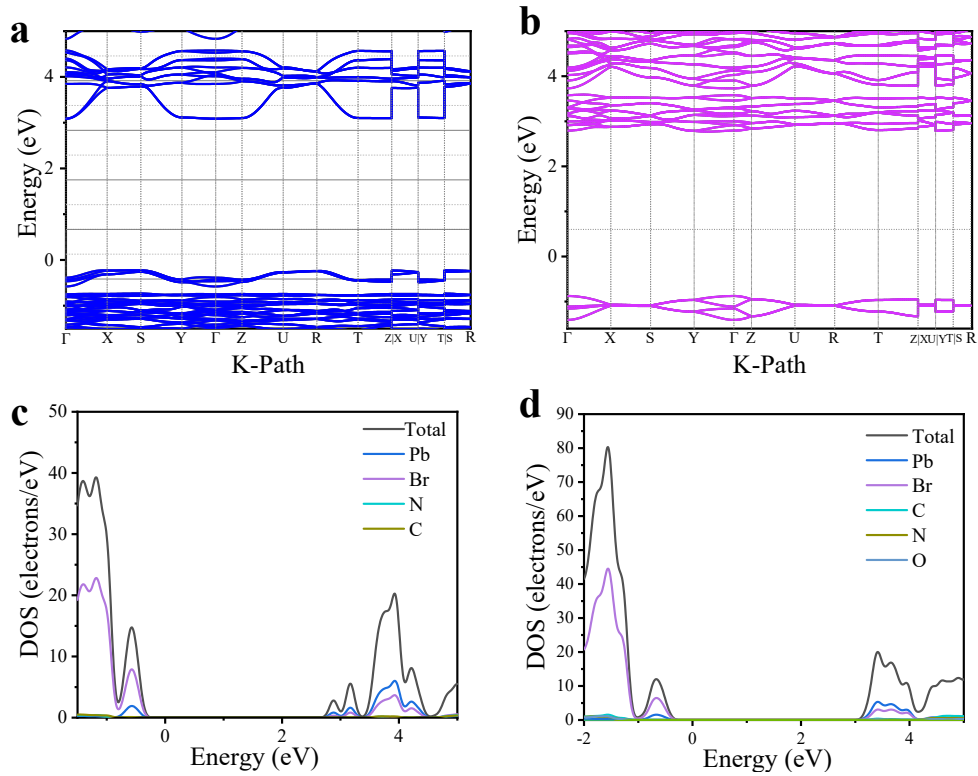


Figure S12. **a-b.** Calculated electronic band structures of (*R*-APD) $PbBr_4$ and (*1R,2R*-DACH) $_2PbBr_6 \cdot 2H_2O$. **c-d.** Calculated partial DOS of (*R*-APD) $PbBr_4$ and (*1R,2R*-DACH) $_2PbBr_6 \cdot 2H_2O$.

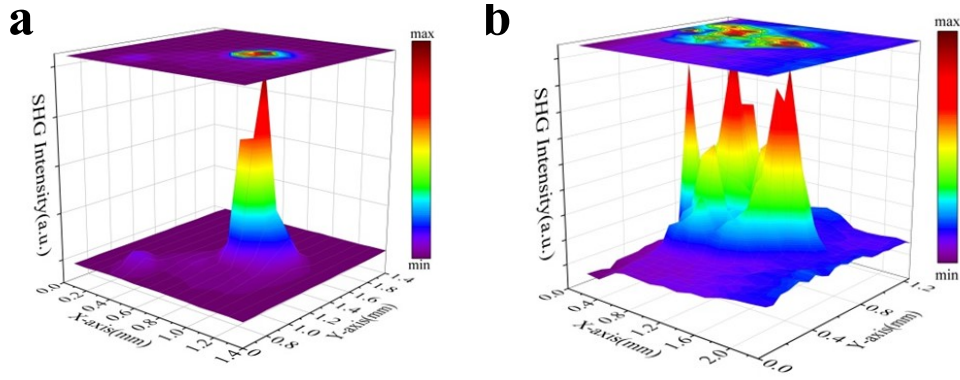


Figure S13. a. The scanned image of (S-APD)PbBr₄ crystal by detecting the SHG signal at 520 nm when pumped 1040 nm. The scanned area is $2.0 \times 1.2 \text{ mm}^2$. **b.** The scanned image of (1S,2S-DACH)₂PbBr₆·2H₂O crystal by detecting the SHG signal at 440 nm when pumped at 880 nm. The scanned area is $2.0 \times 1 \text{ mm}^2$.

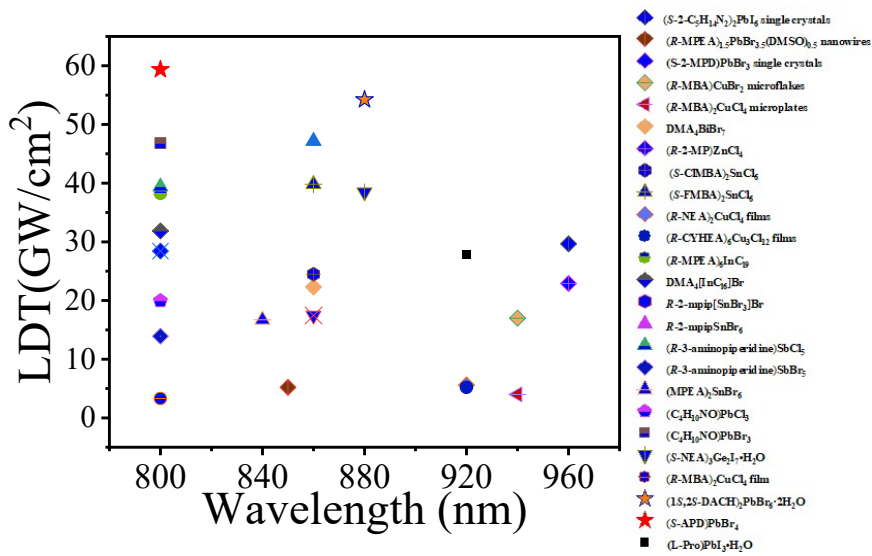


Figure S14. Comparison of LDT of chiral metal halides in literatures and this work.

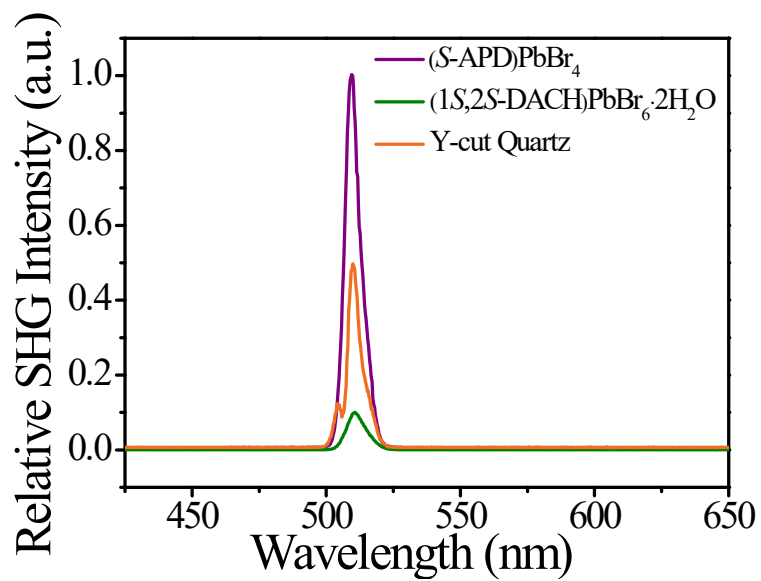


Figure S15. The comparison of SHG signal intensities of $(S\text{-APD})\text{PbBr}_4$ and $(1S,2S\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$ crystals and Y-cut quartz at 1020 nm under the same test conditions.

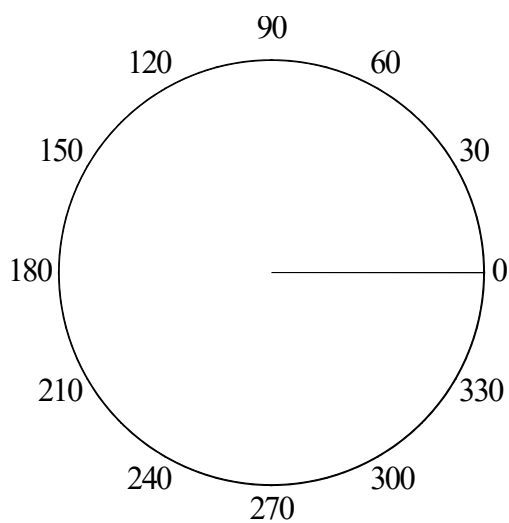


Figure S16. Polarization dependence of the SHG intensity of $(S\text{-APD})\text{PbBr}_4$ crystal as a function of the linear polarization angle pumped at 1040 nm. The purple line is the nonlinear fitting of data points.

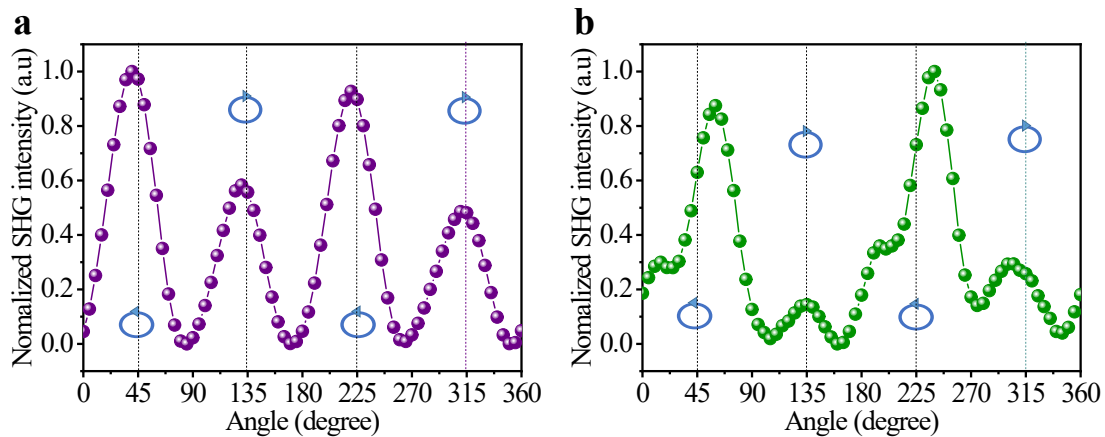


Figure S17. a-b. The SHG intensity of $(R\text{-APD})\text{PbBr}_4$ and $(1R,2R\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$ crystals at various elliptic polarized angles pumped at 1040 nm laser, respectively. The laser is left-handed circularly polarized when the rotation angle is 45 and 225°, while it is right-handed circularly polarized at 135 and 315°, as indicated by the blue arrows. It is noted that the experimental observed SHG-CD spectra can vary significantly when measured at different spots on the same crystal plane, which may be attribute to the anisotropy of the crystal surface. Unless otherwise noted, the SHG-CD results reported herein represents that acquired under optimal condition.

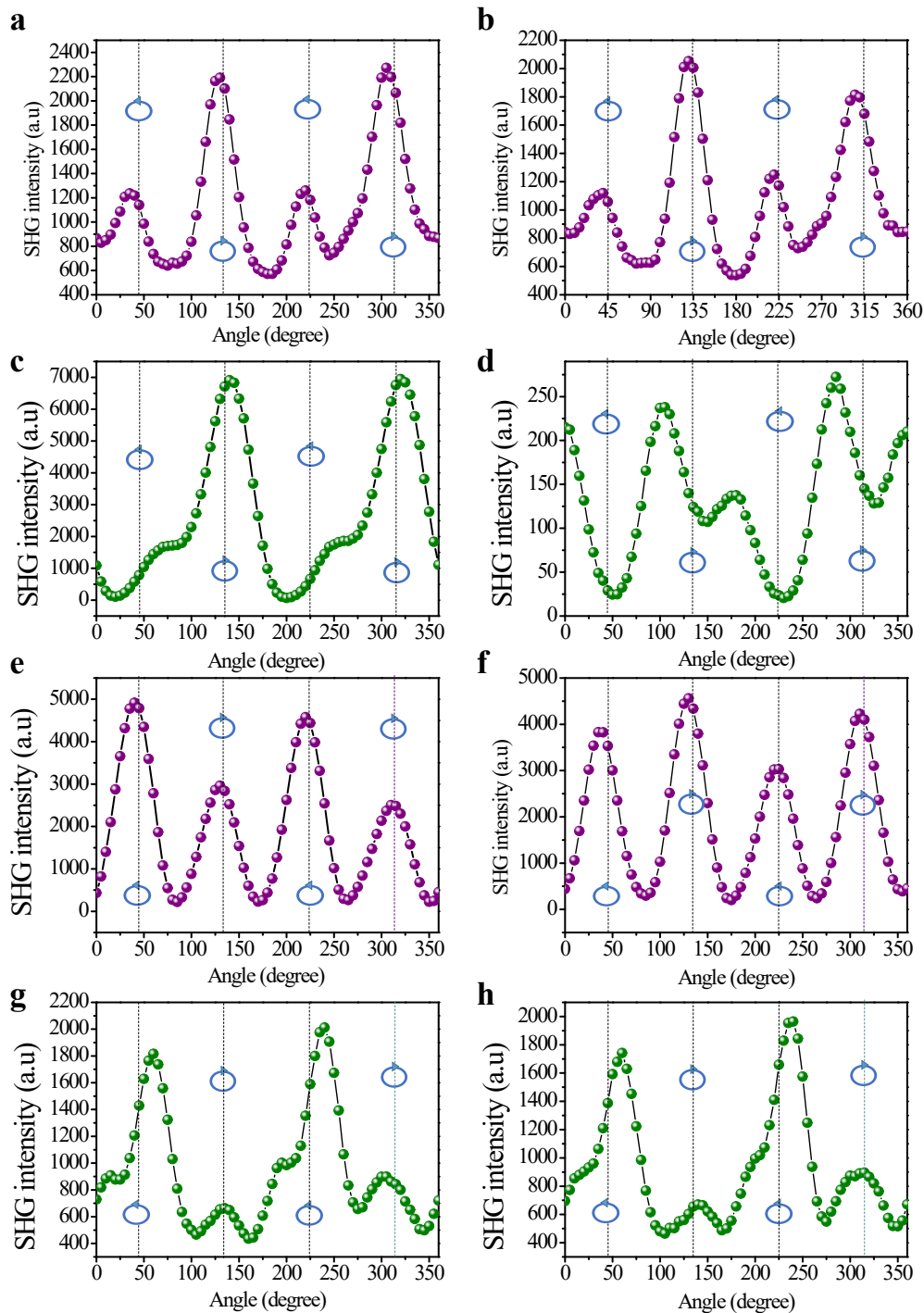


Figure S18. Summary of SHG-CD intensity at various elliptic polarized angles measured on different crystals. **a.** and **b.** The SHG-CD intensity of (*S*-APD)PbBr₄ crystals with thicknesses of ~200 and ~500 μm, respectively. **c.** and **d.** The SHG-CD intensity of (1*S*,2*S*-DACH)₂PbBr₆·2H₂O crystals with thicknesses of ~200 and ~400 μm, respectively. **e.** and **f.** The SHG-CD intensity of (*R*-APD)PbBr₄ crystals with thicknesses of ~250 and ~500 μm, respectively. **g.** and **h.** The SHG-CD intensity of (1*R*,2*R*-DACH)₂PbBr₆·2H₂O crystals with thicknesses of ~300 and ~500 μm, respectively. It is noted that Figures S18a, S18c, S18e and S18g corresponding to Figure 5c, Figure 5d, Figure S17a and Figure S17b, were used only for comparison of the SHG-CD intensity of different crystals.

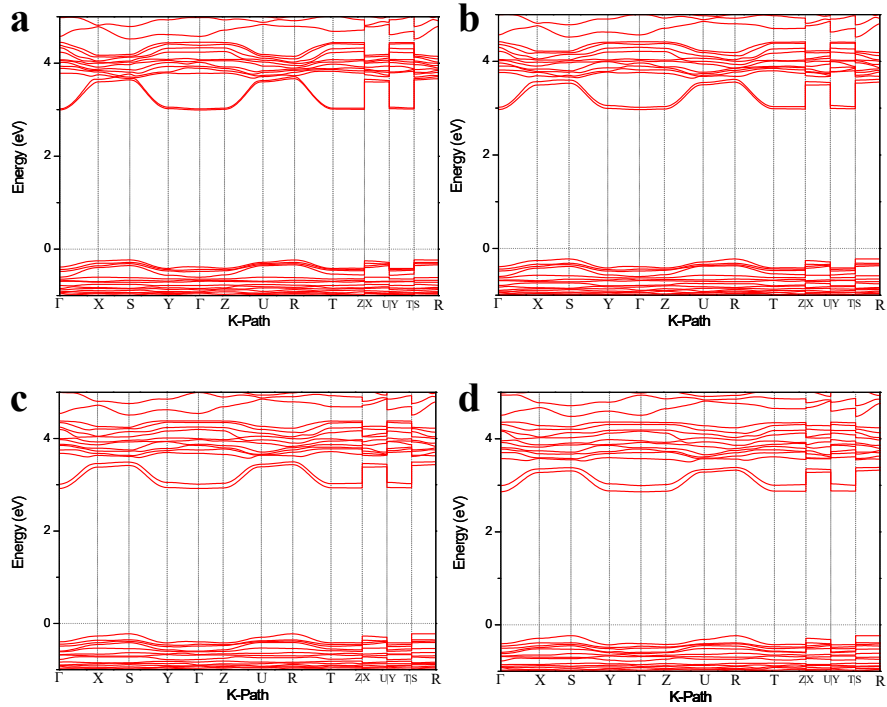


Figure S19. Electro-phonon renormalized band gaps of (S-APD)PbBr₄. Calculated band structure considering the effect of electron-phonon coupling at 0 K (a), 100 K (b), 200 K (c) and 300 K (d). The electron-phonon interactions from ZG displacements were generated accounting for thermal lattice expansion.

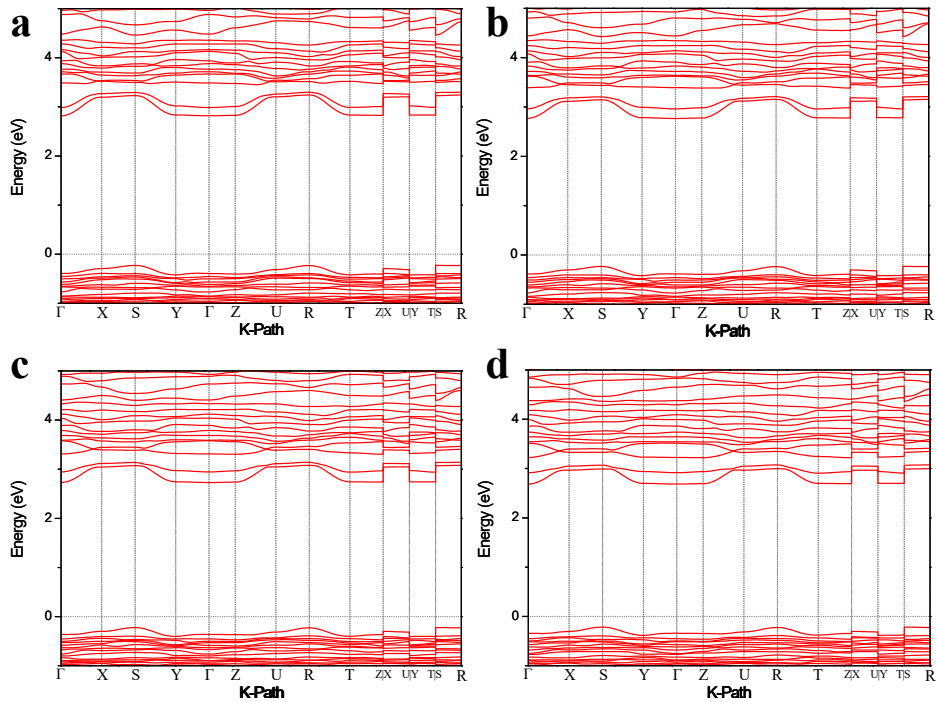


Figure S20. Electro-phonon renormalized band gaps of (S-APD)PbBr₄. Calculated band structure considering the effect of electron-phonon coupling at 400 K (a), 500 K (b), 600 K (c) and 700 K (d). The electron-phonon interactions from ZG displacements were generated accounting for thermal lattice expansion.

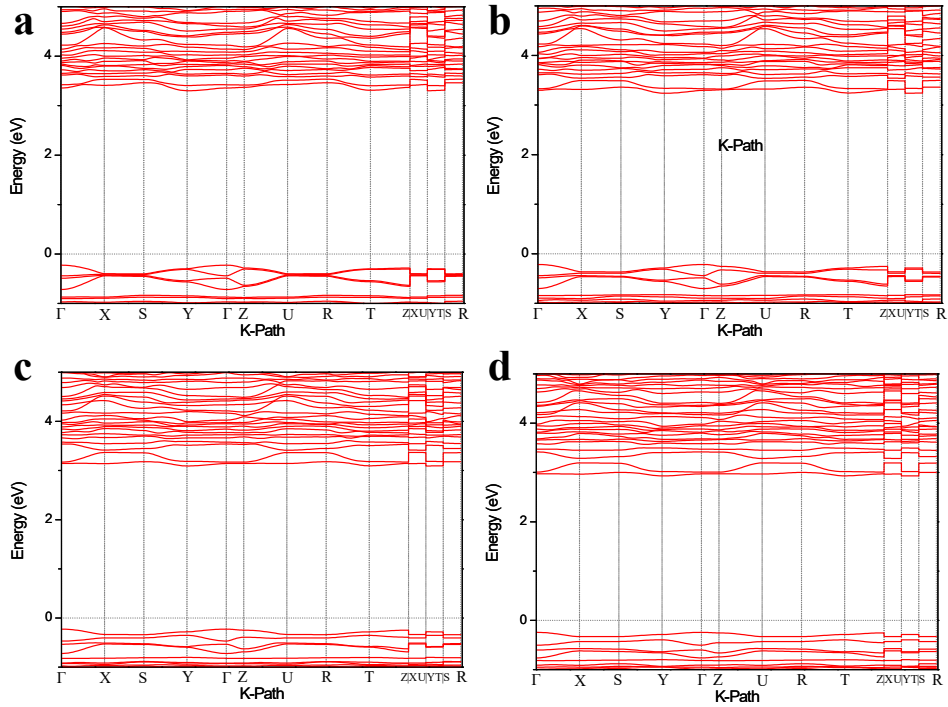


Figure S21. Electro-phonon renormalized band gaps of $(1S,2S\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$. Calculated band structure considering the effect of electron-phonon coupling at 0 K (a), 100 K (b), 200 K (c) and 300 K (d). The electron-phonon interactions from ZG displacements were generated accounting for thermal lattice expansion.

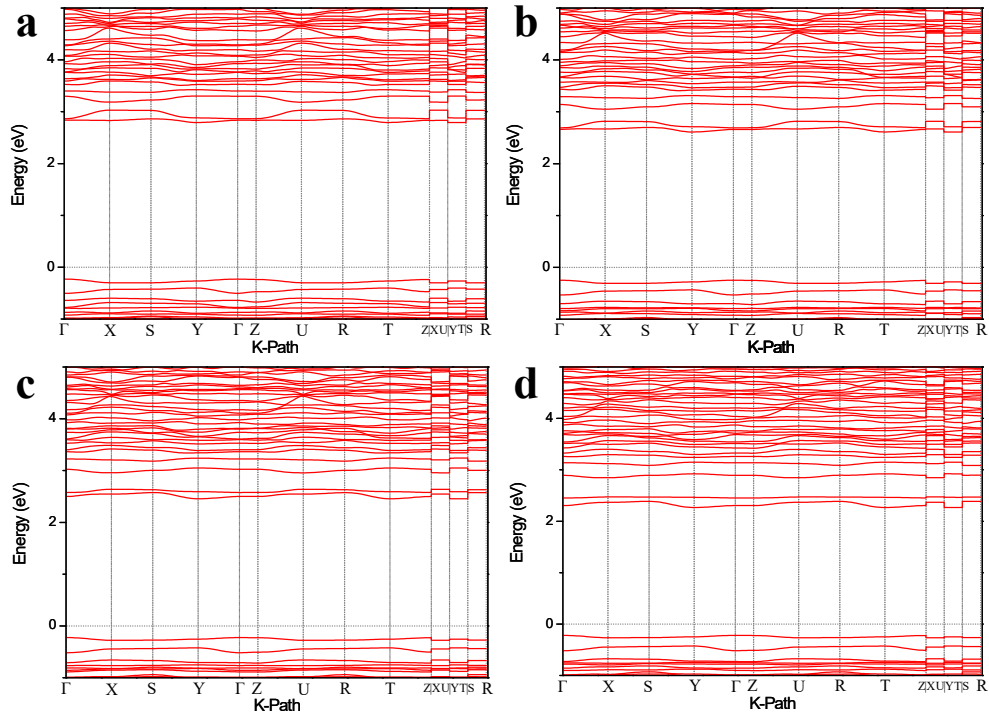


Figure S22. Electro-phonon renormalized band gaps of $(1S,2S\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$. Calculated band structure considering the effect of electron-phonon coupling at 400 K (a), 500 K (b), 600 K (c) and 700 K (d). The electron-phonon interactions from ZG displacements were generated accounting for thermal lattice expansion.

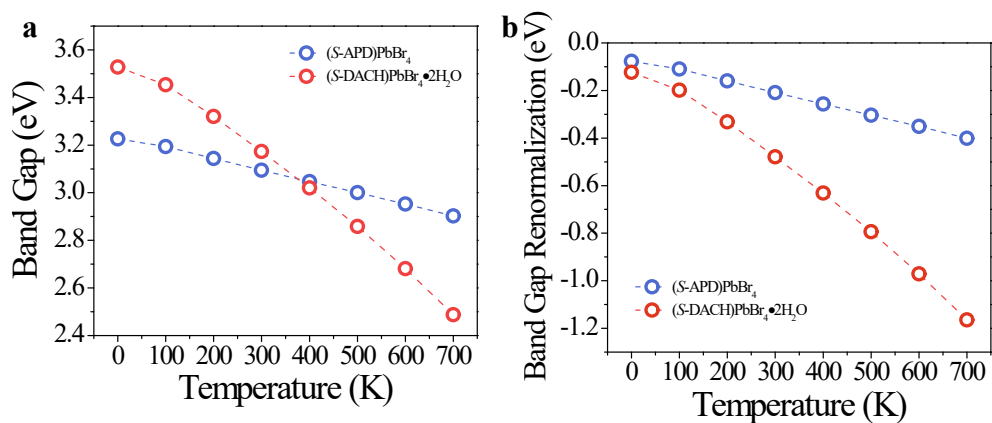


Figure S23. Electro-phonon renormalized band gaps of (S-APD)PbBr₄ and (1S,2S-DACH)₂PbBr₆·2H₂O. Phonon-induced band gap renormalization of (S-APD)PbBr₄ (blue) and (1S,2S-DACH)₂PbBr₆·2H₂O (red).

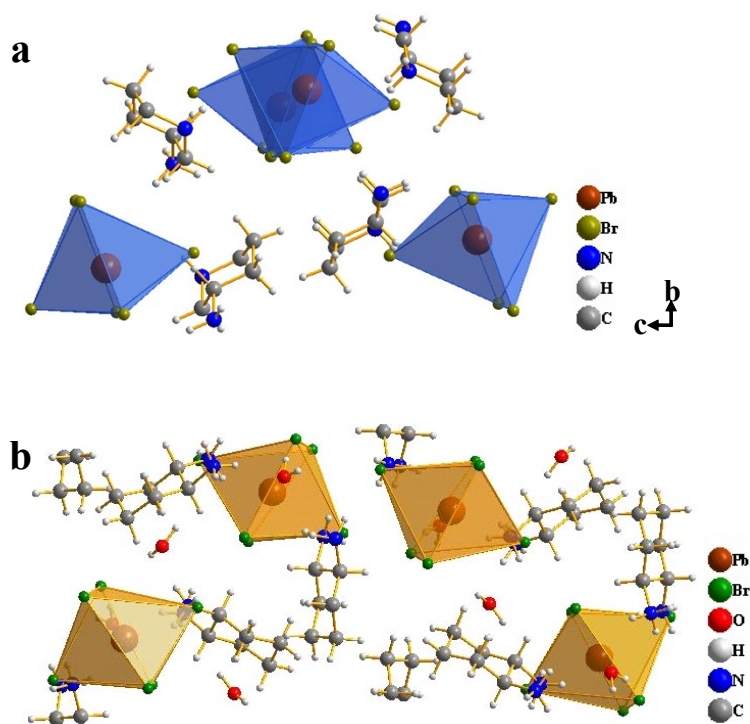


Figure S24. **a.** The unite cell structure of (S-APD)PbBr₄. **b.** The unite cell structure of (1S,2S-DACH)₂PbBr₆·2H₂O.

Table S1. Crystal data and structure refinement of (*R*-/*S*-APD)PbBr₄.

CCDC	2339434	2339435
Identification code	(<i>R</i> -APD)PbBr ₄	(<i>S</i> -APD)PbBr ₄
Empirical formula	C ₅ H ₁₄ N ₂ PbBr ₄	C ₅ H ₁₄ N ₂ PbBr ₄
Formula weight, g·mol ⁻¹	629.01	629.01
Temperature (K)	300	150
Crystal system	orthorhombic	orthorhombic
Space group	<i>P</i> 2 ₁ 2 ₁ 2 ₁	<i>P</i> 2 ₁ 2 ₁ 2 ₁
<i>a</i> (Å)	8.7802(2)	8.7810(3)
<i>b</i> (Å)	10.8321(3)	10.8005(4)
<i>c</i> (Å)	13.9609(4)	13.7783(5)
α (°)	90	90
β (°)	90	90
γ (°)	90	90
<i>V</i> (Å ³)	1327.79(6)	1306.72(8)
<i>Z</i>	4	4
ρ (calculated) (g/cm ³)	3.147	3.197
Absorption coefficient, mm ⁻¹	24.704	25.103
<i>F</i> (000)	1120.0	1120.0
Theta range for data collection	5.48 to 52.74	4.792 to 60.832
	-10 ≤ <i>h</i> ≤ 9	-11 ≤ <i>h</i> ≤ 11
Index ranges	-13 ≤ <i>k</i> ≤ 13	-13 ≤ <i>k</i> ≤ 13
	-17 ≤ <i>l</i> ≤ 17	-15 ≤ <i>l</i> ≤ 19
Reflections collected	9538	9815
Independent reflections	2707 [<i>R</i> _{int} = 0.0443]	3208 [<i>R</i> _{int} = 0.0954]
Data/restraints/parameters	2707/0/110	3208/42/110
Goodness-of-fit on <i>F</i> ²	1.095	1.170
Final <i>R</i> indices [<i>I</i> > 2σ(<i>I</i>)]	<i>R</i> ₁ = 0.0341, w <i>R</i> ₂ = 0.0795	<i>R</i> ₁ = 0.0711, w <i>R</i> ₂ = 0.1867
<i>R</i> indices (all data)	<i>R</i> ₁ = 0.0368, w <i>R</i> ₂ = 0.0804	<i>R</i> ₁ = 0.0797, w <i>R</i> ₂ = 0.1925
Largest diff. peak and hole, e.Å ⁻³	1.22 and -2.66	6.72 and -4.70
Flack parameter	-0.012(8)	-0.02(3)

Table S2. Crystal data and structure refinement of (1*R*,2*R*-/1*S*,2*S*-DACH)₂PbBr₆·2H₂O.

CCDC	2339436	2339439
Identification code	(1 <i>R</i> ,2 <i>R</i> -DACH) ₂ PbBr ₆ ·2H ₂ O	(1 <i>S</i> ,2 <i>S</i> -DACH) ₂ PbBr ₆ ·2H ₂ O
Empirical formula	C ₁₂ H ₃₆ N ₄ O ₂ PbBr ₆	C ₁₂ H ₃₆ N ₄ O ₂ PbBr ₆
Formula weight, g·mol ⁻¹	955.10	955.10
Temperature (K)	150	293
Crystal system	orthorhombic	orthorhombic
Space group	<i>P</i> 2 ₁ 2 ₁ 2 ₁	<i>P</i> 2 ₁ 2 ₁ 2 ₁
<i>a</i> (Å)	9.8774(2)	9.9329(2)
<i>b</i> (Å)	10.8539(2)	10.9337(3)
<i>c</i> (Å)	24.3071(5)	24.3960(6)
α (°)	90	90
β (°)	90	90
γ (°)	90	90
<i>V</i> (Å ³)	2605.92(9)	2649.49(11)
<i>Z</i>	4	4
ρ (calculated) (g/cm ³)	2.434	2.394
Absorption coefficient, mm ⁻¹	15.688	23.099
<i>F</i> (000)	1776	1776
Theta range for data collection	4.11 to 61.462	3.624 to 67.239
	-10 ≤ <i>h</i> ≤ 13	-8 ≤ <i>h</i> ≤ 11
Index ranges	-14 ≤ <i>k</i> ≤ 12	-12 ≤ <i>k</i> ≤ 13
	-31 ≤ <i>l</i> ≤ 29	-28 ≤ <i>l</i> ≤ 29
Reflections collected	23794	16990
Independent reflections	6672 [<i>R</i> _{int} = 0.0284]	4731 [<i>R</i> _{int} = 0.07]
Data/restraints/parameters	6672/0/236	4731 / 0 / 231
Goodness-of-fit on <i>F</i> ²	1.023	1.063
Final <i>R</i> indices [<i>I</i> > 2σ(<i>I</i>)]	<i>R</i> ₁ = 0.0206, <i>wR</i> ₂ = 0.0330	<i>R</i> ₁ = 0.0452, <i>wR</i> ₂ = 0.1141
<i>R</i> indices (all data)	<i>R</i> ₁ = 0.0249, <i>wR</i> ₂ = 0.0336	<i>R</i> ₁ = 0.0469, <i>wR</i> ₂ = 0.1167
Largest diff. peak and hole, e.Å ⁻³	0.69 and -0.63	1.841 and -2.254
Flack parameter	-0.018(3)	-0.053(6)

Table S3. Bond Lengths for (*R*-/*S*-APD)PbBr₄.

<i>(R</i> -APD)PbBr ₄			<i>(S</i> -APD)PbBr ₄		
Atom	Atom	Length/Å	Atom	Atom	Length/Å
Pb1	Br2	3.0136(13)	Pb1	Br1 ¹	3.026(3)
Pb1	Br2 ¹	3.0334(13)	Pb1	Br1	3.021(3)
Pb1	Br3	2.9560(13)	Pb1	Br2	2.943(2)
Pb1	Br4	3.0810(14)	Pb1	Br3	2.898(3)
Pb1	Br5	2.9023(14)	Pb1	Br4	3.083(3)
Pb1	Br6	3.3350(14)	Pb1	Br5	3.309(2)

Table S4. Bond Lengths for (1*R*,2*R*-/1*S*,2*S*-DACH)₂PbBr₆·2H₂O.

(1 <i>R</i> ,2 <i>R</i> -DACH) ₂ PbBr ₆ ·2H ₂ O			(1 <i>S</i> ,2 <i>S</i> -DACH) ₂ PbBr ₆ ·2H ₂ O		
Atom	Atom	Length/Å	Atom	Atom	Length/Å
Pb1	Br1	3.0154(5)	Pb1	Br6	2.9766(17)
Pb1	Br2	3.0463(5)	Pb1	Br1	2.9848(17)
Pb1	Br3	2.9752(5)	Pb1	Br2	3.0195(16)
Pb1	Br4	3.0039(5)	Pb1	Br5	3.0230(16)
Pb1	Br5	2.9911(5)	Pb1	Br3	3.0588(16)
Pb1	Br6	3.0820(5)	Pb1	Br4	3.0717(18)

Table S5. Octahedral distortion calculation for (*R*-/*S*-APD)PbBr₄ and (1*R*,2*R*-/1*S*,2*S*-DACH)₂PbBr₆·2H₂O.

Octahedral Distortion	Δd
(<i>R</i> -APD)PbBr ₄	2.05×10^{-3}
(<i>S</i> -APD)PbBr ₄	1.86×10^{-3}
(1 <i>R</i> ,2 <i>R</i> -DACH) ₂ PbBr ₆ ·2H ₂ O	1.4×10^{-4}
(1 <i>S</i> ,2 <i>S</i> -DACH) ₂ PbBr ₆ ·2H ₂ O	1.34×10^{-4}

Table S6. Comparison of LDT of chiral metal halides in literatures and this work.

	Pulse width/ fs	Repetition rate/ MHz	Laser spot diameter / μm	Power/ mW	Power intensity mJ/cm^2	LDT GW/cm^2	Ref.
$(R\text{-MPEA})_{1.5}\text{PbBr}_{3.5}(\text{DMSO})_{0.5}$ nanowires	100	82	10	67.5	0.52	5.2	1
$(S\text{-}2\text{-C}_5\text{H}_{14}\text{N}_2)_2\text{PbI}_6$ single crystals	100	80	20	745	2.97	29.66	2
$(S\text{-}2\text{-MPD})\text{PbBr}_3$ single crystals	100	80	20	714	2.84	28.42	3
$(R\text{-MBA})\text{CuBr}_2$ microflakes	100	82	-	-	1.7	17	4
$(R\text{-MBA})_2\text{CuCl}_4$ microplates	100	82	-	-	0.4	4	4
$\text{DMA}_4\text{BiBr}_7$	100	80	20	563	2.23	22.3	5
$(R\text{-}2\text{-MP})\text{ZnCl}_4$	100	80	20	575	2.29	22.89	6
$(S\text{-CIMBA})_2\text{SnCl}_6$	100	80	20	615	2.45	24.48	7
$(S\text{-FMBA})_2\text{SnCl}_6$	100	80	20	1000	3.98	39.81	7
$(R\text{-NEA})_2\text{CuCl}_4$ films	100	80	-	-	0.56	5.6	8
$(R\text{-CYHEA})_6\text{Cu}_3\text{Cl}_{12}$ films	100	80	-	-	0.52	5.2	8
$(R\text{-MPEA})_6\text{InCl}_9$	100	80	20	961	3.82	38.26	9
$\text{DMA}_4[\text{InCl}_6]\text{Br}$	100	80	20	800	3.18	31.85	10
$R\text{-}2\text{-mpip}[\text{SnBr}_3]\text{Br}$	100	80	20	438	1.74	17.44	11
$R\text{-}2\text{-mpipSnBr}_6$	100	80	20	1185	4.72	47.17	11
$(R\text{-}3\text{-aminopiperidine})\text{SbCl}_5$	100	80	20	961	3.94	39.4	12
$(R\text{-}3\text{-aminopiperidine})\text{SbBr}_5$	100	80	20	350	1.39	13.93	12
$(\text{MPEA})_2\text{SnBr}_6$	100	80	20	420	1.67	16.72	13
$(\text{C}_4\text{H}_{10}\text{NO})\text{PbCl}_3$	140	80	2	-	-	19.94	14
$(\text{C}_4\text{H}_{10}\text{NO})\text{PbBr}_3$	140	80	2	-	-	46.82	14
$(S\text{-NEA})_3\text{Ge}_2\text{I}_7 \cdot \text{H}_2\text{O}$	100	80	6	-	-	38.46	15
$(R\text{-MBA})_2\text{CuCl}_4$ film	100	80	20	-	-	3.31	16
$(\text{L-Pro})\text{PbI}_3 \cdot \text{H}_2\text{O}$	100	80	20	700	2.8	27.87	17
$(1S,2S\text{-DACH})_2\text{PbBr}_6 \cdot 2\text{H}_2\text{O}$	100	80	20	1362	5.42	54.22	This work
$(S\text{-APD})\text{PbBr}_4$	100	80	20	1491	5.94	59.36	This work

$$Power_{intensity} = \frac{P}{f\pi r^2}$$

$$LDT = \frac{P}{f\pi r^2 t}$$

The laser-induced damage threshold (LDT) was defined as $LDT = P/(F \times \pi r^2 t)$, where P, denoted incident power when the SHG signal is offset from the square dependence, F, r and t are frequency and radius of laser spot, pulse width, respectively.

Table S7. Comparison of $g_{\text{SHG-CD}}$ in the literature and this work.

Chiral compounds	Morphology	Laser conditions	Anisotropic g factor	Ref
[(<i>R/S</i>)-3-aminopiperidine]PbI ₄	single crystal	1064 nm, pulse power of 400mV	$g_{\text{SHG-CD}} = 0.21$	18
(<i>R</i> -MBA) ₄ Bi ₂ Br ₁₀	spiral microplates.	1200 nm, 100 fs, 1 kHz	$g_{\text{SHG-CD}} = 0.58$	19
(<i>R</i> -MBACl) ₂ PbI ₄	thin film	880 nm, 100 fs, 80MHz	$g_{\text{SHG-CD}} = 1.57$	20
<i>P</i> -DMA ₄ [InCl ₆]Br	single crystal	1040 nm, 100 fs, 80 MHz	$g_{\text{SHG-CD}} = 1.56$	10
(<i>R</i> -1-(1-NEA) ₂ CuCl ₄	thin film	880 nm, 100 fs, 80 MHz	$g_{\text{SHG-CD}} = 0.41$	21
(<i>R</i> -1-(2-NEA) ₂ CuCl ₄	thin film	880 nm, 100 fs, 80 MHz	$g_{\text{SHG-CD}} = 0.1$	21
(<i>R/S</i> -MPEA)BAPbBr ₄	thin film	800 nm, 100 fs, 80 MHz	$g_{\text{SHG-CD}} = 0.8$	22
(<i>R</i> -NEA) ₃ Ge ₂ I ₇ ·H ₂ O	single crystal	880 nm, 100 fs, 80 MHz	$g_{\text{SHG-CD}} = 0.48$	15
(<i>S</i> -NEA) ₃ Ge ₂ I ₇ ·H ₂ O	single crystal	880 nm, 100 fs, 80 MHz	$g_{\text{SHG-CD}} = 0.45$	15
(1 <i>S</i> ,2 <i>S</i> -DACH) ₂ PbBr ₆ ·2H ₂ O	single crystal	1040 nm, 100 fs, 80 MHz	$g_{\text{SHG-CD}} = 1.58$	This work
(<i>S</i> -APD)PbBr ₄	single crystal	1040 nm, 100 fs, 80 MHz	$g_{\text{SHG-CD}} = 0.60$	This work

$$g_{\text{SHG-CD}} = 2 \frac{I_{\text{LCP}} - I_{\text{RCP}}}{I_{\text{LCP}} + I_{\text{RCP}}}$$

where I_{LCP} and I_{RCP} represent the SHG intensities under LCP and RCP light excitation, respectively.

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