Supporting Information

for

Dual Tuning of Second-Harmonic Generation and Rashba Spin Splitting in Chiral Hybrid Lead Halide Perovskites via Halogen Site Engineering

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Table S1. Calculated values of E_g^{PBE}, E_g^{SOC}, E_g^{HSE}, static SHG tensor d_{ij}, effective powder SHG response d_{eff}^{p} at static limit and 1064 nm, and birefringence at 1064 nm for theoretical (R)-o-BrMBAPbI$_3$-theory.

Material	E_g^{PBE} (eV)	E_g^{SOC} (eV)	E_g^{HSE} (eV)	d_{ij} (pm/V)	d_{eff}^{p}(pm/V) static limit ($\omega \rightarrow 0$)	d_{eff}^{p}(pm/V) at 1064 nm	Δn at 1064 nm
(R)-o-BrMBAPbI$_3$ -theory	2.85	2.42	3.64	d_{14} = -0.19 d_{16} = *-0.29* d_{21} = *-0.90* d_{22} = *-2.34* d_{23} = *-0.45* d_{25} = *-0.14* d_{34} = *0.15* d_{36} = *-0.20*	1.205 (3.65×d_{eff}^{KDP})	2.682 (8.13×d_{eff}^{KDP})	0.112

Table S2. Bond length values after structure optimization for (S)-o-BrMBAPbI$_3$, as well as the experimental values from this work.

Bonds	(S)-o-BrMBAPbI$_3$ Bond lengths (Å)		
	Experimental	PBE-GGA	DFT-D3
Pb1-I1	3.287	3.273	3.289
Pb1-I2	3.169	3.201	3.198
Pb1-I3	3.481	3.404	3.404
Pb1-I4	3.051	3.121	3.123
Pb1-I5	3.275	3.292	3.292
Pb1-I6	3.192	3.217	3.228
Avg.	3.242	3.251	3.256
C-C	1.421	1.427	1.427
C-Br	1.929	1.916	1.919
N-C	1.512	1.524	1.524
N-H	0.891	1.038	1.037

Table S3. Bond length values after structure optimization for (S)-m-$BrMBA_2PbI_4$, as well as the experimental values obtained from reference[1].

Bonds	(S)-m-$BrMBA_2PbI_4$ Bond lengths (Å)		
	Experimental[1]	PBE-GGA	DFT-D3
Pb1-I1	3.183	3.214	3.216
Pb1-I2	3.182	3.181	3.188
Pb1-I3	3.176	3.192	3.200
Pb1-I4	3.208	3.242	3.236
Pb1-I5	3.308	3.323	3.326
Pb1-I6	3.149	3.169	3.160
Avg.	3.201	3.220	3.221
C-C	1.419	1.426	1.426
C-Br	1.902	1.907	1.910
N-C	1.511	1.512	1.511
N-H	0.910	1.043	1.043

Table S4. Bond length values after structure optimization for (S)-p-$BrMBA_2PbI_4$, as well as the experimental values from this work.

Bonds	(S)-p-$BrMBA_2PbI_4$ Bond lengths (Å)		
	Experimental	PBE-GGA	DFT-D3
Pb1-I1	3.172	3.210	3.205
Pb1-I2	3.291	3.321	3.309
Pb1-I3	3.176	3.203	3.205
Pb1-I4	3.278	3.264	3.284
Pb1-I5	3.243	3.258	3.268
Pb1-I6	3.300	3.304	3.309
Pb2-I1	3.189	3.200	3.195
Pb2-I2	3.122	3.190	3.189
Pb2-I3	3.214	3.246	3.251
Pb2-I4	3.399	3.349	3.367
Pb2-I5	3.235	3.229	3.239
Pb2-I6	3.336	3.357	3.363
Avg.	3.246	3.261	3.265
C-C	1.423	1.426	1.427
C-Br	1.948	1.908	1.910
N-C	1.544	1.516	1.516
N-H	0.891	1.042	1.042

Table S5. Optimized crystal structure data for (S)-o-BrMBAPbI$_3$.

Lattice parameters				Space group	
a=8.680Å *b*=8.058Å *c*=11.561Å *β*=99.83°				*P*2$_1$	
Atoms	*x*	*y*	*z*	Wyckoff sites	Occupancy
Pb	0.4937	0.5960	0.4930	2*a*	1.000
I1	0.5197	0.3668	0.7213	2*a*	1.000
I2	0.7834	0.8061	0.6049	2*a*	1.000
I3	0.7370	0.3523	0.3950	2*a*	1.000
C1	0.1553	0.3522	0.8810	2*a*	1.000
C2	0.0288	0.3672	0.7736	2*a*	1.000
C3	0.2269	0.1989	0.9129	2*a*	1.000
C4	0.2041	0.4883	0.9541	2*a*	1.000
C5	0.3174	0.4738	0.0548	2*a*	1.000
C6	0.3849	0.3189	0.0847	2*a*	1.000
C7	0.3398	0.1817	0.0134	2*a*	1.000
C8	0.9576	0.2044	0.7249	2*a*	1.000
Br	0.1148	0.7034	0.9166	2*a*	1.000
N	0.0935	0.4554	0.6756	2*a*	1.000
H1	0.9359	0.4501	0.7935	2*a*	1.000
H2	0.0136	0.4522	0.5982	2*a*	1.000
H3	0.1967	0.4003	0.6606	2*a*	1.000
H4	0.1189	0.5790	0.6956	2*a*	1.000
H5	0.1936	0.0906	0.8580	2*a*	1.000
H6	0.3527	0.5823	0.1093	2*a*	1.000
H7	0.4723	0.3066	0.1641	2*a*	1.000
H8	0.3920	0.0606	0.0365	2*a*	1.000
H9	0.0433	0.1232	0.6933	2*a*	1.000
H10	0.8625	0.2285	0.6513	2*a*	1.000
H11	0.9081	0.1379	0.7927	2*a*	1.000

Table S6. Optimized crystal structure data for (S)-m-$BrMBA_2PbI_4$.

Lattice parameters				Space group	
a=9.155Å *b*=8.225Å *c*=17.778Å *β*=99.05°				$P2_1$	
Atoms	*x*	*y*	*z*	Wyckoff sites	Occupancy
Pb	0.7515	0.7335	0.0013	2*a*	1.000
I1	0.8388	0.6762	0.1817	2*a*	1.000
I2	0.5488	0.4165	0.9897	2*a*	1.000
I3	0.6725	0.7213	0.8196	2*a*	1.000
I4	0.9227	0.0787	0.0215	2*a*	1.000
C1	0.2718	0.6834	0.2073	2*a*	1.000
C2	0.2357	0.7081	0.2868	2*a*	1.000
C3	0.1679	0.7419	0.4350	2*a*	1.000
C4	0.2054	0.5703	0.3280	2*a*	1.000
C5	0.1723	0.5894	0.4013	2*a*	1.000
C6	0.2000	0.8783	0.3937	2*a*	1.000
C7	0.2350	0.8619	0.3204	2*a*	1.000
C8	0.4366	0.6870	0.2037	2*a*	1.000
C9	0.2174	0.6511	0.7629	2*a*	1.000
C10	0.2955	0.6994	0.6974	2*a*	1.000
C11	0.2934	0.8618	0.6733	2*a*	1.000
C12	0.3718	0.9053	0.6154	2*a*	1.000
C13	0.4511	0.7916	0.5799	2*a*	1.000
C14	0.4484	0.6294	0.6026	2*a*	1.000
C15	0.3714	0.5832	0.6610	2*a*	1.000
C16	0.0515	0.6808	0.7479	2*a*	1.000
Br1	0.1314	0.4012	0.4574	2*a*	1.000
Br2	0.3647	0.1266	0.5825	2*a*	1.000
N1	0.1931	0.8080	0.1533	2*a*	1.000
N2	0.2838	0.7432	0.8339	2*a*	1.000

Table S6. The optimized crystal structure data for (S)-m-$BrMBA_2PbI_4$ (continued).

Atoms	*x*	*y*	*z*	Wyckoff sites	Occupancy
H1	0.2257	0.5659	0.1857	2*a*	1.000
H2	0.0801	0.8048	0.1564	2*a*	1.000
H3	0.2043	0.7791	0.0977	2*a*	1.000
H4	0.2337	0.9260	0.1641	2*a*	1.000
H5	0.1412	0.7543	0.4922	2*a*	1.000
H6	0.2080	0.4499	0.3025	2*a*	1.000
H7	0.1982	0.9984	0.4196	2*a*	1.000
H8	0.2612	0.9706	0.2898	2*a*	1.000
H9	0.4582	0.6595	0.1460	2*a*	1.000
H10	0.4923	0.5952	0.2427	2*a*	1.000
H11	0.4847	0.8064	0.2206	2*a*	1.000
H12	0.2564	0.8663	0.8303	2*a*	1.000
H13	0.3991	0.7343	0.8429	2*a*	1.000
H14	0.2443	0.6979	0.8816	2*a*	1.000
H15	0.2397	0.5221	0.7766	2*a*	1.000
H16	0.2328	0.9544	0.6998	2*a*	1.000
H17	0.5128	0.8280	0.5352	2*a*	1.000
H18	0.5086	0.5391	0.5748	2*a*	1.000
H19	0.3713	0.4563	0.6789	2*a*	1.000
H20	0.0266	0.8097	0.7359	2*a*	1.000
H21	0.9999	0.6450	0.7971	2*a*	1.000
H22	0.0020	0.6091	0.6985	2*a*	1.000

Table S7. Optimized crystal structure data for (S)-p-BrMBA$_2$PbI$_4$.

Lattice parameters				Space group	
a=12.722Å *b*=12.768Å *c*=32.749Å *β*=93.92°				*C*2	
Atoms	*x*	*y*	*z*	Wyckoff sites	Occupancy
Pb1	0.7452	0.1606	0.9987	4*c*	1.000
Pb2	0.7526	0.9734	0.4988	4*c*	1.000
I1	0.7837	0.9576	0.5979	4*c*	1.000
I2	0.6916	0.2298	0.5006	4*c*	1.000
I3	0.1859	0.9107	0.0014	4*c*	1.000
I4	0.7637	0.9778	0.4016	4*c*	1.000
I5	0.7518	0.1675	0.9010	4*c*	1.000
I6	0.7698	0.1344	0.0962	4*c*	1.000
I7	0.5000	0.0853	0.0000	2*a*	1.000
I8	0.5000	0.7136	0.0000	2*a*	1.000
I9	0.5000	0.5340	0.5000	2*b*	1.000
I10	0.5000	0.8963	0.5000	2*b*	1.000
C1	0.5140	0.2090	0.6515	4*c*	1.000
C2	0.8723	0.0048	0.7892	4*c*	1.000
C3	0.4454	0.2379	0.6813	4*c*	1.000
C4	0.6163	0.1762	0.6643	4*c*	1.000
C5	0.5784	0.1979	0.7347	4*c*	1.000
C6	0.6488	0.1704	0.7057	4*c*	1.000
C7	0.0778	0.9517	0.8173	4*c*	1.000
C8	0.9523	0.9995	0.7622	4*c*	1.000
C9	0.0553	0.9735	0.7758	4*c*	1.000
C10	0.4763	0.2080	0.6067	4*c*	1.000
C11	0.5212	0.4190	0.8887	4*c*	1.000
C12	0.5133	0.1137	0.5833	4*c*	1.000
C13	0.4632	0.3195	0.8991	4*c*	1.000
C14	0.8958	0.9807	0.8303	4*c*	1.000
C15	0.4983	0.4535	0.8448	4*c*	1.000
C16	0.4769	0.2329	0.7230	4*c*	1.000
C17	0.3863	0.8717	0.2777	4*c*	1.000
C18	0.6323	0.0196	0.2173	4*c*	1.000
C19	0.6415	0.9215	0.2359	4*c*	1.000
C20	0.3443	0.7792	0.2608	4*c*	1.000

Table S7. The optimized crystal structure data for (S)-p-BrMBA$_2$PbI$_4$ (continued).

Atoms	*x*	*y*	*z*	Wyckoff sites	Occupancy
C21	0.6140	0.8302	0.2143	4*c*	1.000
C22	0.3351	0.6896	0.2846	4*c*	1.000
C23	0.4155	0.7828	0.3433	4*c*	1.000
C24	0.3715	0.6917	0.3257	4*c*	1.000
C25	0.5252	0.9469	0.1097	4*c*	1.000
C26	0.5768	0.8381	0.1733	4*c*	1.000
C27	0.4633	0.7829	0.3869	4*c*	1.000
C28	0.5674	0.9359	0.1538	4*c*	1.000
C29	0.4212	0.8728	0.3191	4*c*	1.000
C30	0.5957	0.0260	0.1763	4*c*	1.000
C31	0.5646	0.7194	0.3924	4*c*	1.000
C32	0.4228	0.8888	0.0992	4*c*	1.000
Br1	0.6927	0.9149	0.2920	4*c*	1.000
Br2	0.3011	0.7715	0.2039	4*c*	1.000
Br3	0.6234	0.1852	0.7913	4*c*	1.000
Br4	0.9172	0.0231	0.7053	4*c*	1.000
N1	0.4948	0.5057	0.9178	4*c*	1.000
N2	0.5083	0.3079	0.5854	4*c*	1.000
N3	0.3856	0.7419	0.4159	4*c*	1.000
N4	0.6077	0.9112	0.0814	4*c*	1.000
H1	0.7921	0.0252	0.7784	4*c*	1.000
H2	0.3656	0.2642	0.6720	4*c*	1.000
H3	0.6716	0.1523	0.6420	4*c*	1.000
H4	0.7281	0.1436	0.7150	4*c*	1.000
H5	0.1582	0.9311	0.8280	4*c*	1.000
H6	0.1168	0.9680	0.7544	4*c*	1.000
H7	0.3897	0.2104	0.6041	4*c*	1.000
H8	0.5513	0.5650	0.9163	4*c*	1.000
H9	0.4987	0.4787	0.9476	4*c*	1.000
H10	0.9204	0.0383	0.9114	4*c*	1.000
H11	0.1065	0.9073	0.8946	4*c*	1.000
H12	0.4816	0.1160	0.5513	4*c*	1.000

Table S7. The optimized crystal structure data for (S)-p-BrMBA$_2$PbI$_4$ (continued).

Atoms	*x*	*y*	*z*	Wyckoff sites	Occupancy
H13	0.4833	0.0426	0.5972	4*c*	1.000
H14	0.5994	0.1085	0.5837	4*c*	1.000
H15	0.5001	0.2982	0.5540	4*c*	1.000
H16	0.5852	0.3322	0.5931	4*c*	1.000
H17	0.4572	0.3688	0.5922	4*c*	1.000
H18	0.4842	0.2945	0.9307	4*c*	1.000
H19	0.4858	0.2568	0.8786	4*c*	1.000
H20	0.3774	0.3293	0.8953	4*c*	1.000
H21	0.8318	0.9826	0.8508	4*c*	1.000
H22	0.4230	0.2562	0.7458	4*c*	1.000
H23	0.3931	0.9415	0.2590	4*c*	1.000
H24	0.6527	0.0904	0.2344	4*c*	1.000
H25	0.6207	0.7539	0.2291	4*c*	1.000
H26	0.8021	0.1177	0.2713	4*c*	1.000
H27	0.8656	0.1198	0.3435	4*c*	1.000
H28	0.5145	0.0305	0.1026	4*c*	1.000
H29	0.5547	0.7664	0.1568	4*c*	1.000
H30	0.4782	0.8640	0.3969	4*c*	1.000
H31	0.4542	0.9450	0.3323	4*c*	1.000
H32	0.5890	0.1026	0.1617	4*c*	1.000
H33	0.3624	0.6647	0.4102	4*c*	1.000
H34	0.3189	0.7901	0.4142	4*c*	1.000
H35	0.4182	0.7466	0.4458	4*c*	1.000
H36	0.5508	0.6368	0.3844	4*c*	1.000
H37	0.5990	0.7237	0.4241	4*c*	1.000
H38	0.6221	0.7510	0.3722	4*c*	1.000
H39	0.4312	0.8041	0.1038	4*c*	1.000
H40	0.3938	0.9028	0.0673	4*c*	1.000
H41	0.3630	0.9171	0.1191	4*c*	1.000
H42	0.6364	0.8360	0.0881	4*c*	1.000
H43	0.5774	0.9125	0.0513	4*c*	1.000
H44	0.3289	0.9636	0.9163	4*c*	1.000

Table S8. Contributions of individual atoms to the largest SHG component in (S)-o-$BrMBAPbI_3$, (S)-m-$BrMBA_2PbI_4$, and (S)-p-$BrMBA_2PbI_4$. A_τ is the contribution (in %) from a single atom τ, and C_A that from all atoms of the same type. $^{VB}A_T$ is the contribution (in %) of the VBs, and $^{CB}A_T$ from the CBs. The contributions from the *s*, *p*, and *d* states of the atom τ to of $^{VB}A_T$ and $^{CB}A_T$ are also shown. W_A refers to the number of the same type of atoms in a unit cell.

Material	Atom	W_A	A_T	C_A	$^{VB}A_T$	$^{CB}A_T$	$^{VB}{}_sA_T$	$^{VB}{}_pA_T$	$^{VB}{}_dA_T$	$^{CB}{}_sA_T$	$^{CB}{}_pA_T$	$^{CB}{}_dA_T$
(S)-o-$BrMBAPbI_3$ d_{22}	Pb	2	7.90	15.80	3.78	4.12	1.87	0.61	1.30	0.10	3.59	0.43
	I	6	8.67	52.04	6.66	2.01	-0.75	7.41	0.00	0.15	0.69	1.18
	C	16	1.10	17.59	0.32	0.78	-0.05	0.37	0.00	0.09	0.68	0.00
	Br	2	2.67	5.34	2.24	0.43	-0.22	2.47	-0.01	-0.08	-0.19	0.70
	N	2	3.15	6.29	2.80	0.34	-0.02	2.82	0.00	-0.03	0.38	0.00
	H	22	0.13	2.94	-0.03	0.17	-0.04	0.01	0.00	0.09	0.07	0.00
(S)-m-$BrMBA_2PbI_4$ d_{22}	Pb	2	6.85	13.69	0.77	6.08	-2.42	-0.40	3.59	0.16	5.59	0.34
	I	8	0.49	3.96	-2.04	2.53	-1.13	-0.90	0.00	0.23	1.38	0.93
	C	32	1.26	40.20	0.47	0.79	-0.05	0.52	0.00	0.20	0.60	0.00
	Br	4	4.65	18.58	1.32	3.32	-0.12	1.45	0.00	0.24	1.91	1.17
	N	4	2.83	11.31	1.82	1.01	-0.02	1.84	0.00	0.59	0.43	0.00
	H	44	0.28	12.24	0.12	0.16	0.11	0.01	0.00	0.09	0.07	0.00
(S)-p-$BrMBA_2PbI_4$ d_{16}	Pb	4	2.85	11.39	6.86	-4.01	6.57	-0.28	0.57	-0.10	-3.93	0.02
	I	16	3.76	60.13	3.57	0.18	-0.39	3.97	0.00	-0.16	0.22	0.12
	C	64	0.33	21.22	-0.17	0.51	-0.03	-0.14	0.00	0.11	0.40	0.00
	Br	8	1.92	15.38	-0.89	2.81	-0.03	-0.85	0.00	0.25	1.86	0.70
	N	8	-0.73	-5.86	-1.03	0.29	-0.02	-1.01	0.00	0.02	0.27	0.00
	H	88	-0.03	-2.28	-0.07	0.04	-0.06	-0.01	0.00	0.03	0.01	0.00

Table S9. Octahedral distortion parameters (β and $\Delta\beta$) for 2D (S)-o-BrMBAPbI$_3$*, (S)-m-BrMBA$_2$PbI$_4$ and (S)-p-BrMBA$_2$PbI$_4$.

Material	β (°)	$\Delta\beta$ (°)
(S)-m-BrMBA$_2$PbI$_4$	138.63,152.69	14.06
(S)-p-BrMBA$_2$PbI$_4$	145.76,151.20,156.41 146.00,152.33,153.74	10.65

The β angle (Pb–I–Pb bond angle) disparity $(\Delta\beta)^2$ are calculated by following equations:

$$\Delta\beta = \beta_{max} - \beta_{min}$$

*In 1D (S)-o-BrMBAPbI$_3$, the adjacent PbI$_6$ octahedra are connected via three coplanar I atoms, and therefore neither the β angle nor $\Delta\beta$ is defined.

Table S10. Crystal structure data for the samples investigated in the present paper. Data for (S)-m-$BrMBA_2PbI_4$ have been taken from reference[1].

	(S)-o-$BrMBAPbI_3$	(S)-m-$BrMBA_2PbI_4$	(S)-p-$BrMBA_2PbI_4$
Formula weight	787.97	1116.96	1116.96
Wavelength (Å)	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1$	$P2_1$	$C2$
Lattice parameters (Å)	a = 8.6940 (11) b = 8.0422 (10) c = 11.5683 (16) β= 99.985 (5)	a = 9.1551 (5) b = 8.2245 (5) c = 17.7782 (10) β = 99.051 (3)	a = 12.7224 (14) b = 12.7678 (14) c = 32.749 (4) β= 93.919 (5)
Cell Volume ($Å^3$)	796.59 (18)	1321.96 (13)	5307.1 (11)
Z	2	2	8
CCDC code	2501013	2222721	2479653

Figures S1-S9

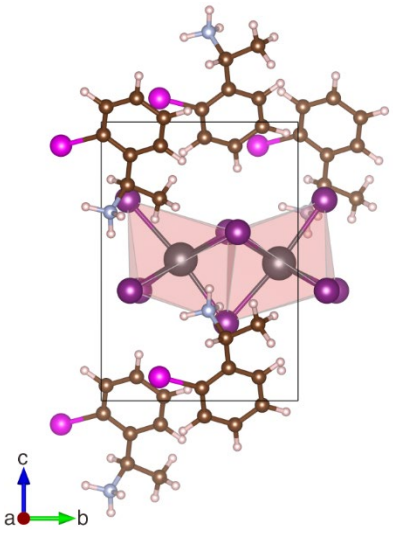

Figure S1. Crystal structures of (R)-o-BrMBAPbI$_3$-theory.

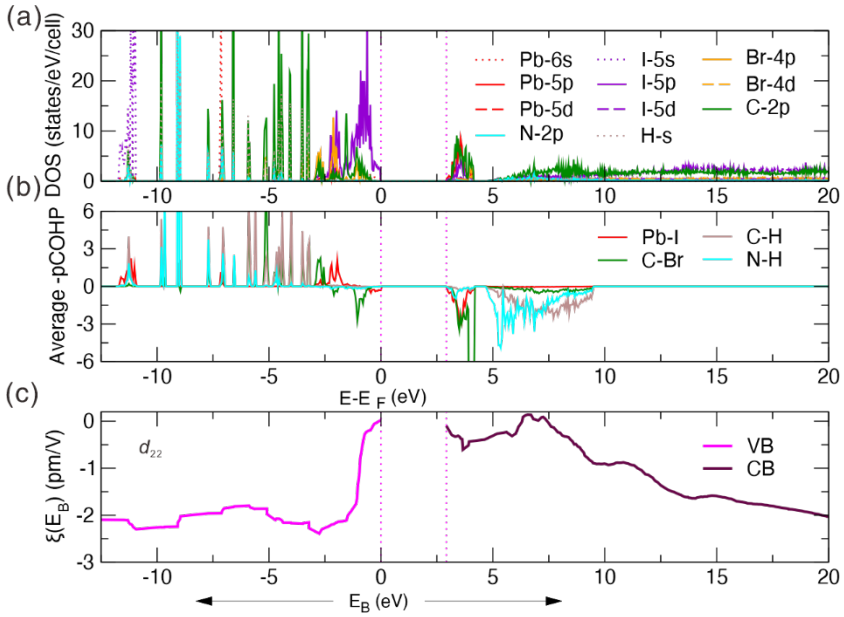

Figure S2. (a) PDOS (b) COHP analysis describing the average chemical bonding, and (c) $\zeta(E_B)$-vs-E_B plot for the largest SHG component d_{22} of (R)-o-BrMBAPbI$_3$-theory.

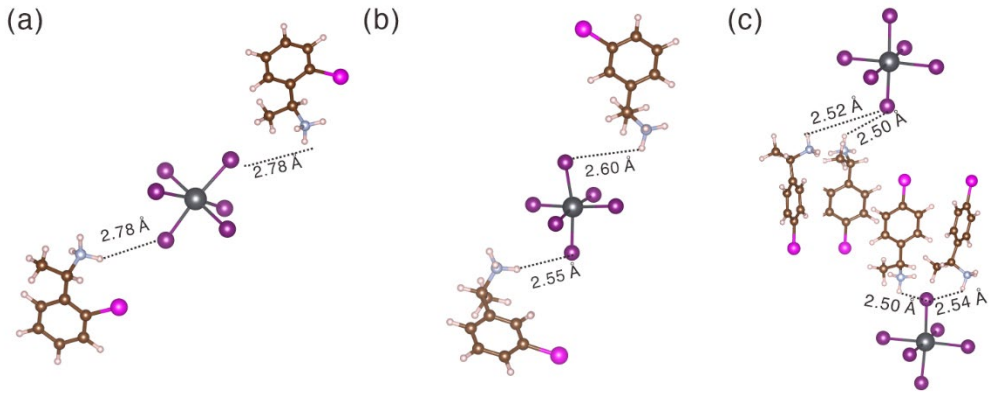

Figure S3. Coordination environments of the PbI_6 octahedra and I···NH weak interactions in (a) (S)-o-BrMBAPbI_3, (b) (S)-m-BrMBA_2PbI_4, and (c) (S)-p-BrMBA_2PbI_4.

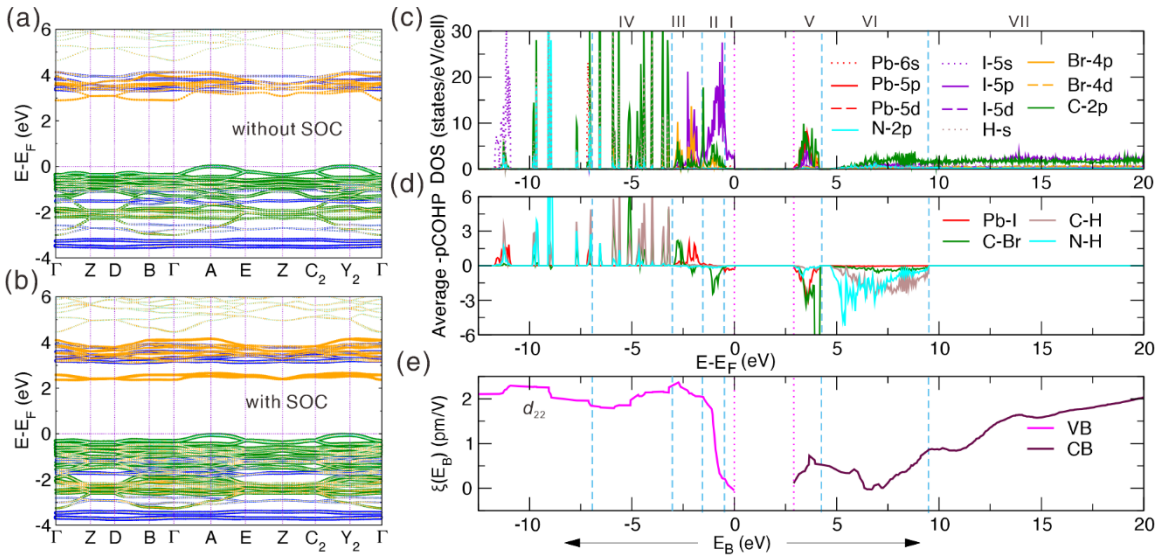

Figure S4. Electronic structures of (S)-o-BrMBAPbI$_3$. (a) Band structure without SOC and (b) with SOC. High-symmetry points: Γ = (0, 0, 0), Z = (0, 0.5, 0), D = (0, 0.5, 0.5), B = (0, 0, 0.5), A = (-0.5, 0, 0.5), E = (-0.5, 0.5, 0.5), C_2 = (-0.5, 0.5, 0), and Y_2 = (-0.5, 0, 0). Key: green = I 5p; orange = Pb 6p; blue = C 2p, cyan = Pb 6s. (c) Calculated PDOS. (d) COHP analysis describing the average chemical bonding characteristics. (e) PRF analysis for the largest SHG tensor component d_{22} of (S)-o-BrMBAPbI$_3$.

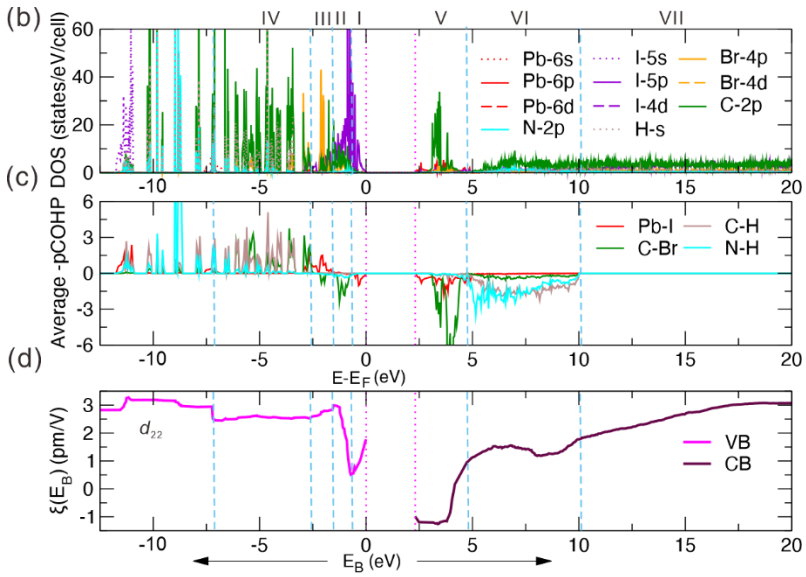

Figure S5. (a) Calculated PDOS. (b) COHP analysis describing the average chemical bonding characteristics. (c) PRF analysis for the largest SHG tensor component d_{22} of (S)-m-$BrMBA_2PbI_4$.

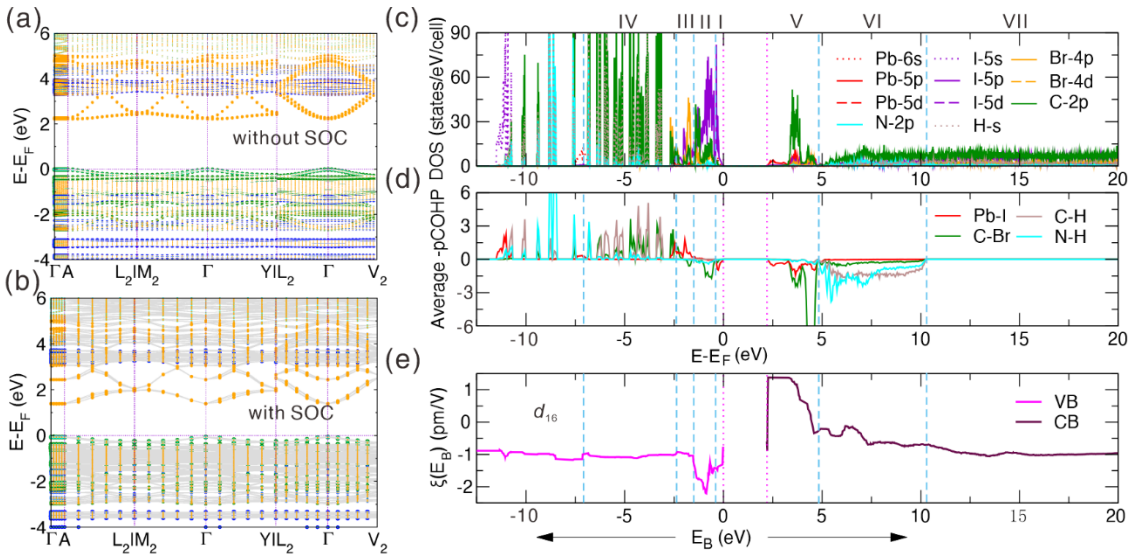

Figure S6. Band structures of (S)-p-BrMBA$_2$PbI$_4$: (a) without SOC and (b) with SOC. Γ = (0, 0, 0), A = (0, 0, 0.5), I_2 = (0.495, 0.495, 0.5), I = (−0.495, 0.504, 0.5), M_2 = (−0.5, 0.5, 0.5), Y = (0.5, 0.5, 0), L_2 = (0, 0.5, 0.5), and V_2 = (0, 0.5, 0). Key: green = I 5p; orange = Pb 6p; blue = C 2p, cyan = Pb 6s. (c) Calculated PDOS. (d) COHP analysis describing the average chemical bonding characteristics. (e) PRF for the largest SHG component d_{16} of (S)-p-BrMBA$_2$PbI$_4$.

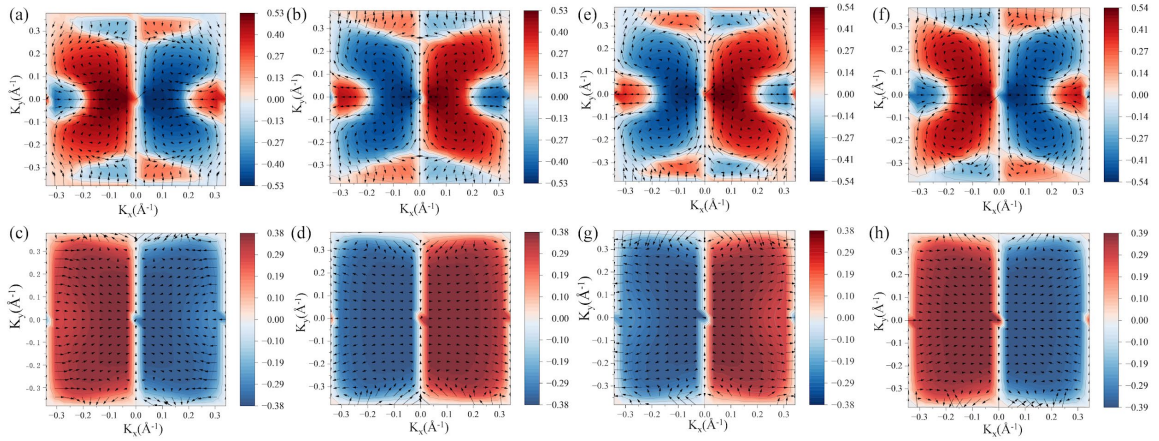

Figure S7. Spin texture of the valence and conduction band branches for (R/S)-m-BrMBA$_2$PbI$_4$. (a-d) (R)-m-BrMBA$_2$PbI$_4$: (a) outer and (b) inner VBM, (c) outer and (d) inner CBM; (e-h) (S)-m-BrMBA$_2$PbI$_4$: (e) outer and (f) inner VBM, (g) outer and (h) inner CBM. The red and blue indicate positive and negative S_z components, respectively.

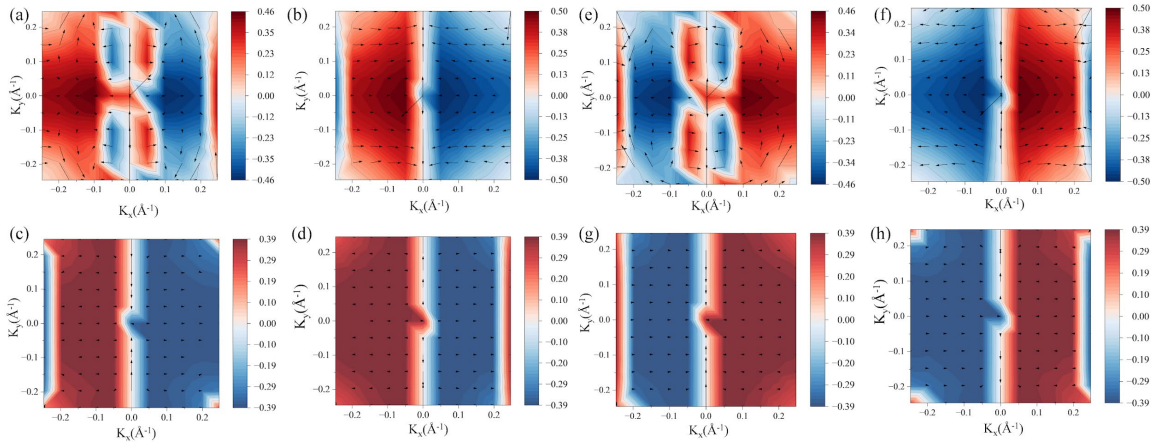

Figure S8. Spin texture of the valence and conduction band branches for (R/S)-p-$BrMBA_2PbI_4$. (a-d) (R)-p-$BrMBA_2PbI_4$: (a) outer and (b) inner VBM, (c) outer and (d) inner CBM; (e-h) (S)-p-$BrMBA_2PbI_4$: (e) outer and (f) inner VBM, (g) outer and (h) inner CBM. The red and blue indicate positive and negative S_z components, respectively.

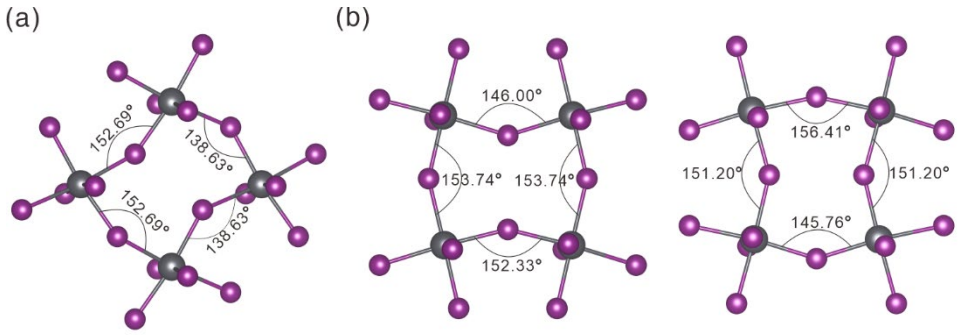

Figure S9. In-plane views of chiral OIHMH perovskite layers with *β* angles (Pb–I–Pb bond angles) for (d) (S)-m-BrMBA_2PbI_4 and (e) (S)-p-BrMBA_2PbI_4

References

(1) Liu, S. et al. Bright circularly polarized photoluminescence in chiral layered hybrid lead-halide perovskites. *Sci. Adv.* **9**, eadh5083 (2023).
(2) Jana, M. K. et al. Structural descriptor for enhanced spin-splitting in 2D hybrid perovskites. *Nat. Commun.* **12**, 4982 (2021).