Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information for:

Theoretical Design of New Two-Dimensional Topological Insulator Family

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I. The parity analysis

In this work, we employ the package Quantum Espresso to calculate the band parities at TRIM points by using the relativistic ultrosoft pseudopotentials. The four TRIM points at the Brillouin zone for two-dimensional materials would be (0, 0), (0, 0.5), (0.5, 0), (0.5, 0.5).

Table S1 shows the band parity analysis of the other three systems: hydrogenated monolayer Pb_2InSb, Pb_2GaSb, Pb_2GaBi. One can observe that these are similar to those in hydrogenated monolayer Pb_2InBi, which is presented in the paper. The nontrivial topological invariant Z_2 can be observed, indicating the topological phase.

Table S1 Parities of twenty occupied spin-degenerate bands at four TRIM points for hydrogenated monolayer Pb_2InSb, Pb_2GaSb, Pb_2GaBi.

Pb_2GaSb	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	+	-	-	**(-)**
	-	+	+	-	-	+	+	-	-	+	+	-	+	-	+	-	+	-	+	-	**(+)**
	-	+	+	-	-	+	+	-	-	+	+	-	+	-	+	-	+	-	+	-	**(+)**
	-	+	+	-	+	-	+	-	-	+	-	+	-	+	-	+	-	+	+	-	**(+)**
Pb_2GaBi	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	+	-	-	**(-)**
	-	+	+	-	-	+	+	-	-	+	+	-	+	-	+	-	+	-	+	-	**(+)**
	-	+	+	-	-	+	+	-	-	+	+	-	+	-	+	-	+	-	+	-	**(+)**
	-	+	+	-	-	+	+	-	-	+	+	-	-	+	-	+	+	-	+	-	**(+)**
Pb_2InSb	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	+	-	-	**(-)**
	+	-	-	+	+	-	-	+	+	-	-	+	-	+	-	+	-	+	-	+	**(+)**
	+	-	-	+	+	-	-	+	+	-	-	+	-	+	-	+	-	+	-	+	**(+)**
	+	-	-	+	+	-	+	-	-	+	+	-	-	+	-	+	-	+	+	-	**(+)**

Table S2 shows the band parity analysis of hydrogenated monolayer Pb_2GaBi with strain deformation, while the negative value means the compressive stress and the positive ones denote the tensile stress. As the value increase from -8% to 8%, which means from compressive to tensile stress, a transition from trivial phase to nontrivial phase can be observed while the critical point stands at around -4%.

Table S2 The strain dependence of band parities at Gamma point of hydrogenated monolayer Pb_2GaBi.

Strain (%)	Parities of twenty occupied spin-degenerate bands at Gamma point																					Z_2
-8	+	-	+	-	-	+	+	-	+	-	+	+	-	+	-	-	-	+	-	+	**(+)**	**trivial**
-6	+	-	+	-	-	+	+	-	+	-	+	-	+	+	-	-	-	+	-	+	**(+)**	**trivial**
-4	+	-	+	-	-	+	+	-	+	-	+	+	-	+	-	-	-	+	-	-	**(-)**	**nontrivial**
-2	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	+	-	-	**(-)**	**nontrivial**
0	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	+	-	-	**(-)**	**nontrivial**
2	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	+	-	-	**(-)**	**nontrivial**
4	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	+	-	-	**(-)**	**nontrivial**
6	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	+	-	-	**(-)**	**nontrivial**
8	+	-	-	+	-	+	+	-	+	-	+	+	-	-	+	-	-	-	+	-	**(-)**	**nontrivial**

II. The strain dependence of electronic band structures

Here we present the electronic band structures of the other three systems as influenced by strain effect, at the range of [-8%, 8%], while the negative value means the compressive stress and the positive ones denote the tensile stress.

The case of hydrogenated monolayer Pb_2InBi is similar with that of hydrogenated monolayer Pb_2GaBi, as demonstrated in the paper. From -8% to 8% strain deformation, a metal-semiconductor transition can be observed, while the critical point stands at around -4%.

For hydrogenated monolayer Pb_2GaSb and Pb_2InSb, the semiconductive properties are kept with strain deformation. The value of bandgap keeps decreasing from -8% to 0% (compressive condition), and then increases until a stable value in the tensile range of [0%, 8%].

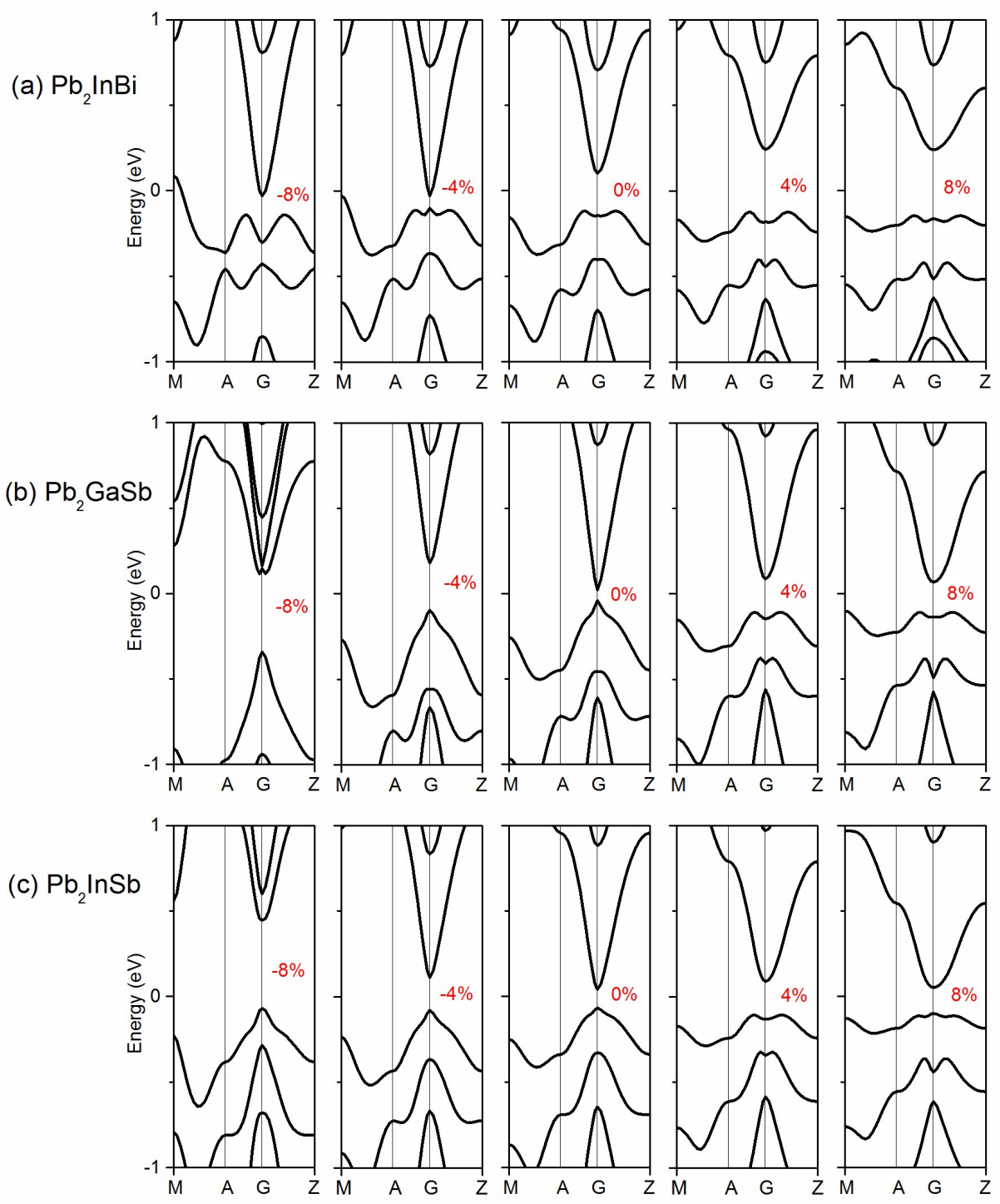

Fig. S1 The electronic band structure of hydrogenated (a) Pb_2InBi; (b) Pb_2GaSb and (c) Pb_2InSb with strain deformation in the range of [-8%, 8%] along the high-symmetry point "M(-0.5, -0.5, 0.5)-A(-0.5, 0, 0)-G(0, 0, 0)-Z(0, -0.5, 0.5)".

III. The atomistic structures at high temperature

Ab initio molecular dynamics method is adopted to calculate the atomistic structures of the new proposed family at high temperature. From Figure S2, it can be observed that this new family can preserve the crystal structure at room temperature. However, as temperature increase to 700K, the instability is induced due to thermal excitations, which is similar with the case shown in the previous work.[1]

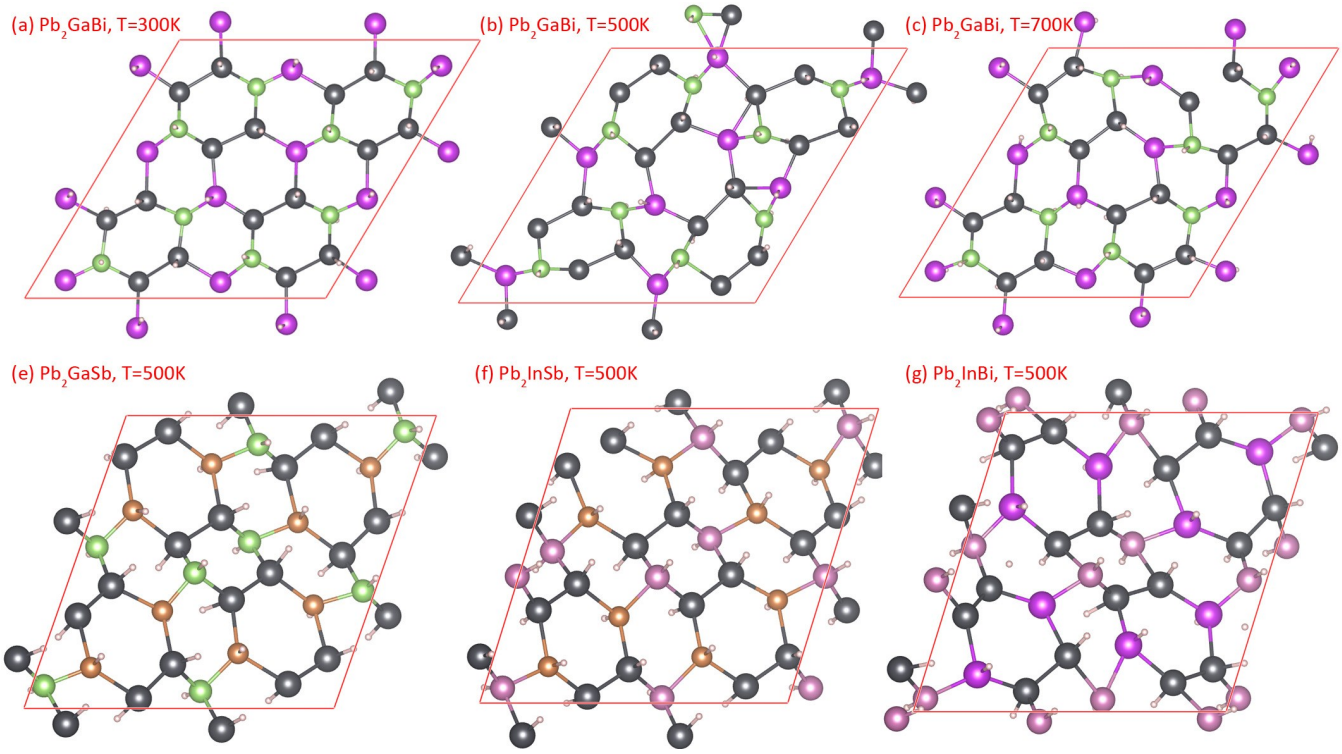

Fig. S2 The atomistic structures calculated by ab initio MD methods. (a)~(c) denote the hydrogenated monolayer Pb_2GaBi structures at 300K, 500K and 700K. The structures at 500K for hydrogenated monolayer Pb_2GaSb, Pb_2InSb and Pb_2InBi are shown in (e)~(g), respectively.

References

1. Aktürk, E.; Aktürk, O. Ü.; Ciraci, S. *Phys. Rev. B* **2016,** 94, (1), 014115.