Supplementary Information

Strong Thickness-Dependent Quantum Confinement in All-inorganic Perovskite Cs_2PbI_4 with Ruddlesden-Popper Structure

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Figure S1. The tested atoms structure without dangling bonds for monolayer perovskite $CsPbI_4$.

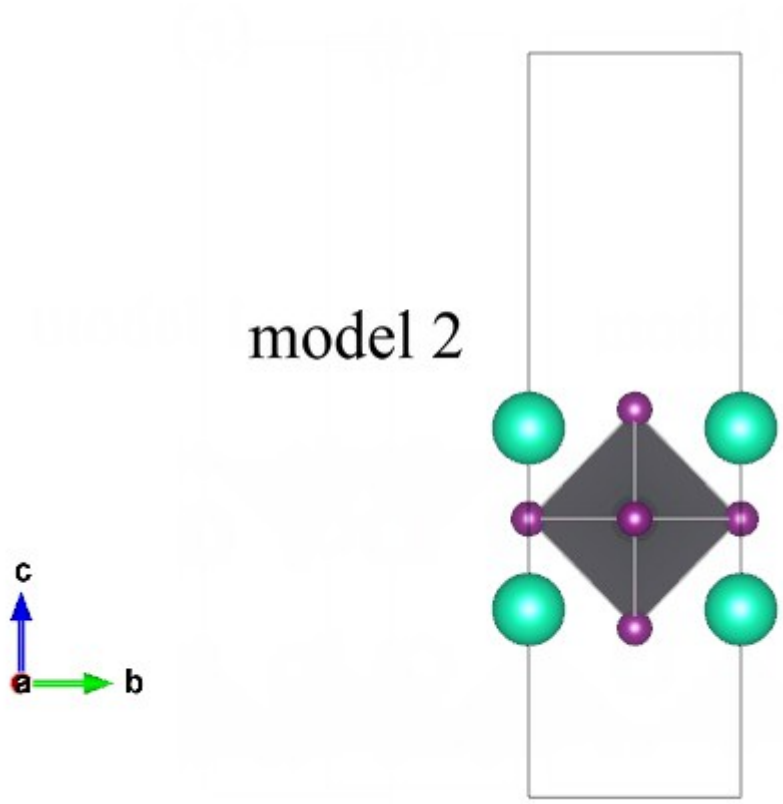

Figure S2. The band structure of model in our manuscript with PBE (a) and PBE+SOC (b). The band structure of model 2 without dangling bond with PBE (c) and PBE+SOC (d) for one layer perovskite $CsPbI_4$.

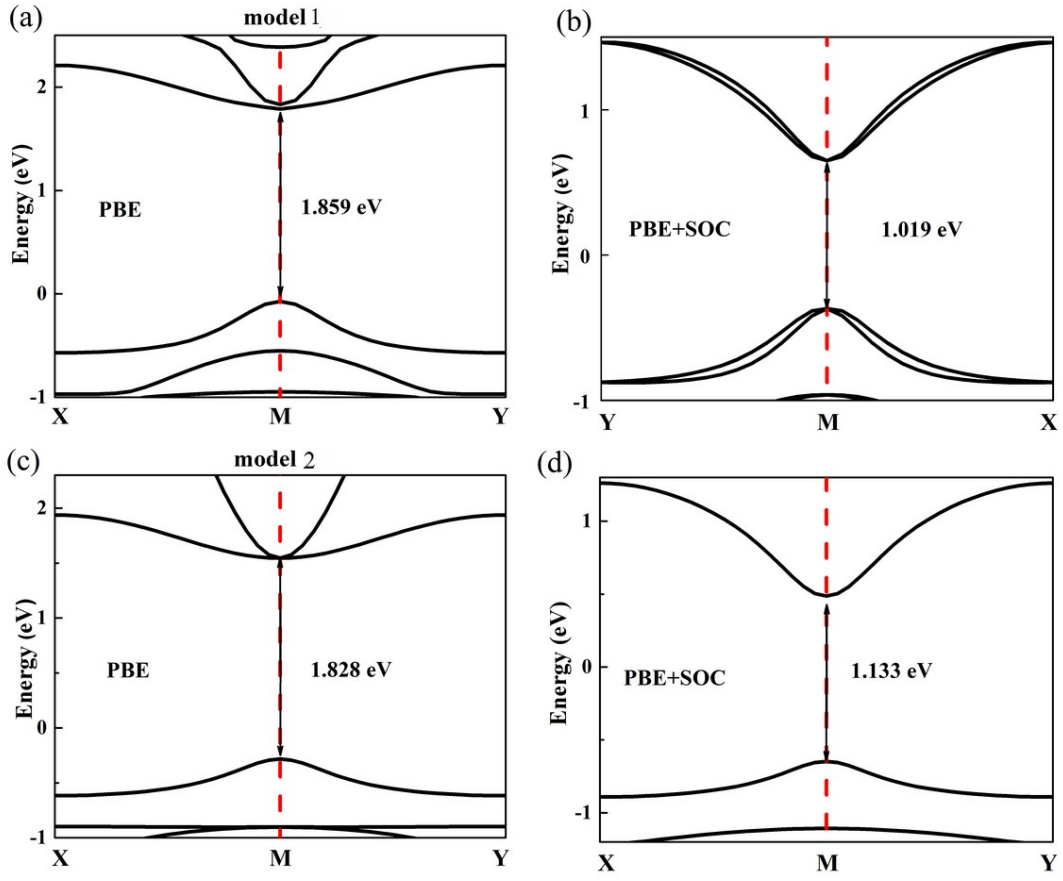

Figure S3. The calculated band structures of all-inorganic 2D layered RP perovskite Cs_2PbI_4 (L=4, 5) by PBE.

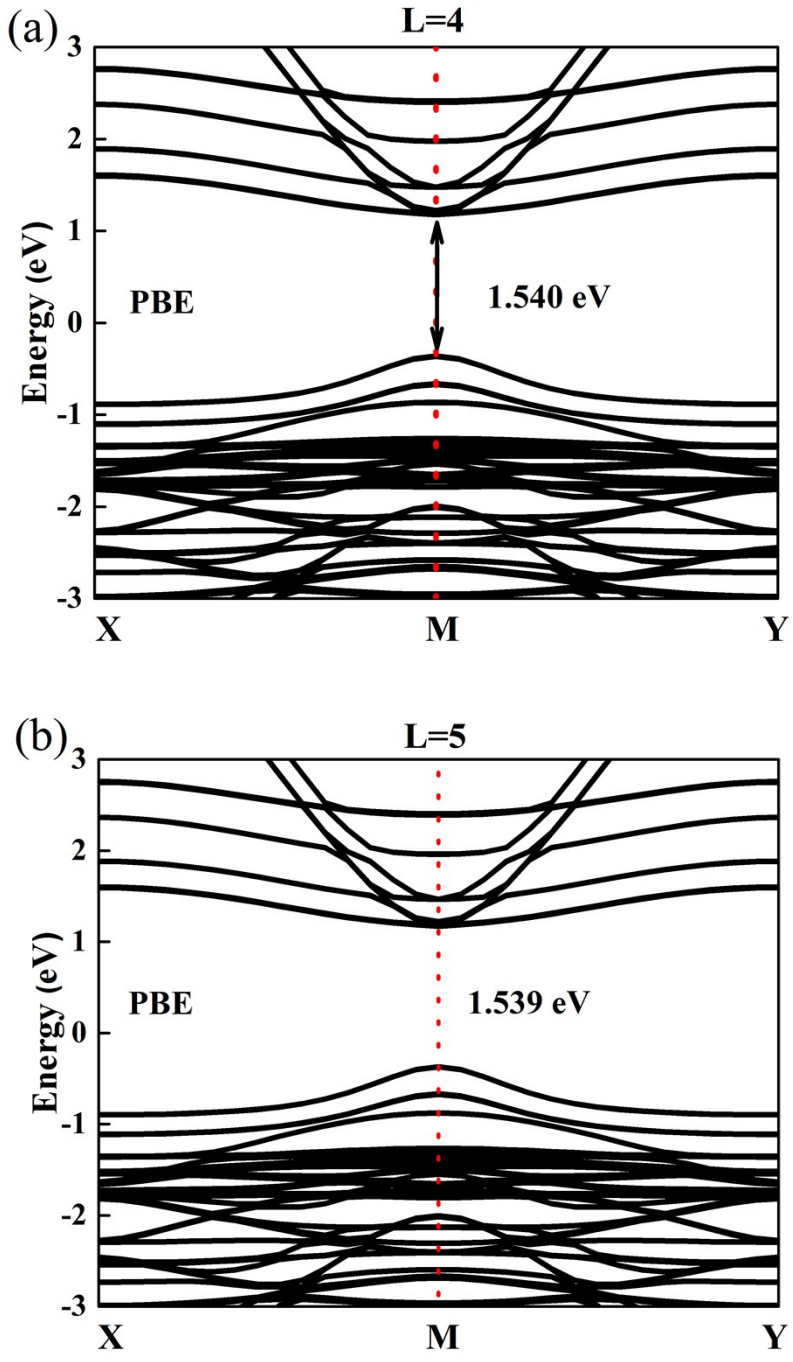

Figure S4. The band structrures with LDA functionals for one layer (a), two layers (b) and three layers (c). The values of bandgaps are shown in the figure.

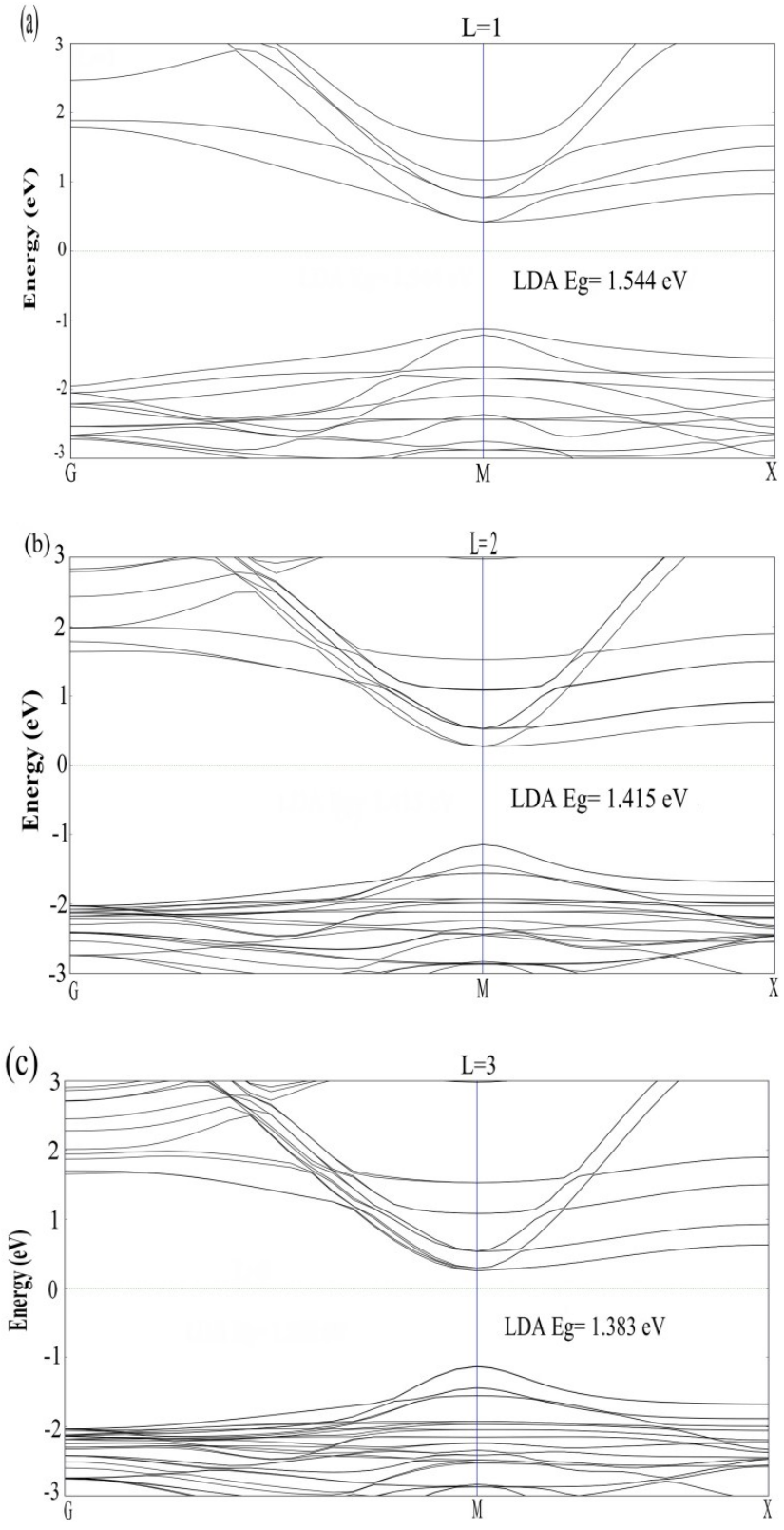

Figure S5. (a) The band edge energy shift of VB and CB with respect to the lattice dilation along the a_0 direction for the bilayer perovskite. Solid lines represent the linear fit, which defines DP constants. (b) The total energy as a function of lattice deformation along the a_0 directions. Solid lines are the parabola fittings, which give elastic constant. I_0 is the lattice constant in the transport direction, and $\Delta I=I_0-I$ is the deformation of I_0.

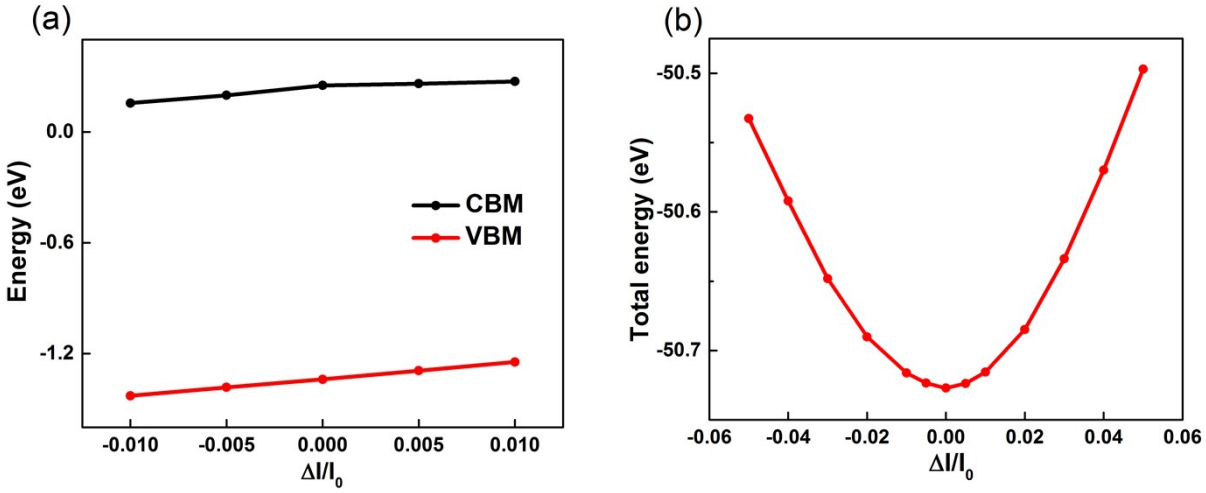

Figure S6. (a) The band edge positions of VB and CB with respect to the lattice dilation along the a_0 direction for the trilayer perovskite. Solid lines represent the linear fit, which defines DP constants. (b) The total energy as a function of lattice deformation along the a_0 direction. Solid lines are the parabola fittings, which give elastic constant.

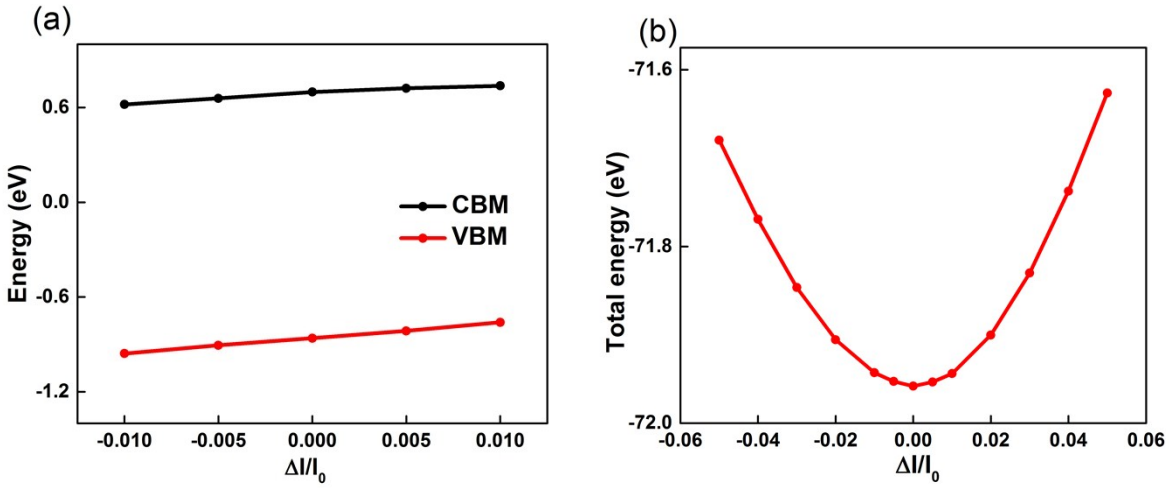

Table S1. The bandgap values of perovskite Cs_2PbI_4 with different thicknesses calculated by PBE, PBE+SOC, LDA and HSE+SOC.

Layer	PBE (eV)	PBE+SOC (eV)	LDA (eV)	HSE+SOC (eV)
1L	1.859	1.019	1.544	1.544
2L	1.592	0.823	1.415	1.333
3L	1.558	0.804	1.383	1.310
4L	1.540			
5L	1.539			