Electronic Supplementary Information

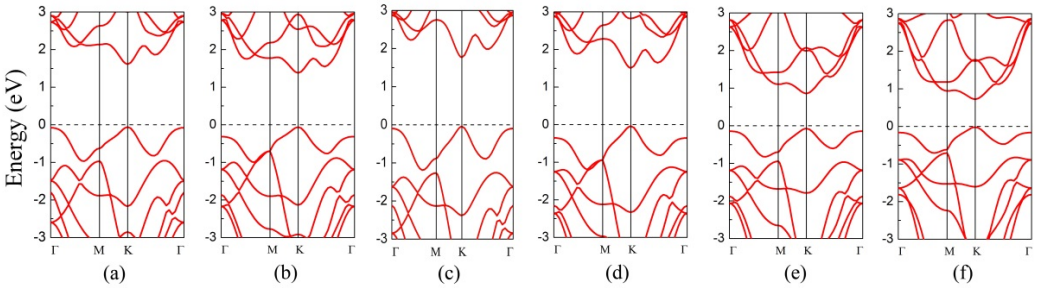

Figure S1. Computed electronic band structures of (a) MoS_2, (b) $MoSe_2$, (c) WS_2, (d) WSe_2, (e) CrS_2, and (f) $CrSe_2$ monolayer, based on PBE functional. All monolayers exhibits a direct bandgap.

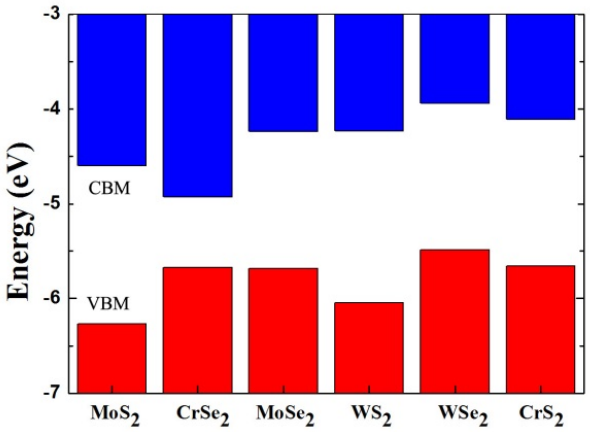

Figure S2. Computed positions of CBM and VBM of TMD monolayers, respectively, using PBE functional. The vacuum level is set to zero.

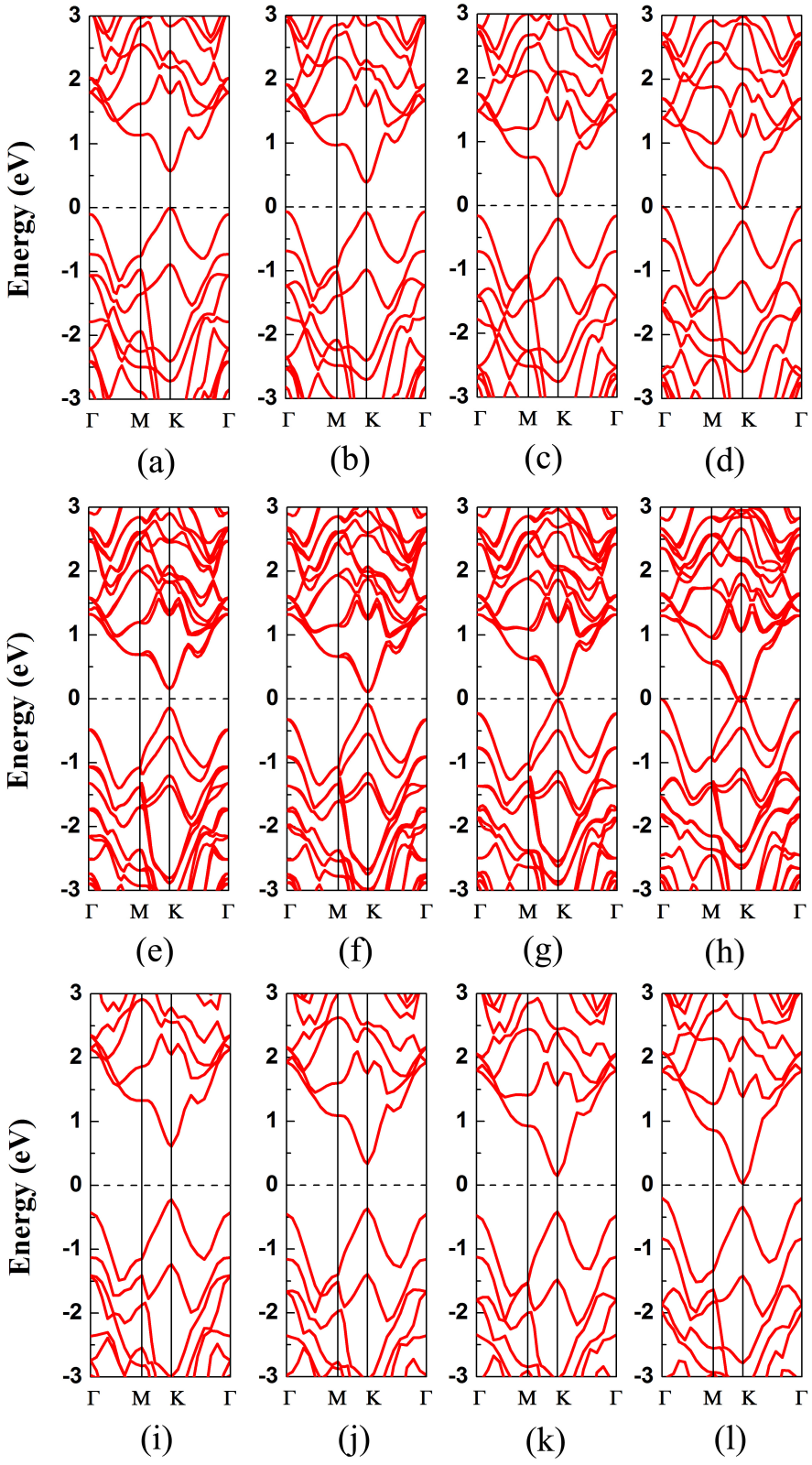

Figure S3. Computed electronic band structures (PBE) of WSe_2/MoS_2 heterobilayer with (a) 0%, (b) 1%, (c) 2%, and (d) 4% biaxial strain. Computed band structure (PBE) of WSe_2/MoS_2 with inclusion of spin-orbit coupling (SOC) effect and with (e) 0%, (f) 1%, (g) 2%, and (h) 4% biaxial strain. Computed band structures (**HSE06**) of WSe_2/MoS_2 with (i) 0%, (j) 1%, (k) 2%, and (l) 4% biaxial strain.

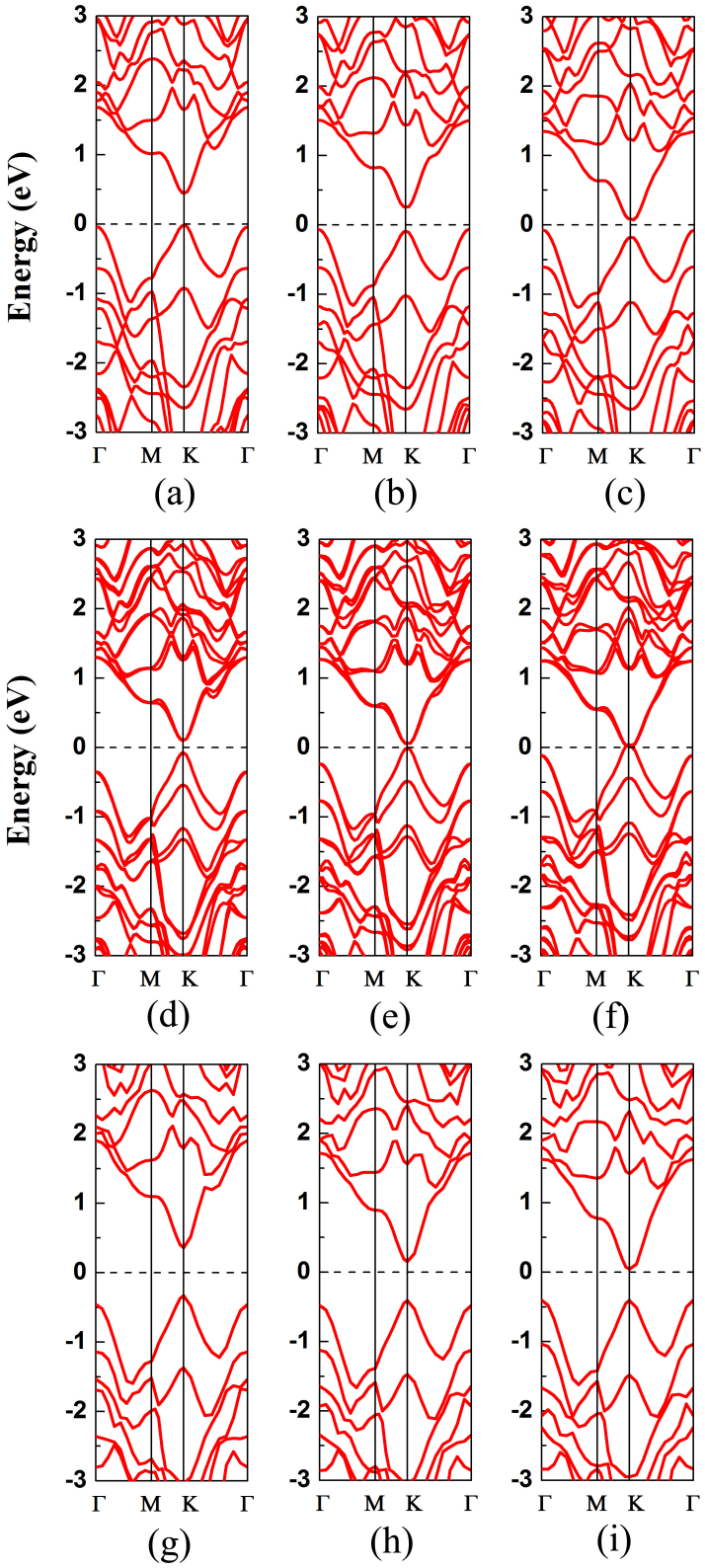

Figure S4. Computed electronic band structures (PBE) of WSe_2/MoS_2 heterobilayer with (a) 2%, (b) 4%, and (c) 6% strain along the *x*-direction. Computed band structures of WSe_2/MoS_2 with inclusion of SOC effect and with (d) 2%, (e) 4%, and (f) 6% strain along *x*-direction. Calculated band structures (HSE06) of WSe_2/MoS_2 with (g) 2%, (h) 4%, and (i) 6% strain along *x*-direction.

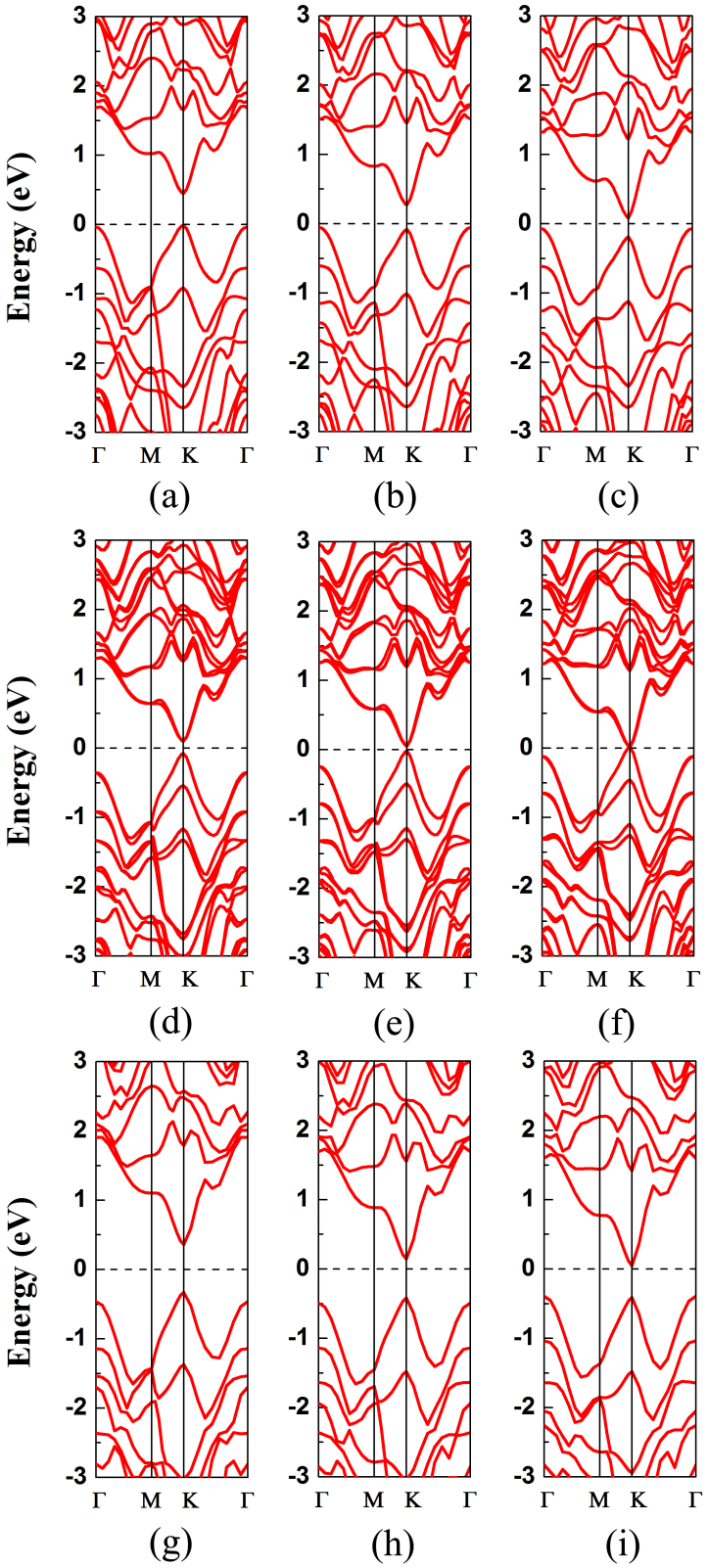

Figure S5. Computed electronic band structures (PBE) of WSe_2/MoS_2 heterobilayer with (a) 2%, (b) 4%, and (c) 6% strain along the *y*-direction. Computed band structures of WSe_2/MoS_2 with inclusion of SOC effect and with (d) 2%, (e) 4%, and (f) 6% strain along *y*-direction. Calculated band structures (HSE06) of WSe_2/MoS_2 with (g) 2%, (h) 4%, and (i) 6% strain along *y*-direction.

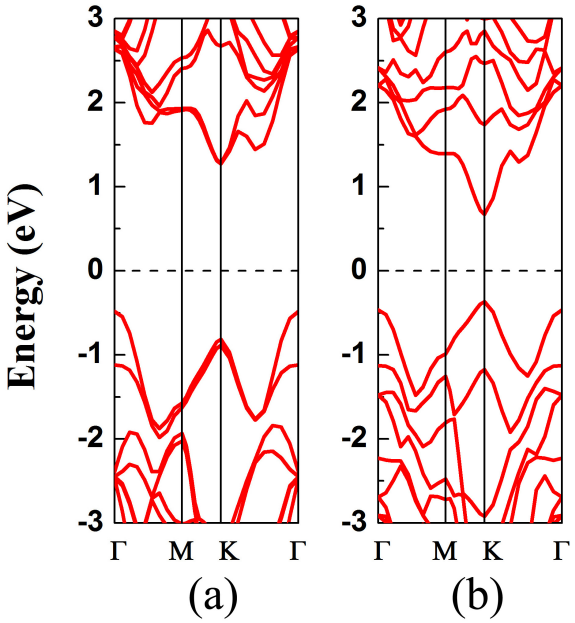

Figure S6. (a) Computed electronic band structures (HSE06) of MoS_2 bilayer. Note that the measured indirect bandgap of MoS_2 bilayer is about 1.53 eV (Ref. 25). So the computed HSE06 indirect bandgap (1.77 eV) overestimates the bandgap of MoS_2 bilayer. (b) Computed band structures (HSE06) of $MoSe_2/MoS_2$ heterobilayer. The $MoSe_2/MoS_2$ heterobilayer exhibits a direct bandgap. PBE calculation suggests $MoSe_2/MoS_2$ heterobilayer possesses a quasi-direct bandgap (Figure 3(a)).

Table S1.Cell parameter *a* in Å and the distance d_{S-X} in Å between the two TMD heterobilayers.

Heterobilayer	*a*	d_{S-X}
$MoSe_2/MoS_2$	3.26	3.12
WS_2/MoS_2	3.19	3.10
WSe_2/MoS_2	3.26	3.11
CrS_2/MoS_2	3.13	3.18
$CrSe_2/MoS_2$	3.21	3.16
FeS_2/MoS_2	3.17	3.12
VS_2/MoS_2	3.19	3.08
VSe_2/MoS_2	3.26	3.14

Table S2.Computed Bader charge transfer between $MoSe_2$ and MoS_2 layer under different external electric field (normal to the plane of the bilayer).

Electric field (V/ Å)	**0**	**0.1**	**0.2**	**0.3**	**0.4**	**0.5**	**0.6**
Charge (e)	0.0099	0.0125	0.0149	0.0172	0.0201	0.0216	0.0272

Table S3. Computed bandgap (PBE) of the $MoSe_2/MoS_2$ heterobilayer with and without geometric optimization under the electric field.

Electric field (V/ Å)	**0.1**	**0.2**	**0.3**	**0.4**	**0.5**
Direct gap	0.67	0.58	0.49	0.42	0.34
Indirect gap	0.70	0.65	0.59	0.54	0.49
Direct gap after geometric optimization	0.67	0.58	0.49	0.43	0.31
Indirect gap after geometric optimization	0.70	0.65	0.59	0.55	0.47