Defects of monolayer PbI_2: A computational study

Han Wang[a,#] Xiao Wang[b,#,*], Da Li[a,*]

[a]State Key Laboratory of Superhard Materials, College of Physics, Jilin University, Changchun, 130012, P. R. China

[b]Shenzhen Key Laboratory of Nanobiomechanics, Shenzhen Institute of Advanced Technology, Chinese Academy of Sciences, Shenzhen 518055, P. R. China

Table S1. Comparison of the band gap by different methods.

Methods	PBE	PBE+SOC	HSE	HSE+SOC
Bandgap (eV)	2.52 eV	1.86 eV	3.29 eV	2.60 eV

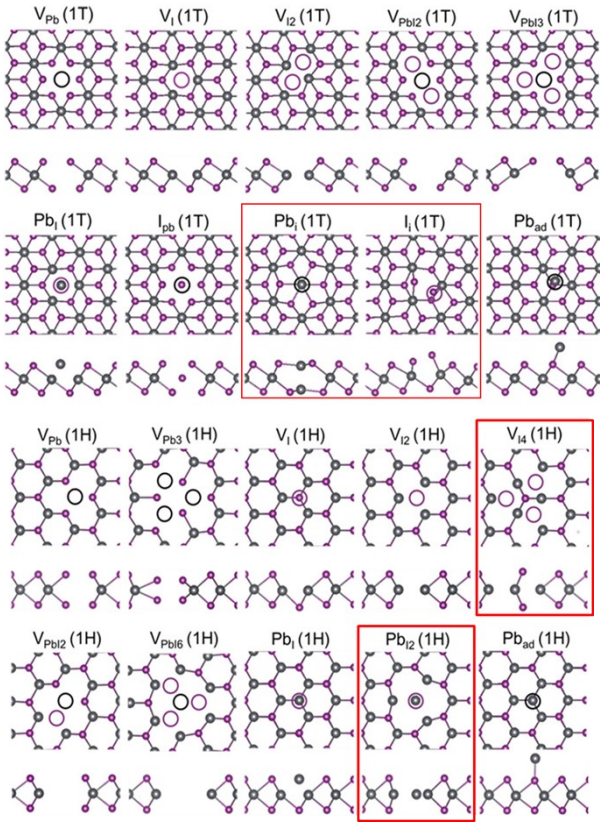

Figure S1. Optimized geometries of defects at the neutral state in (a) 1T and (b) 1H PbI_2 phases: vacancies, antisites, interstitials, and adatoms are all shown. The Pb and I atoms are colored in black and purple, respectively. Positions of vacancy, interstitial, antisite or adatom defects in the unit cell are marked in solid circle. The reconstructed structures are labelled with red box.

Table S2. The cohesive energies of the defects of PbI_2 in both 1T and 1H phases.

1T		1H	
Structure	Cohesive energy (eV)	Structure	Cohesive energy (eV)
perfect	2.63	perfect	2.56
V_{Pb}	2.59	V_{Pb}	2.52
V_I	2.62	V_{Pb3}	2.46
V_{I2}	2.61	V_I	2.55
V_{PbI2}	2.62	V_{I2}	2.55
V_{PbI3}	2.61	V_{I4}	2.54
Pb_I	2.62	V_{PbI2}	2.54
I_{Pb}	2.58	V_{PbI6}	2.52
Pb_i	2.62	Pb_I	2.55
I_i	2.61	Pb_{I2}	2.55
Pb_{ad}	2.61	Pb_{ad}	2.54

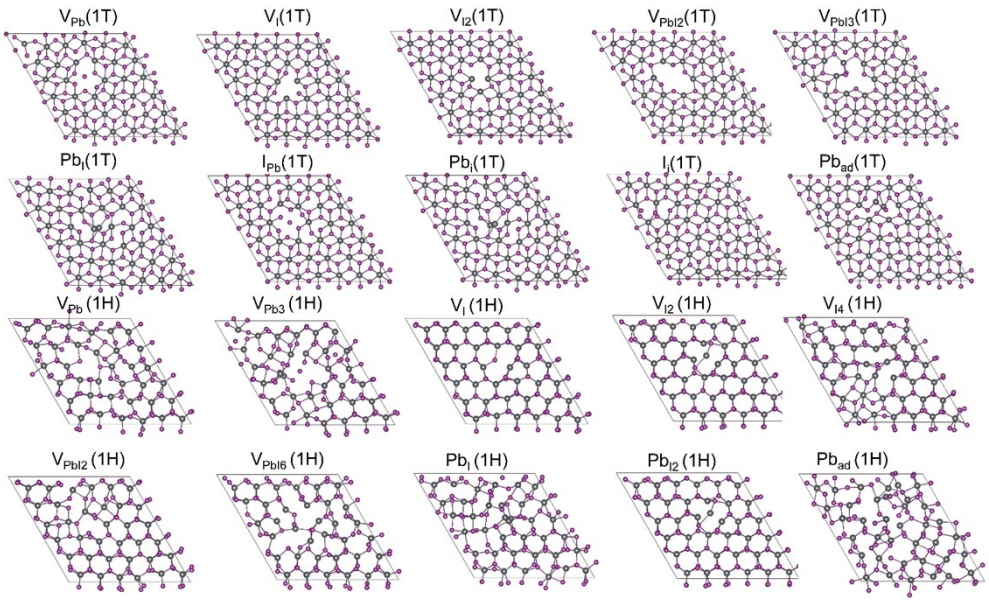

Figure S2. Snapshots for the equilibrium structures of various point defects in 1T and1H phase PbI_2 under room-temperature with time running for 5 ps.

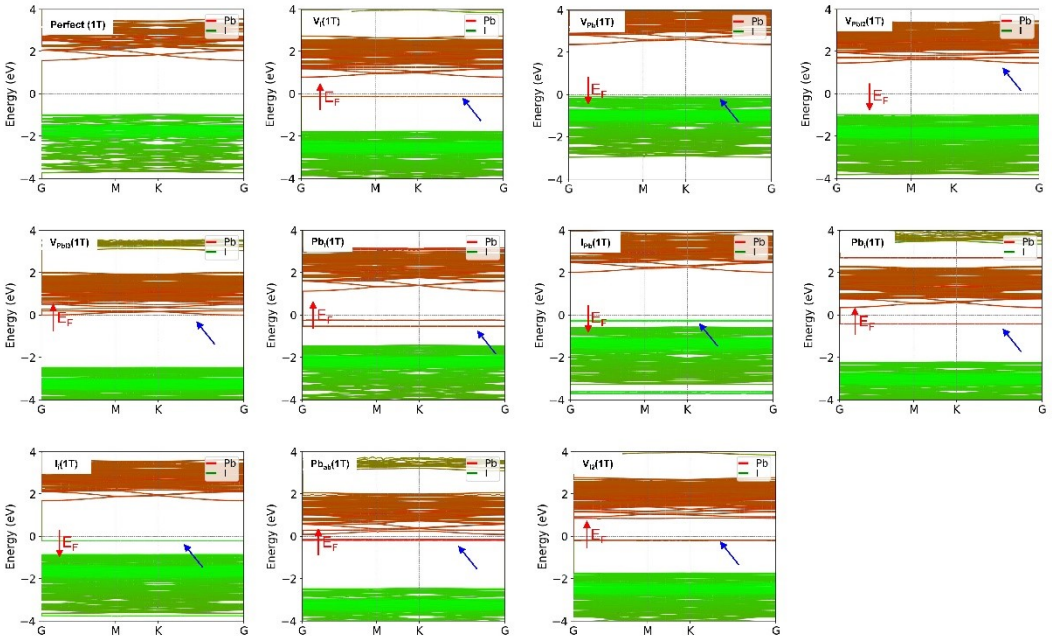

Figure S3. Band structures of various point defects in 1T phase PbI_2. The shift in Fermi energy levels is marked by a red arrow, the electron and hole states caused by defects are marked by blue arrows.

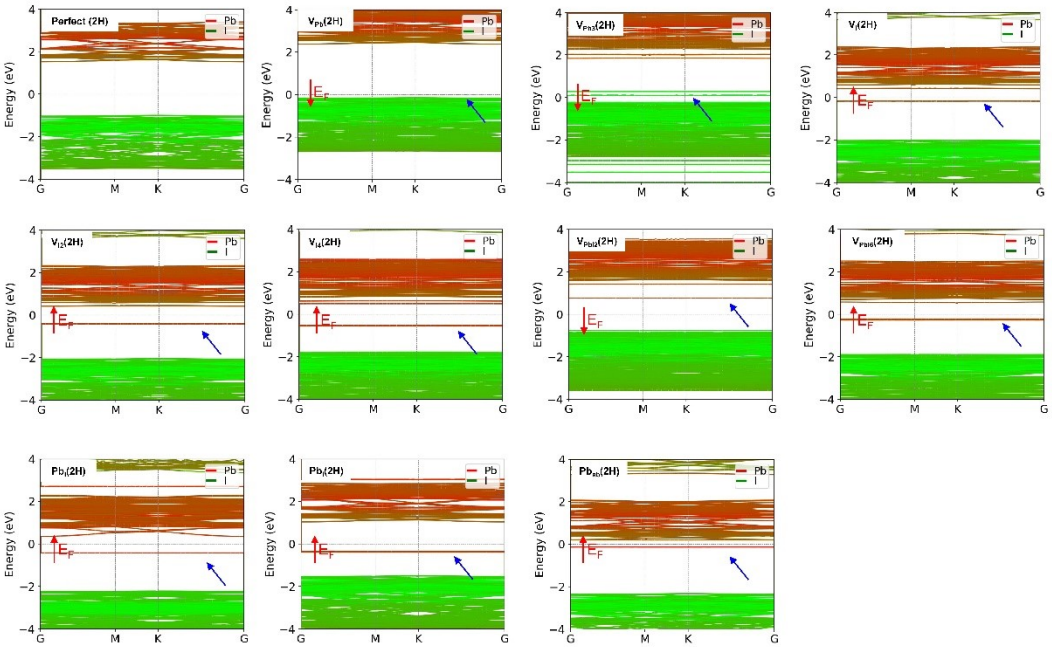

Figure S4. Band structures of various point defects in 1H phase PbI_2. The shift in Fermi energy levels is marked by a red arrow, the electron and hole states caused by defects are marked by blue arrows.

Table S3. The Comparison of the formation energy of defects between PbI_2 and MoS_2

Chemical potential	Formation energy (eV)					
	MoS_2[1]		PbI_2			
			1T		1H	
	V_{Mo}	V_S	V_{Pb}	V_I	V_{Pb}	V_I
S rich /I rich	~5.7	~2.4	1.1	2.2	0.7	2.4
S poor/I poor	~7.7	~1.3	4.0	0.9	3.9	0.7

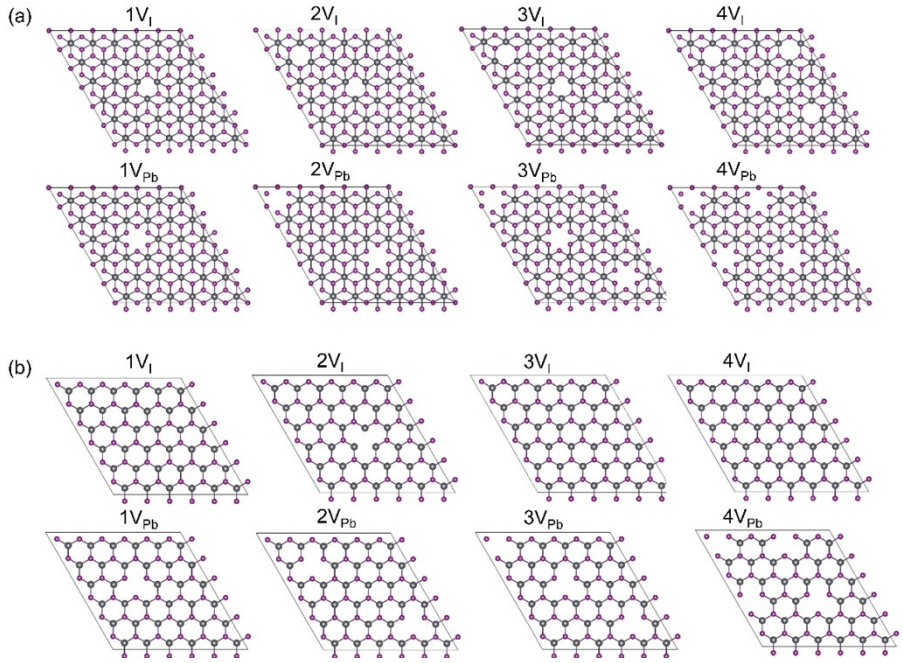

Figure S5. The optimized structures with different concentration of defects of (a) 1T and (b) 1H phase.

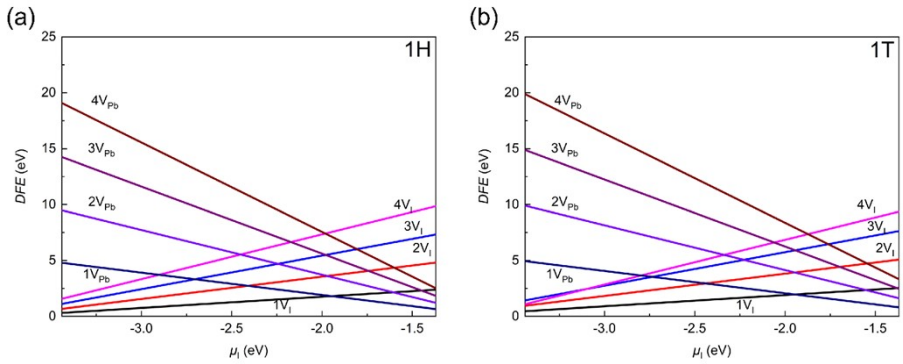

Figure S6. Formation energies of V_I and V_{Pb} in PbI_2 of (a) 1T and (b) 1H phase at their neutral charge state as a function of defect concentration.

References

1 H.-P. Komsa and A. V. Krasheninnikov, *Phys. Rev. B*, 2015, **91**, 125304.