Supplementary Information:

High-performance photovoltaic device for the 2D all-inorganic Ruddlesen-Popper perovskite heterostructure $Cs_2PbI_2Cl_2/MAPbI_3$

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Table S1. The calculated lattice parameters of 2D $Cs_2PbI_2Cl_2$ monolayer, 3D $MAPbI_3$ slab, and 2D/3D heterostructure. The a and b stand for the lattice direction.

	2D $Cs_2PbI_2Cl_2$ (Å)	**3D $MAPbI_3$ (Å)**	**2D/3D Her. (Å)**
a	5.64	8.80	8.37
b	5.64	8.80	8.37

Table S2. Calculated band gaps of 2D monolayer/3D MAI terminated and 2D trilayer/3D MAI terminated heterostructures using the PBE method , respectively.

	E_g (eV)
2D monolayer/3D MAI Her.	1.20
2D trilayer/3D MAI Her.	1.19

Table S3. The work function before and after compression of 2D $Cs_2PbI_2Cl_2$., 3D MAI-terminated and 3D PbI-terminated.

	2D (eV)	**3D-MAI (eV)**	**3D-PbI (eV)**
Before compression	4.88	4.09	5.21
After compression	4.97	3.57	5.49

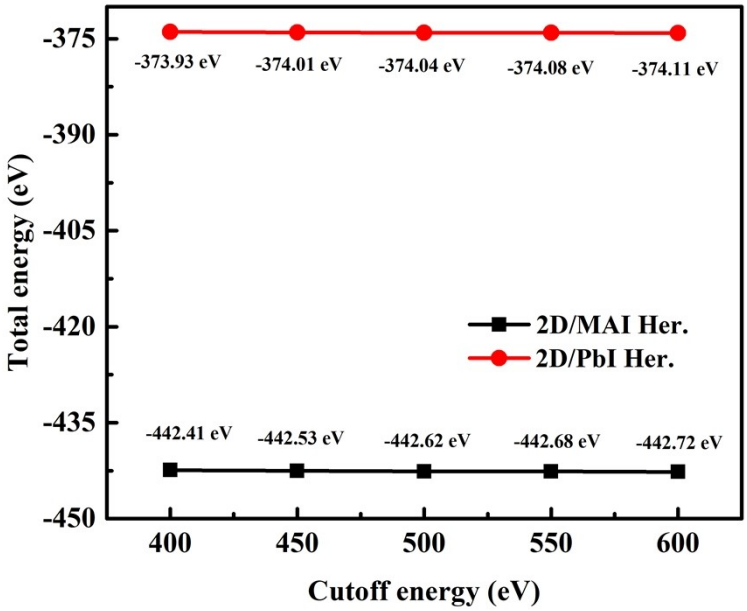

Fig. S1 The total energy of 2D/MAI and 2D/PbI heterostructures for various energy cutoffs.

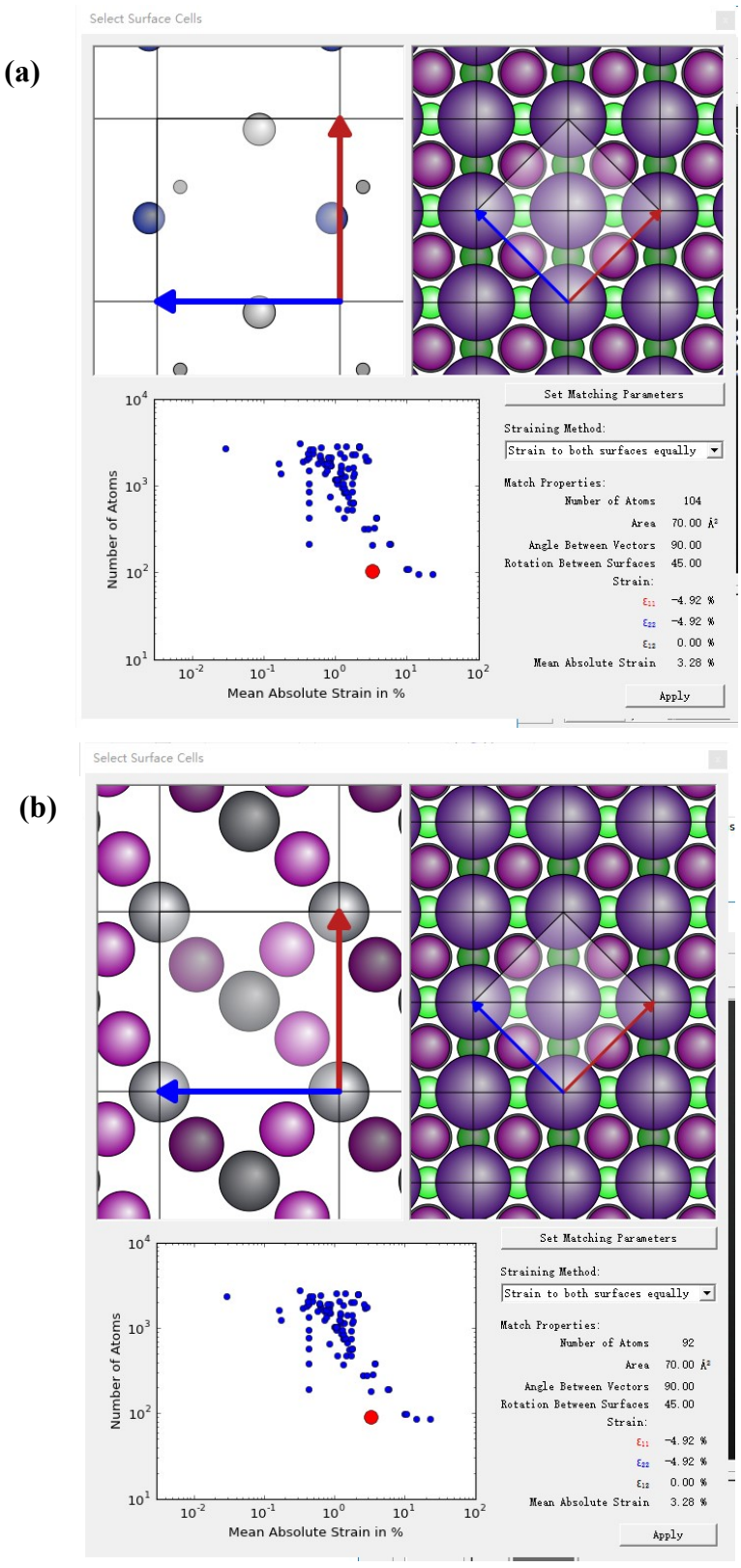

Fig. S2 The different match between the 2D $Cs_2PbI_2Cl_2$. monolayer and (a) 3D MAI-teminated, (b) 3D PbI-terminated. The red dot present the supercell dimension with the minimum number of atoms and mean absolute strain.

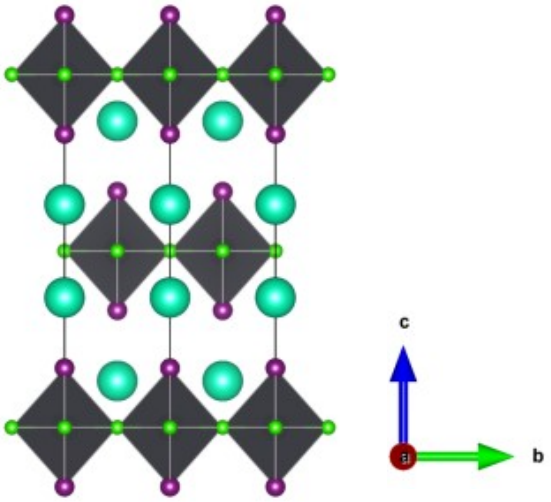

Fig. S3 The periodic structure of 2D $Cs_2PbI_2Cl_2$.

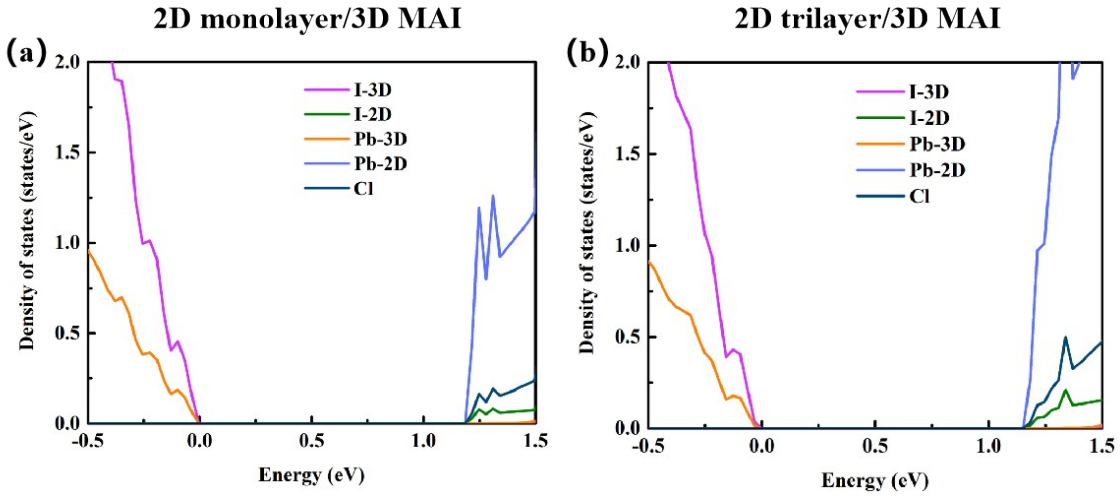

Fig. S4 Calculated projected density of states (PDOS) of (a) 2D monolayer/3D MAI terminated and (b) 2D trilayer/3D MAI terminated heterostructures using the PBE method , where I-3D, I-2D, Pb-3D and Pb-2D represent I atom in 3D $MAPbI_3$, I atom in 2D $Cs_2PbI_2Cl_2$, Pb atom in 3D $MAPbI_3$ and Pb atom in 2D $Cs_2PbI_2Cl_2$, repectively.

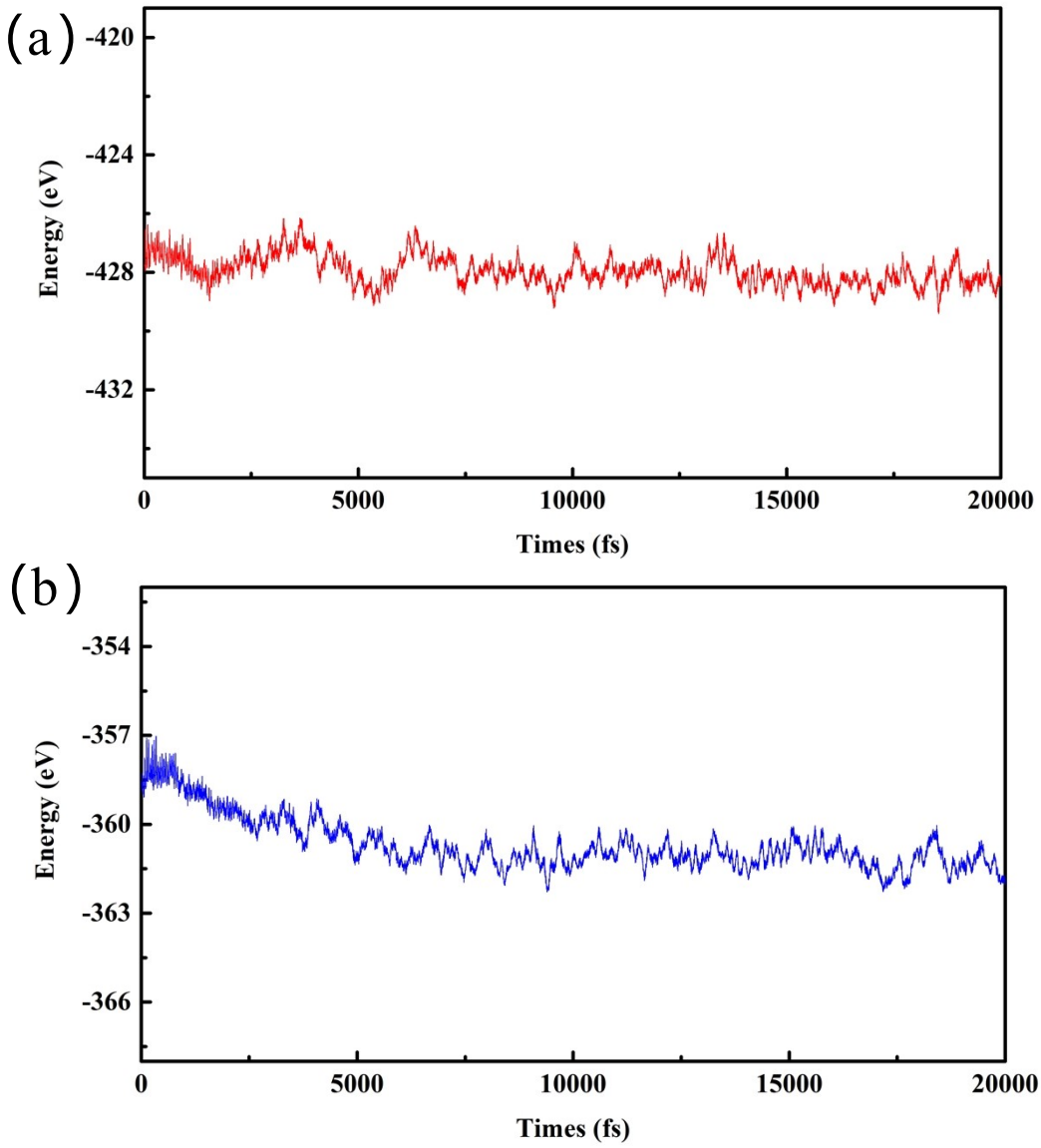

FIG. S5 Variations of the energy against the time for AIMD simulations of (a) 2D/MAI and (b) 2D/PbI heterostructures. The simulation is run under 298.15 K and the time step is 1 fs.

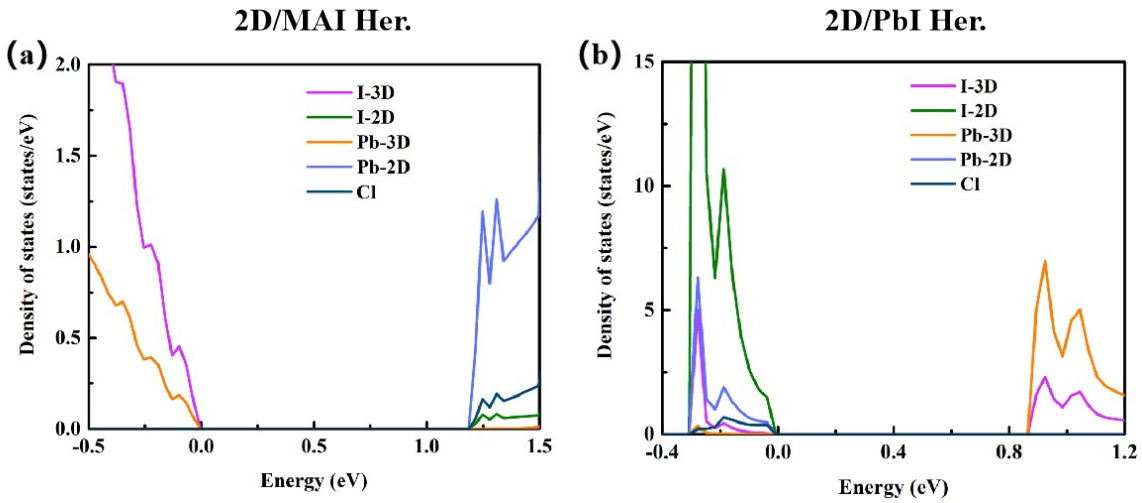

Fig. S6 Calculated projected density of states (PDOS) of (a) 2D/MAI and (b) 2D/PbI heterostructures using the PBE method.

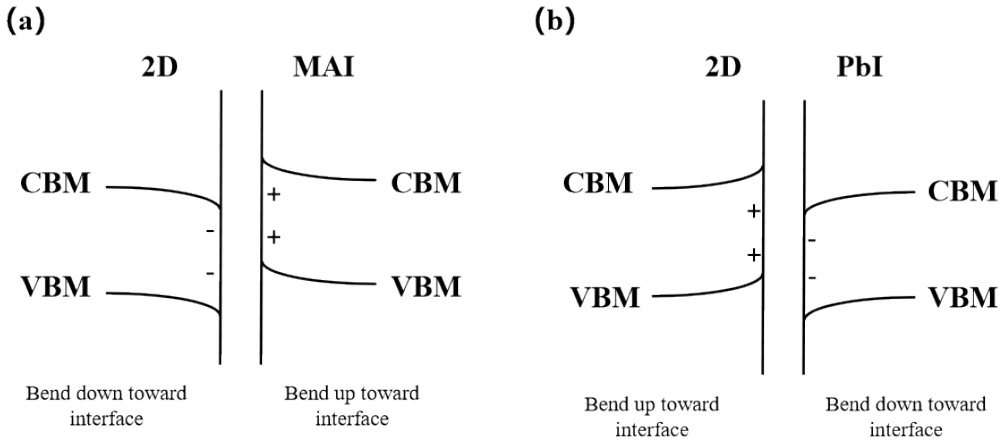

Fig. S7 The band bending diagrams of (a) 2D/MAI and (b) 2D/PbI heterostructures, respectively.