

Supplementary materials

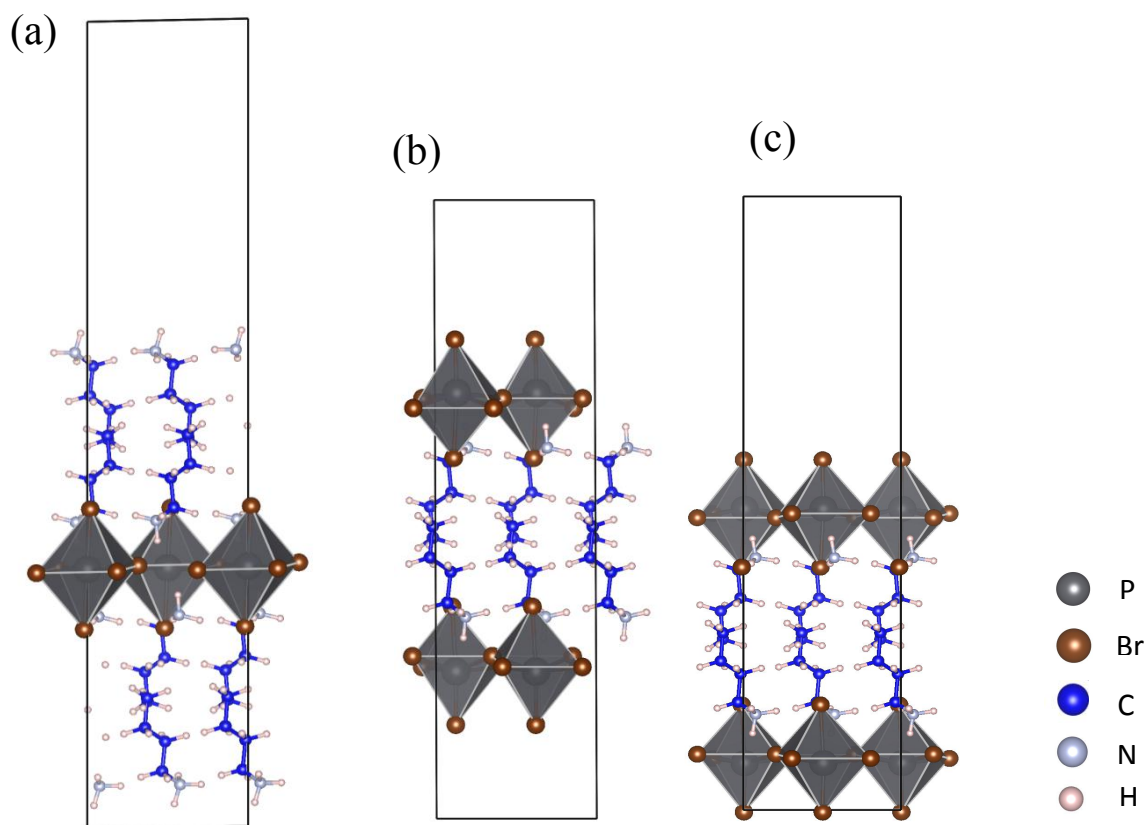


Fig.1 The optimized atomic arrangement of two dimensional single layered hybrid perovskite $(C_4H_9NH_3)_2PbBr_4$, (a) the 2D structure with plane lattice parameters ($a=8.12 \text{ \AA}$, $b=8.01 \text{ \AA}$), in which the middle inorganic layer is clamped by bilateral organic layers. (b) and (c) are the 2D structure with plane lattice parameters ($a=7.96 \text{ \AA}$, $b=7.81 \text{ \AA}$), ($a=7.99 \text{ \AA}$, $b=7.83 \text{ \AA}$) respectively, in which the middle organic groups are clamped by bilateral inorganic octahedron framework. The calculated results show the different calculated 2D model will lead to the contradiction conclusion (the cell expanding or shrinking upon the 3D-to-2D structure transformation).

note: the calculated plane lattice parameters of corresponding 3D phase are $a=8.09 \text{ \AA}$, $b=7.93 \text{ \AA}$ respectively

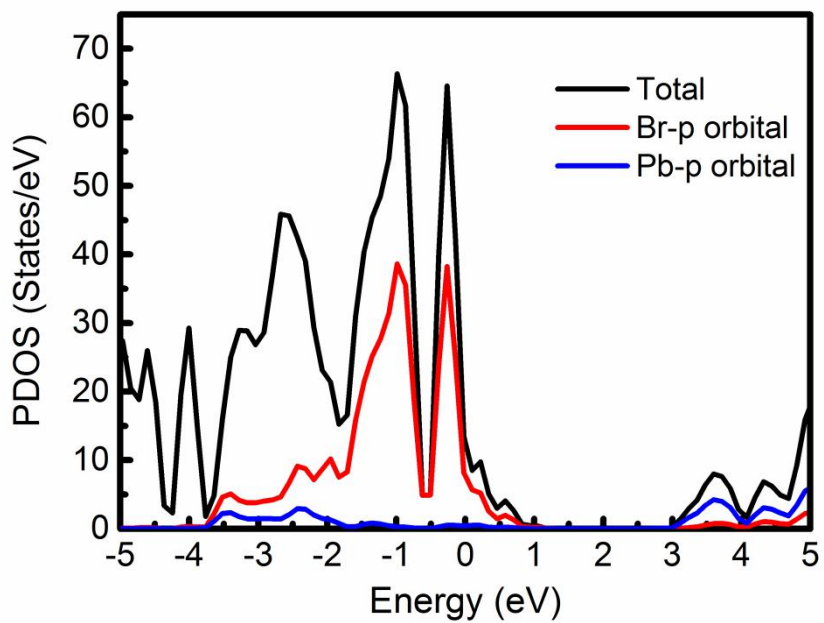


Fig.2 The projected density of states (PDOS) of two dimensional single layered hybrid perovskites $(C_4H_9NH_3)_2PbBr_4$, which denotes the leading optical transition from the occupied p-orbitals of Br to unoccupied one in Pb.