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Quasi-Low-Dimensional Perovskite with Enhanced Stability Against Lattice Strain and Desired Band-Edge by A-Site Design

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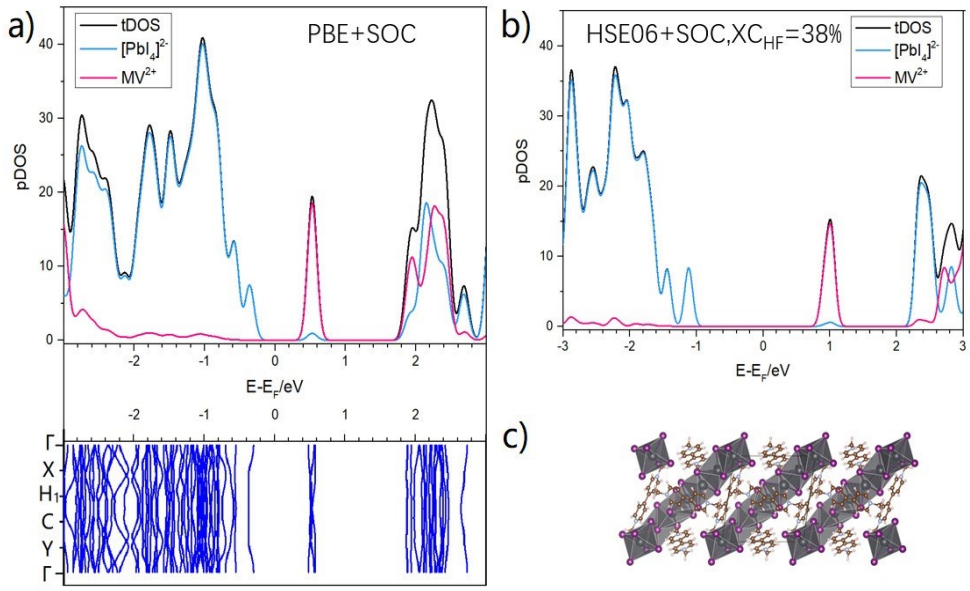

Figure S1: Computed (a) projected density-of-states and band structure at PBE+SOC level of theory, and (b) projected density-of-states at HSE06+SOC level of theory with an XC_{HF} fraction being 0.38 of the (c) structure of $MV(PbI_3)_2$ according to *Nature Communications* (2024) ***15*** : 2753.

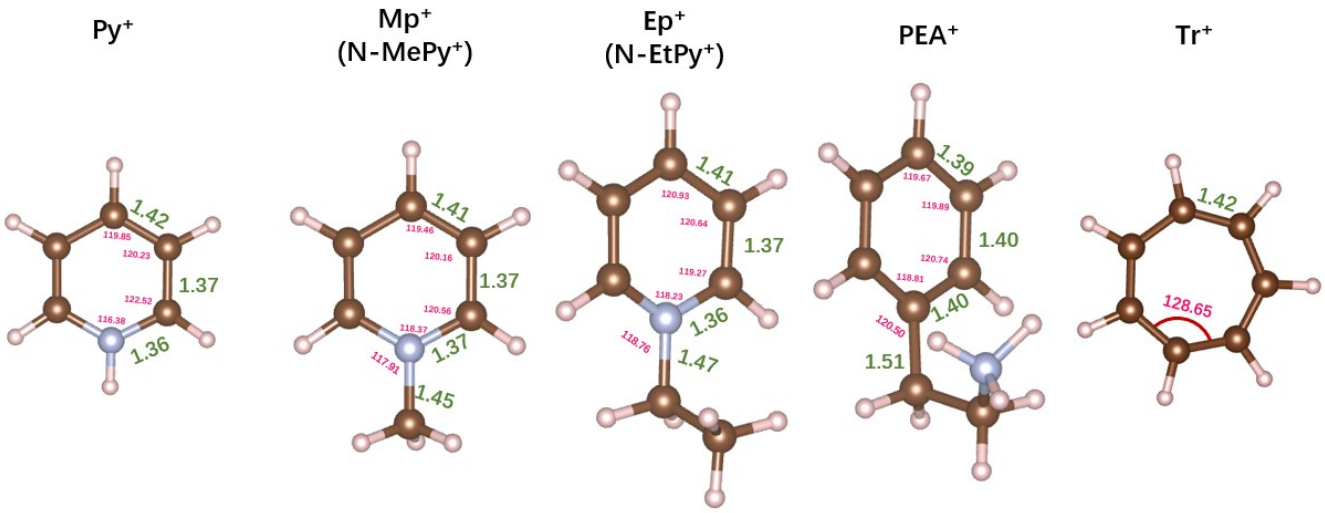

Figure S2: Optimized geometric parameters of the spacer cations adopted in this research. All the spacers are optimized as its iodide salt.

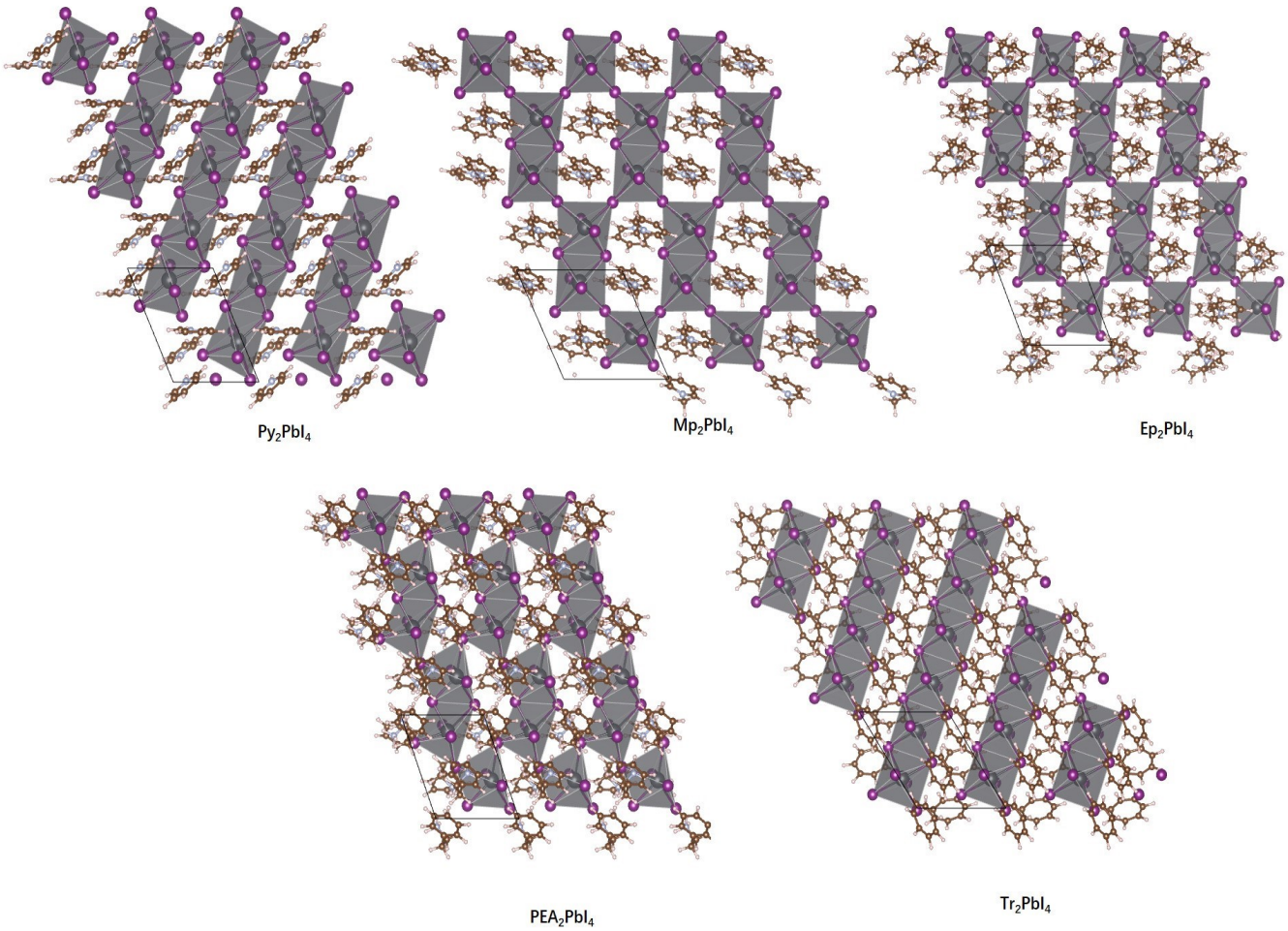

Figure S3: Top view of optimized CPAC-based QLD perovskite A_2PbI_4 supercells with various spacers.

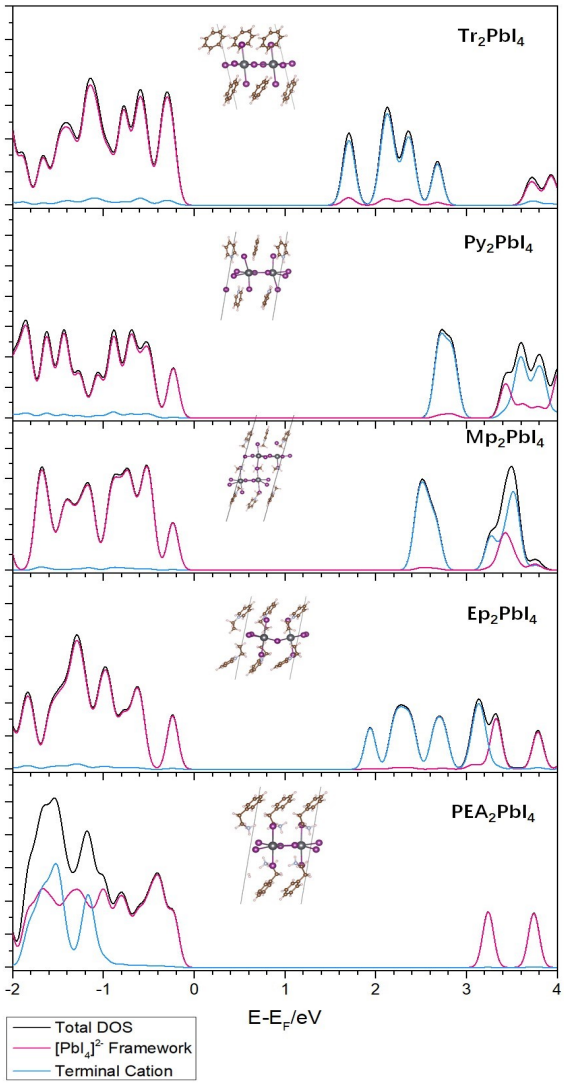

Figure S4: Computed projected density-of-states of rigid QLD A_2PbI_4 *A* = Py, Mp, Ep, Tr, and PEA for benchmark at HSE06+SOC level of theory with an XC_{HF} fraction being 0.38.

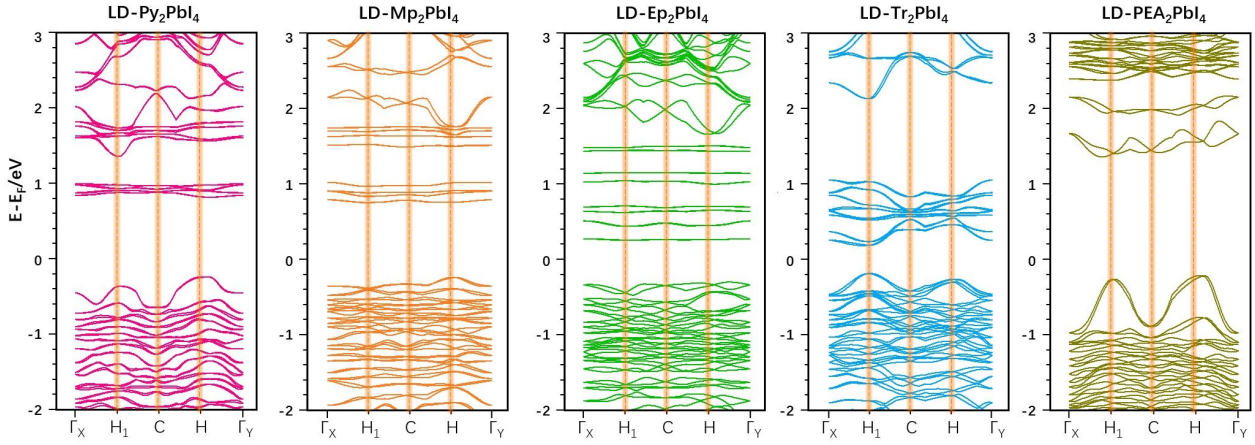

Figure S5: Computed band structure of rigid QLD A_2PbI_4 structures with A = Py, Mp, Ep, Tr, and PEA for benchmark at the PBE+SOC level of theory.

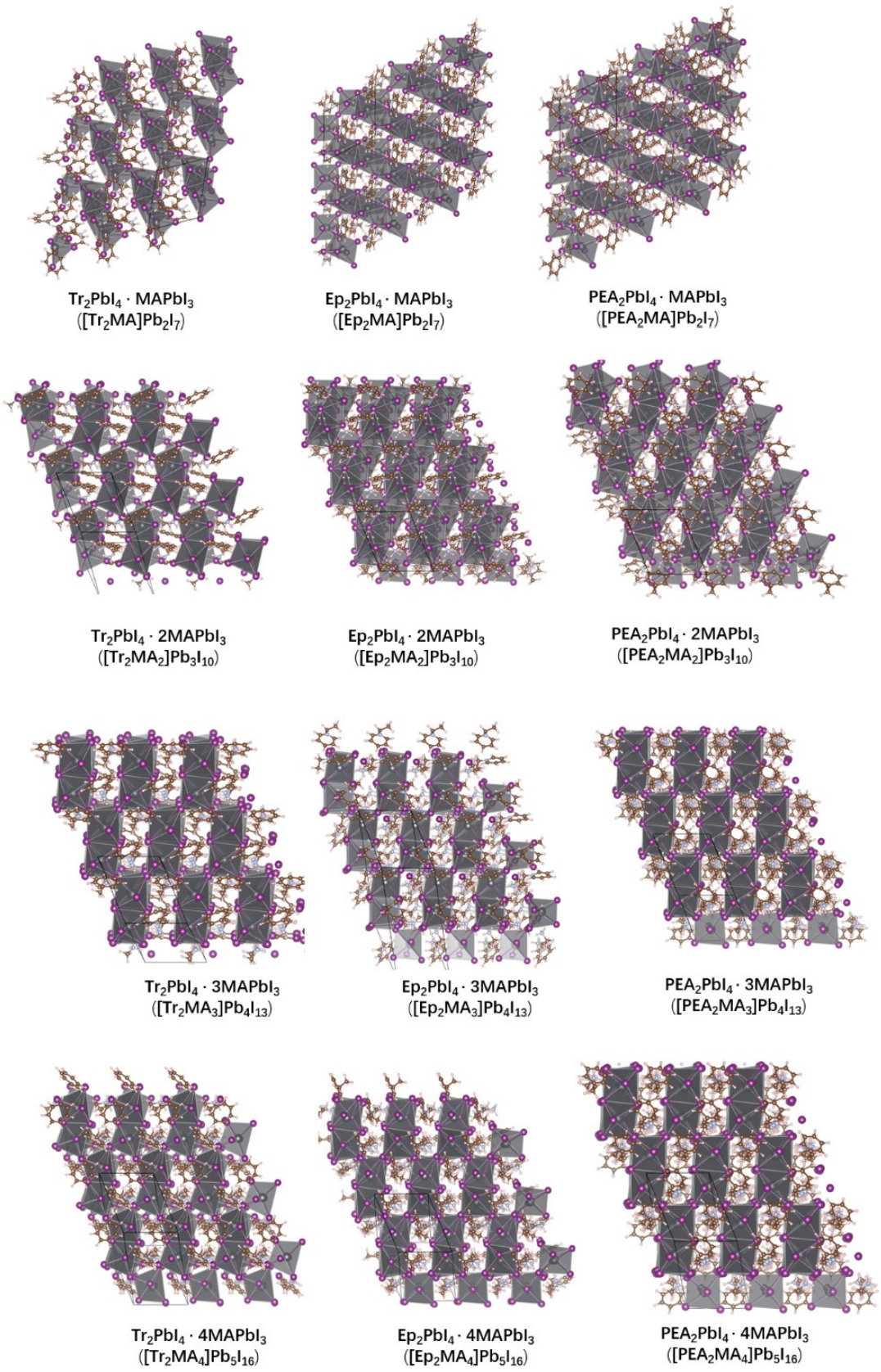

Figure S6: Optimized conformation (top view) of QLD perovskite structures $A_2PbI_4 \bullet n MAPbI_3$ *A* = Py, Mp, Ep, Tr with *n* = 1 to 4.

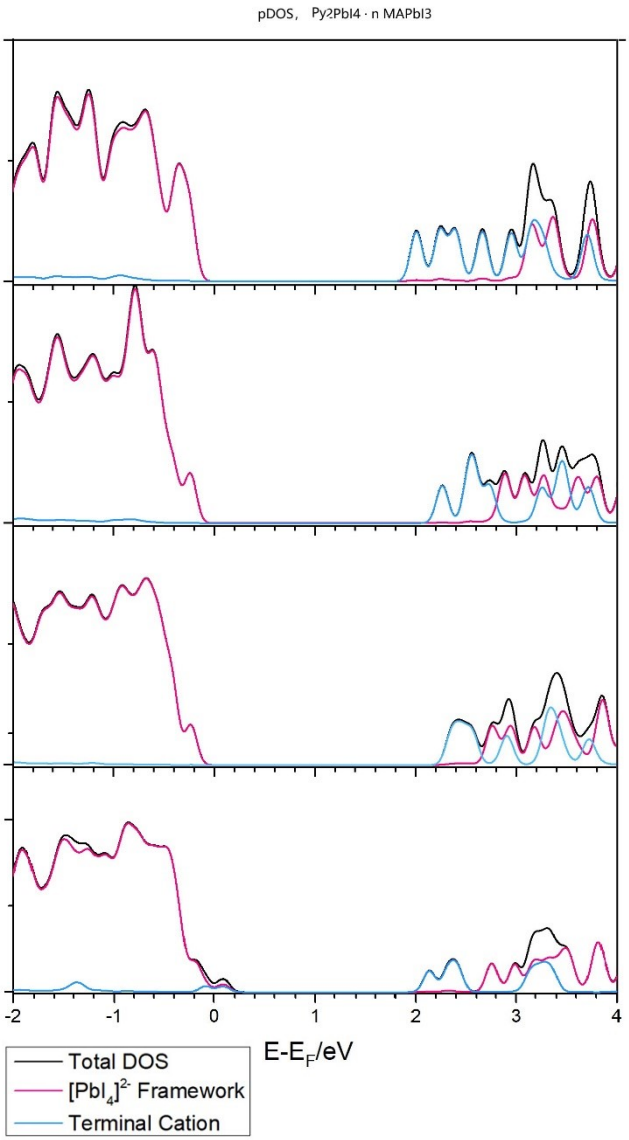

Figure S7: Computed density-of-states of QLD $Py_2PbI_4 \bullet n MAPbI_3$ with n = 1 to 4 (top - bottom panels) at HS E06+SOC level of theory with an XC_{HF} fraction being 0.38

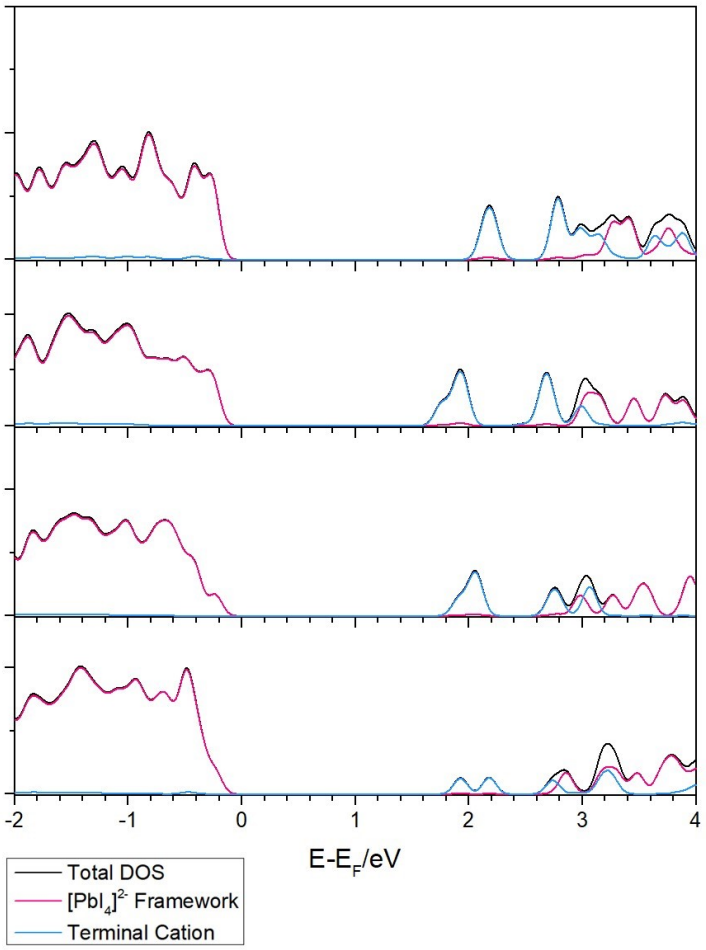

Figure S8: Computed density-of-states of QLD $Mp_2PbI_4 \bullet n$MAPbI$_3$ with n = 1 to 4 (top - bottom panels) at HSE06+SOC level of theory with an XC_{HF} fraction being 0.38

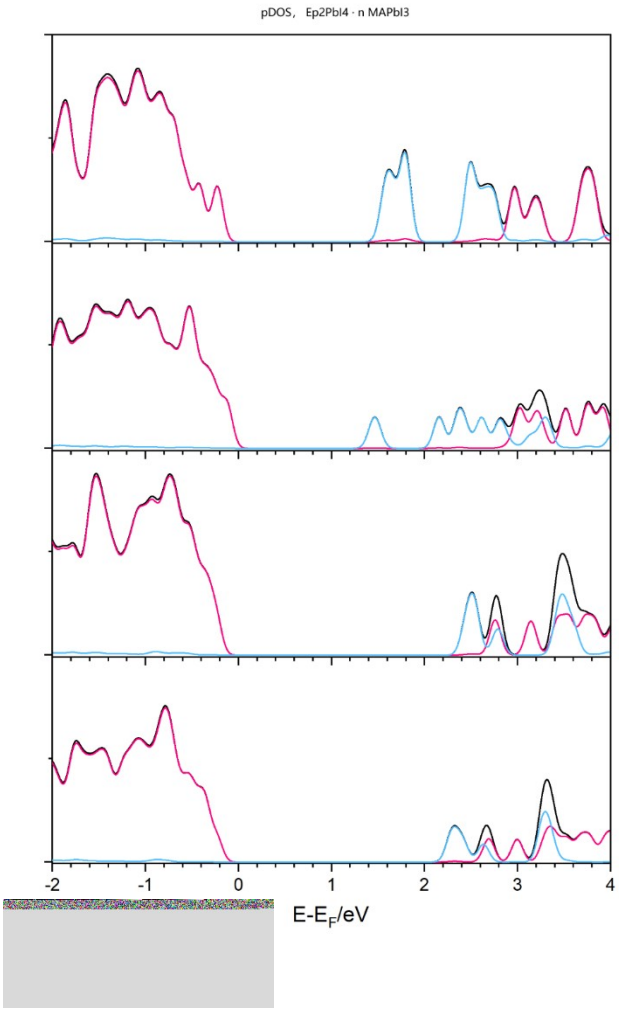

Figure S9: Computed density-of-states of QLD Ep_2PbI_4 • *n*$MAPbI_3$ with *n* = 1 to 4 (top - bottom panels) at HSE06+SOC level of theory with an XC_{HF} fraction being 0.38

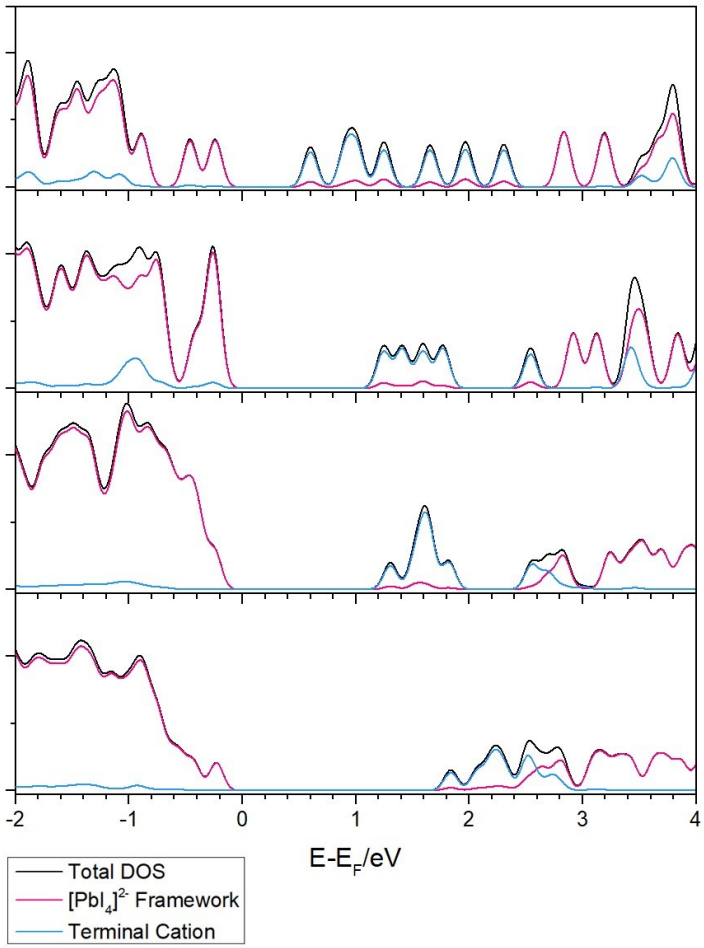

Figure S10: Computed density-of-states of QLD Tr_2PbI_4 • *n*$MAPbI_3$ with *n* = 1 to 4 (top - bottom panels) at HSE06+SOC level of theory with an XC_{HF} fraction being 0.38.

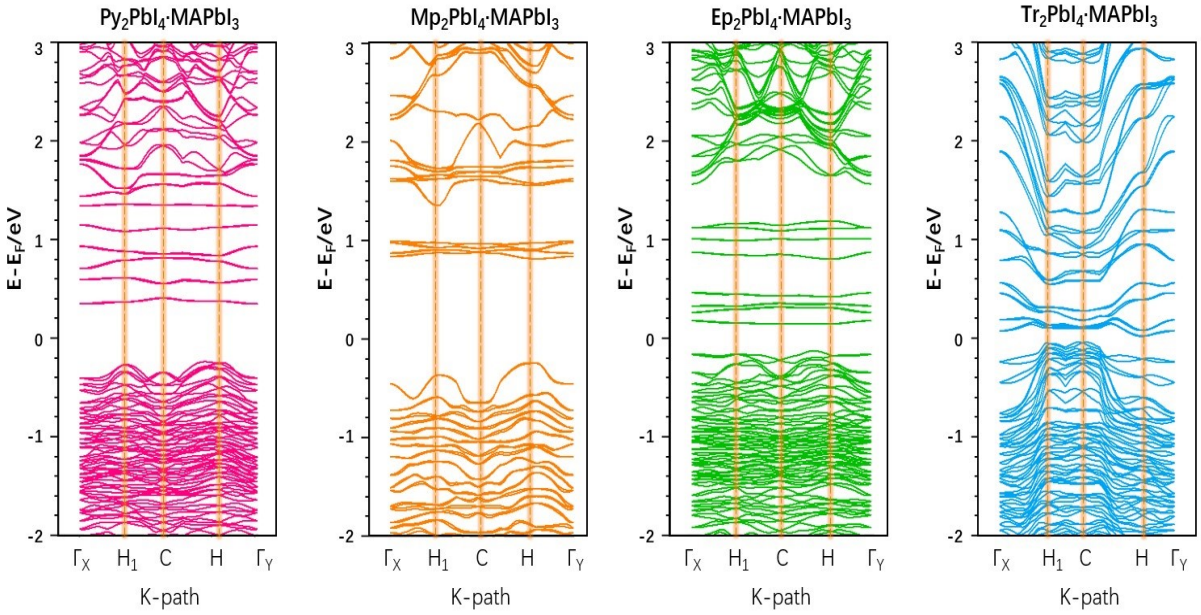

Figure S11: Computed band structure of QLD A_2PbI_4 • $MAPbI_3$ (*n*=1) structures with A = Py, Mp, Ep, Tr at PBE+SOC level of theory.

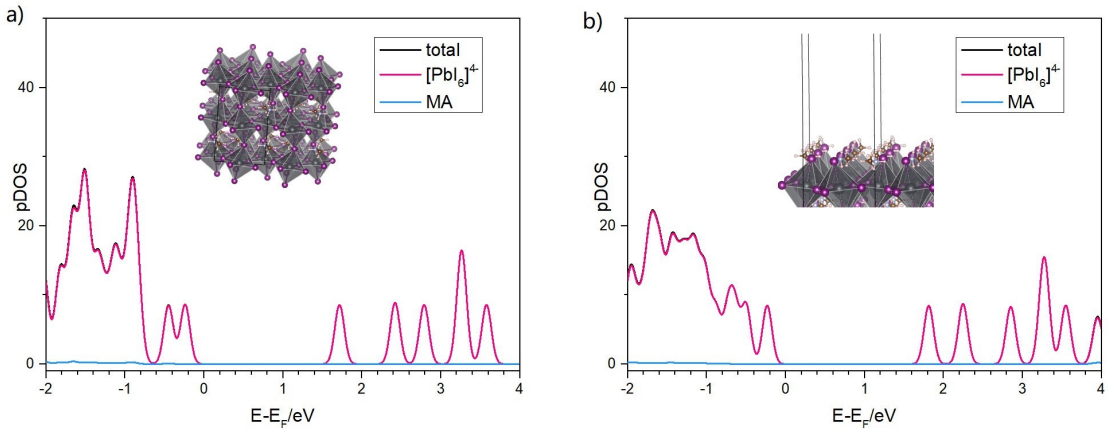

Figure S12: Computed projected density-of-state of $MAPbI_3$ with (a) bulk 3D structure and (b) QLD structure with 5 layers and a vacuum gap of 15 Å in the supercell.

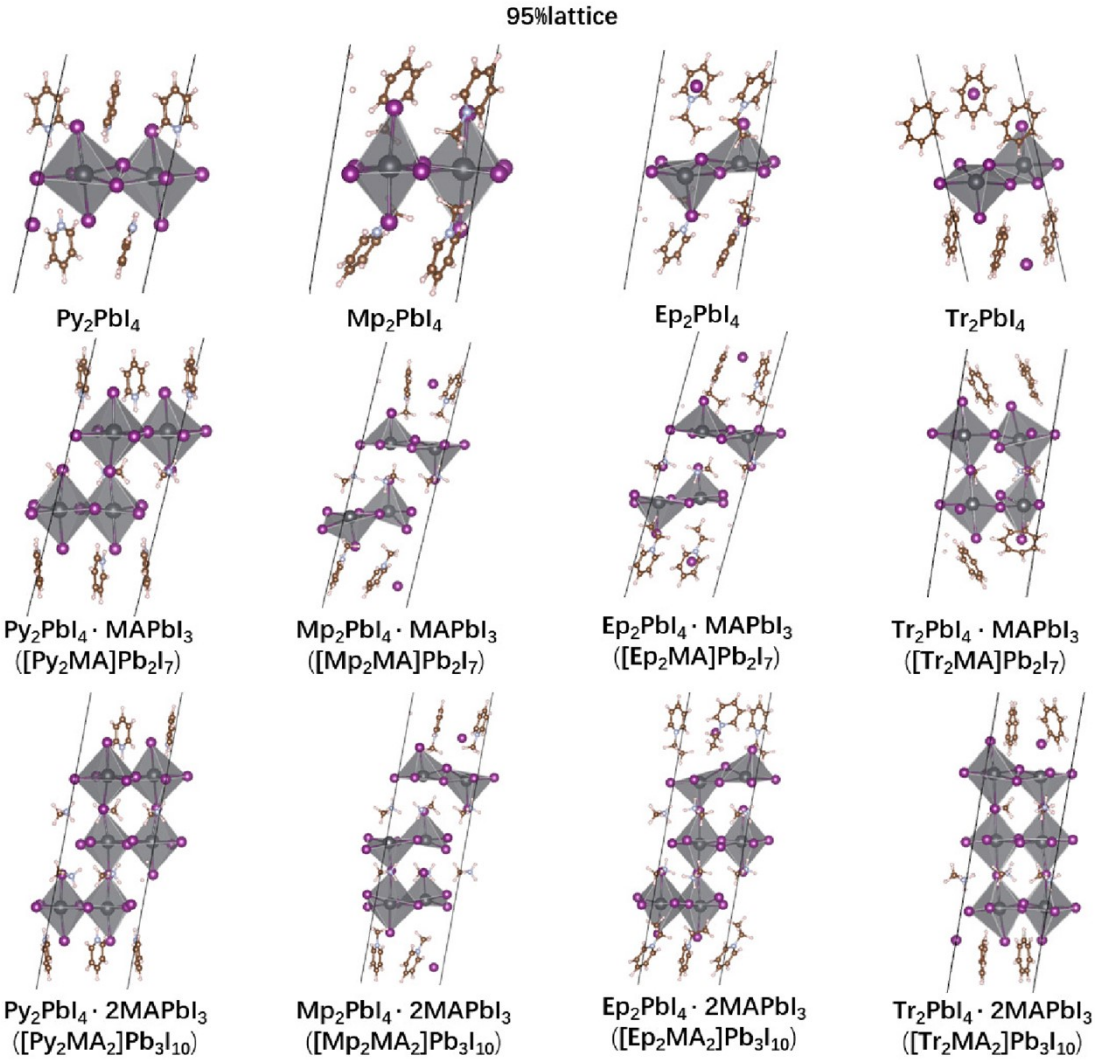

Figure S13: Optimized conformation (side view) of QLD perovskite structure $A_2PbI_4 \bullet n MAPbI_3$ *A* = Py, Mp, Ep, Tr with *n* = 0 to 2 and with 95% lattice constant.

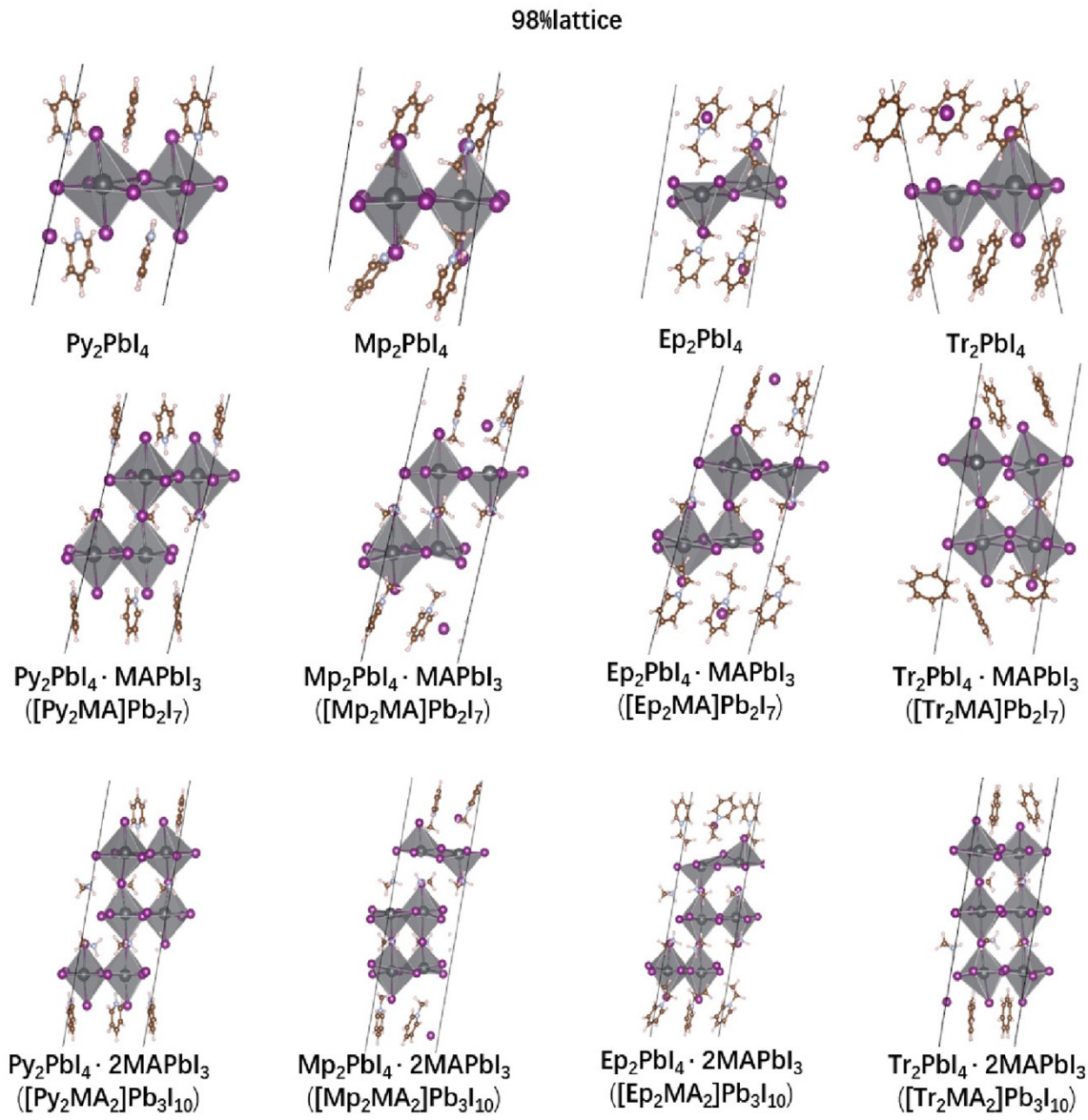

Figure S14: Optimized conformation (side view) of QLD perovskite structure $A_2PbI_4 \bullet n MAPbI_3$ *A* = Py, Mp, Ep, Tr with *n* = 0 to 2 and with 98% lattice constant.

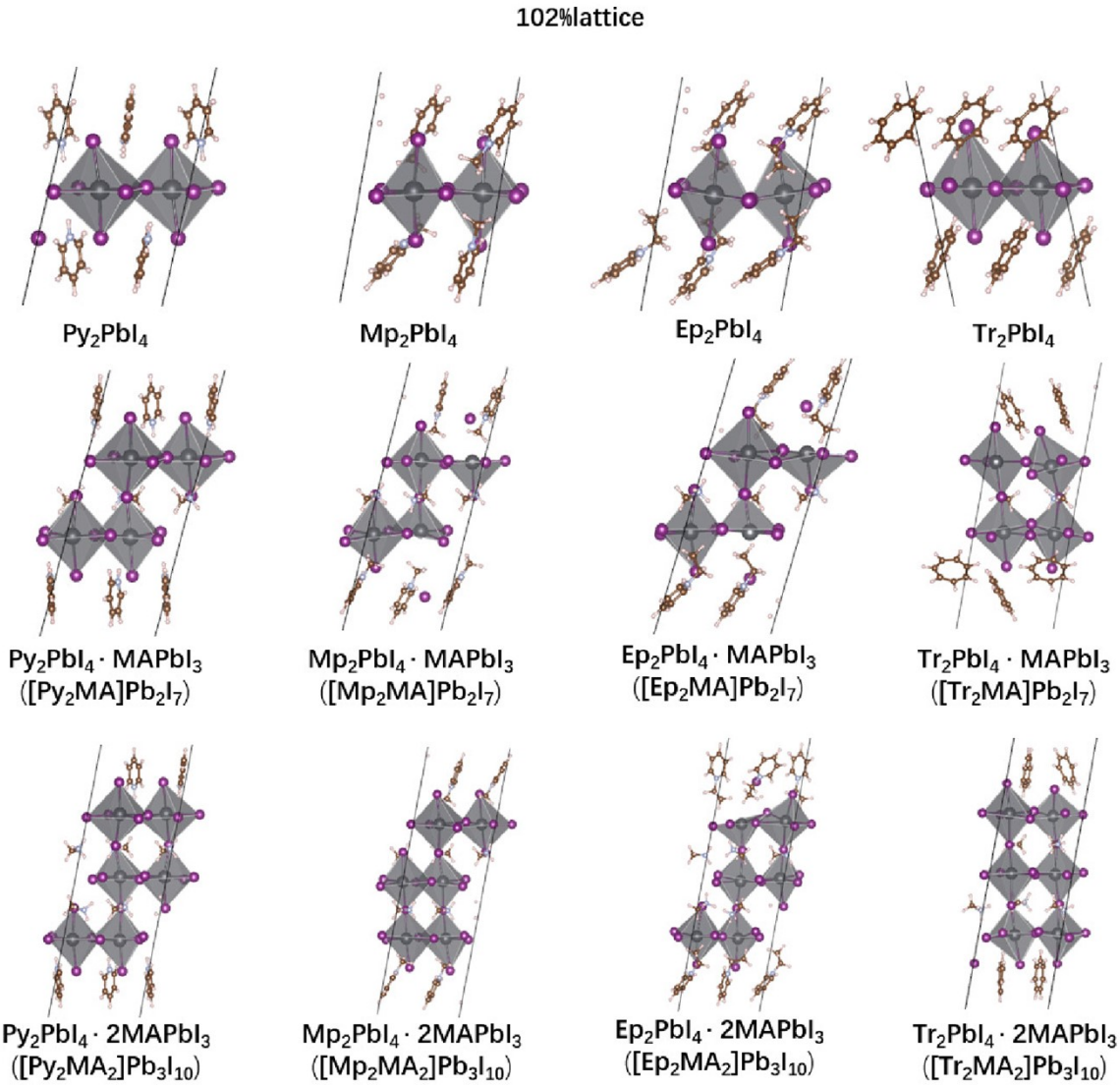

Figure S15: Optimized conformation (side view) of QLD perovskite structure $A_2PbI_4 \bullet n MAPbI_3$ *A* = Py, Mp, Ep, Tr with *n* = 0 to 2 and with 102% lattice constant.

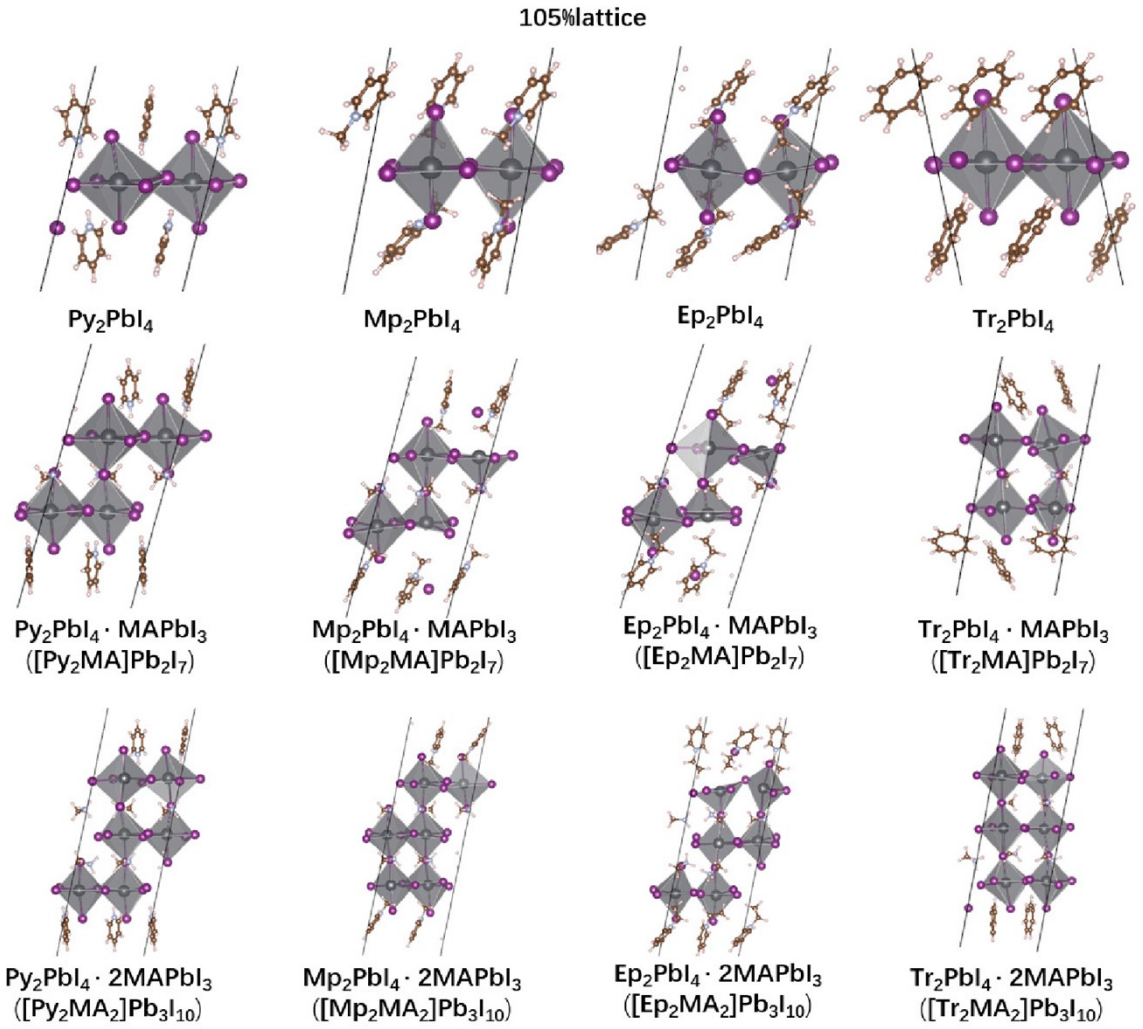

Figure S16: Optimized conformation (side view) of QLD perovskite structure $A_2PbI_4 \bullet n MAPbI_3$ *A* = Py, Mp, Ep, Tr with *n* = 0 to 2 and with 105% lattice constant.

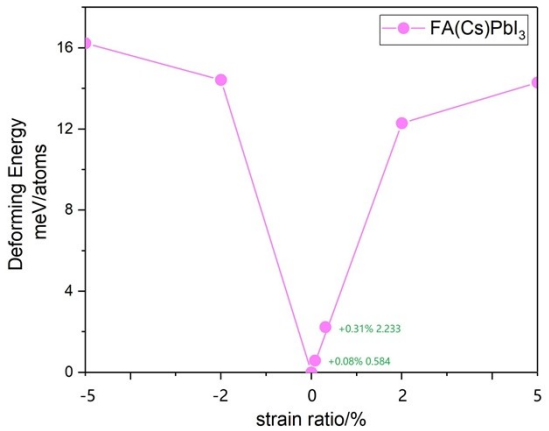

Figure S17: Computed deformation energy of $FACsPbI_3$ adopted in *Science* (2025) ***387***, 1069 at various strain ratios. The observed strain ratios in this article are marked out.

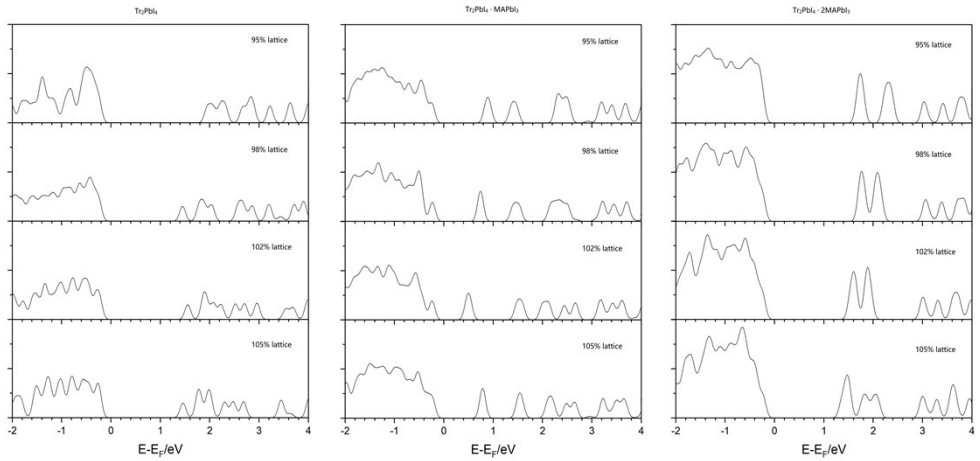

Figure S18: Computed density-of-state of $Tr_2PbI_4 \bullet n MAPbI_3$ with n = 0 to 2 and with various strain ratios.

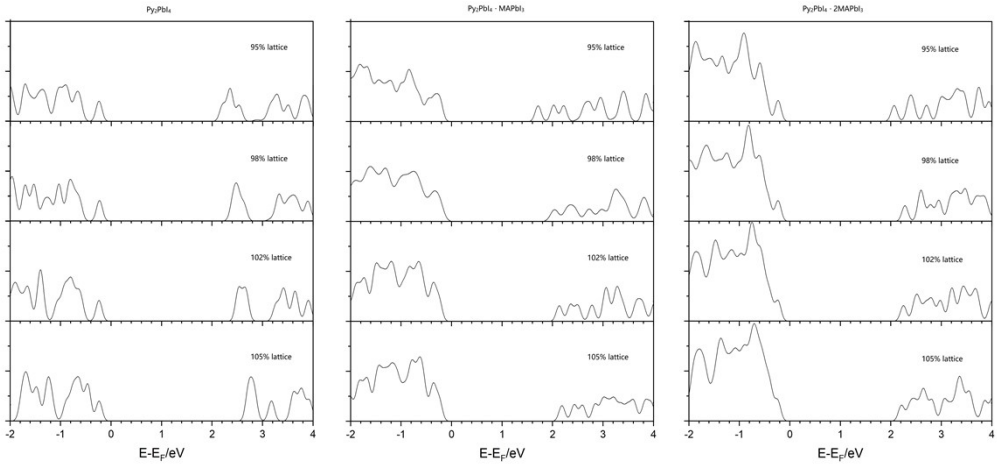

Figure S19: Computed density-of-state of $Py_2PbI_4 \bullet n MAPbI_3$ with n = 0 to 2 and with various strain ratios.

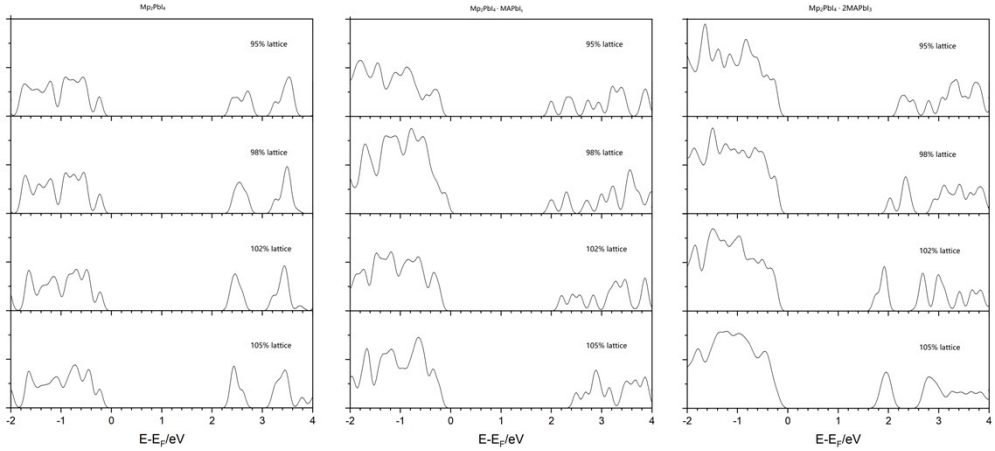

Figure S20: Computed density-of-state of $Mp_2PbI_4 \bullet n MAPbI_3$ with n = 0 to 2 and with various strain ratios.

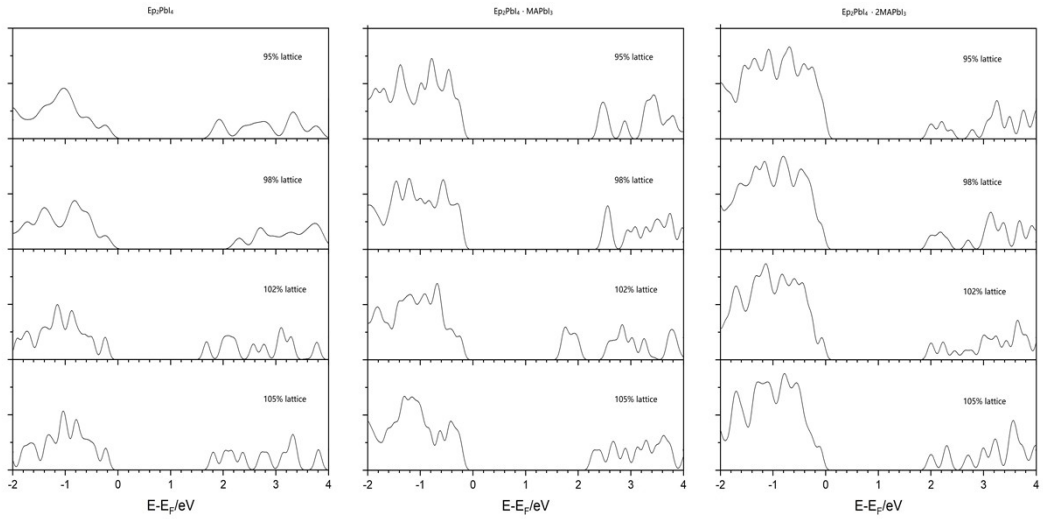

Figure S21: Computed density-of-state of Ep_2PbI_4 • *n*$MAPbI_3$ with *n* = 0 to 2 and with various strain ratios.

Table S1: Calculated geometric data of the optimized conformations of QLD CPAC perovskite A_2PbI_4 • *n* $MAPbI_3$ structures with *n* = 1 to 4

Spacer Category		Py	Mp	Ep	Tr
Max d_{Pb-I}/Å	n=1	3.45	4.14	3.76	3.62
	n=2	3.59	3.57	5.06	3.59
	n=3	3.31	3.46	6.20	3.84
	n=4	3.34	4.91	6.53	4.60
Average d_{Pb-I}/Å	n=1	3.30	3.28	3.31	3.29
	n=2	3.33	3.28	3.39	3.28
	n=3	3.25	3.29	3.21	3.24
	n=4	3.26	3.32	3.23	3.32
∠I-Pb-I/deg.	n=1	97.1	98.6	98.7	100.8
	n=2	98.6	96.48	116.34	116.63
	n=3	96.27	92.78	105.32	100.90
	n=4	100.17	106.71	111.79	101.96
d_{A-I}/ Å	n=1	3.54(2.86)	3.79	4.05	3.70(2.85)
	n=2	3.57(2.58)	3.36	3.78	3.69(2.85)
	n=3	3.43(2.39)	4.44	3.98	3.88(2.73)
	n=4	3.71(2.58)	4.28	3.99	3.90(2.82)
h/ Å	n=1	18.47	21.33	20.46	21.13
	n=2	24.16	26.79	27.21	26.75
	n=3	30.08	32.44	34.41	34.21
	n=4	38.73	40.24	40.37	39.76

Table S2: Calculated free energies (in eV) of the optimized conformations of QLD PAC perovskite A_2PbI_4 • *n* $MAPbI_3$ structures with *n* = 1 to 4

Spacer Category	Py	Mp	Ep	Tr
n = 1	-429.18	-494.74	-562.90	-481.22
n = 2	-531.75	-596.84	-664.76	-580.14
n = 3	-635.06	-700.86	-767.65	-680.97
n = 4	-738.59	-810.18	-870.13	-786.71

Table S3: Calculated formation energies (in meV/atoms) of the optimized conformations of QLD PAC perovskite A_2PbI_4 • *n* $MAPbI_3$ structures with *n* = 1 to 4

E_{form} follows the procession of $2AI + nMAI + (n+1)PbI_2 \rightarrow A_2PbI_4 \bullet n\ MAPbI_3$

Spacer Category	Py	Mp	Ep	Tr
n = 1	-96.2975	-70.8127	-134.9811	-60.0866
n = 2	-89.4830	-86.6542	-123.5330	-85.8491
n = 3	-125.5692	-108.9014	-123.4488	-87.1335
n = 4	-121.7064	-158.9325	-171.2101	-118.3771

Table S4: Calculated phase-segregation energies (in meV/atoms) of the optimized conformations of QLD PAC perovskite A_2PbI_4 • *n* $MAPbI_3$ structures with *n* = 1 to 4

E_{seg} follows the procession of $A_2PbI_4 \bullet n\ MAPbI_3 \rightarrow A_2PbI_4 + (MAPbI_3)_n$

Spacer Category	Py	Mp	Ep	Tr
n = 1	107.61638	10.53029	42.01524	58.81978
n = 2	91.77215	27.32841	52.62792	77.29956
n = 3	77.48840	23.14085	51.42889	88.77147
n = 4	74.69879	19.63120	53.18080	84.03623

Table S5: Calculated layer-segregation energies (in meV/atoms) of the optimized conformations of QLD PAC perovskite A_2PbI_4 • *n* $MAPbI_3$ structures with *n* = 1 to 4

E_{seg} follows the procession of A_2PbI_4 • n $MAPbI_3$ → A_2PbI_4 • (n-1) $MAPbI_3$ + $MAPbI_3$

Spacer Category	Py	Mp	Ep	Tr
n = 1	107.61638	10.53029	42.01524	58.81978
n = 2	17.30024	20.20840	20.12557	56.30756
n = 3	6.49481	0.30483	8.51692	27.72553
n = 4	3.51592	37.46852	9.76681	12.69667

Table S6: Calculated deformation energies (in meV/atoms) of the optimized conformations of QLD PAC perovskite A_2PbI_4 • *n* $MAPbI_3$ structures with *n* = 0 to 2 and with various strain ratios

$E_{dist.}$ follows the procession of A_2PbI_4 • n $MAPbI_{3\ pris.}$ → A_2PbI_4 • n $MAPbI_{3\ str.}$

thickness	strain ratio	Py	Mp	Ep	Tr
n=0	-5%	106.67	5.25	7.13	72.85
	-2%	68.73	1.14	0.45	62.31
	+2%	48.49	0.35	8.36	48.40
	+5%	39.38	17.17	6.11	39.47
n=1	-5%	53.88	27.53	36.16	28.55
	-2%	24.27	19.75	29.56	20.71
	+2%	14.88	14.67	33.50	16.40
	+5%	12.22	13.07	24.38	13.59
n=2	-5%	68.56	34.91	22.93	52.95
	-2%	32.27	24.03	14.62	31.61
	+2%	21.65	20.93	14.53	22.38
	+5%	21.85	13.25	13.89	20.22

Table S7: Calculated maximum surface Pb-I distance (in Å) of the optimized conformations of QLD PAC perovskite A_2PbI_4 • *n* $MAPbI_3$ structures with *n* = 0 to 2 and with various strain ratios

thickness	strain ratio	Py	Mp	Ep	Tr
n=0	-5%	3.37	3.59	7.27	7.39
	-2%	3.35	3.48	6.39	5.31
	+2%	3.34	3.48	3.59	3.92
	+5%	3.46	3.51	3.56	3.73
n=1	-5%	3.30	6.70	8.08	3.63
	-2%	3.38	4.17	7.90	3.54
	+2%	3.52	4.17	3.91	3.63
	+5%	3.60	4.48	6.72	3.60
n=2	-5%	3.41	4.89	5.60	3.88
	-2%	3.51	4.33	5.31	3.58
	+2%	3.66	3.47	5.05	3.61
	+5%	3.56	3.61	5.18	3.67

Table S8: Calculated band gaps (in eV) of the optimized conformations of QLD PAC perovskite A_2PbI_4 • *n* $MAPbI_3$ structures with *n* = 0 to 2 and with various strain ratios

At HSE06+SOC level of theory with an XC_{HF} fraction of 0.38

thickness	strain ratio	Py	Mp	Ep	Tr
n=0	-5%	1.99	2.17	1.66	1.21
	-2%	2.19	2.26	1.92	1.23
	+2%	2.52	2.37	1.56	1.78
	+5%	2.54	2.43	1.51	1.69
n=1	-5%	1.50	1.78	2.24	0.62
	-2%	1.76	1.84	2.28	0.65
	+2%	2.01	2.16	1.50	0.91
	+5%	2.04	2.27	2.07	1.00
n=2	-5%	1.85	2.02	2.00	1.47
	-2%	2.06	1.85	1.78	1.50
	+2%	2.08	1.53	1.45	1.36
	+5%	1.99	1.58	1.53	1.59