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

Supporting Information for:

Study of photogenerated exciton dissociation in transition metal dichalcogenide van der Waals heterojunction A2-MWS_4: A first-principle study

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Tab. S 1 Self-consistent energy of A2-MWS_4 with different values of cut-off energy.

Cut-off energy (eV)	Energy (eV)
400	-43.387
450	-43.388
500	-43.391
550	-43.391
600	-43.391
650	-43.392
700	-43.392

As indicated in **Tab. S 1**, the cut-off energy of A2-MWS_4 was set to 500eV because the calculation method was performed using the HSE (06) and GW0+BSE.[1, 2]

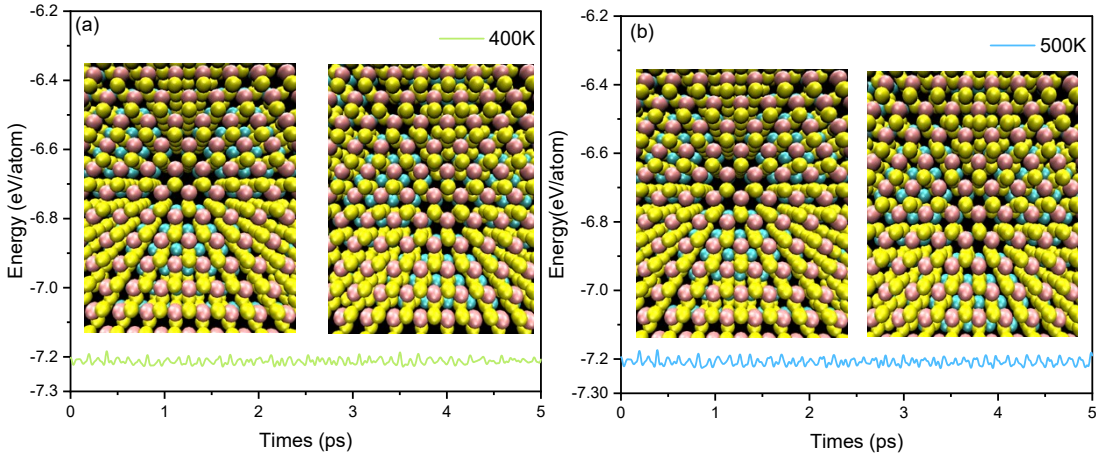
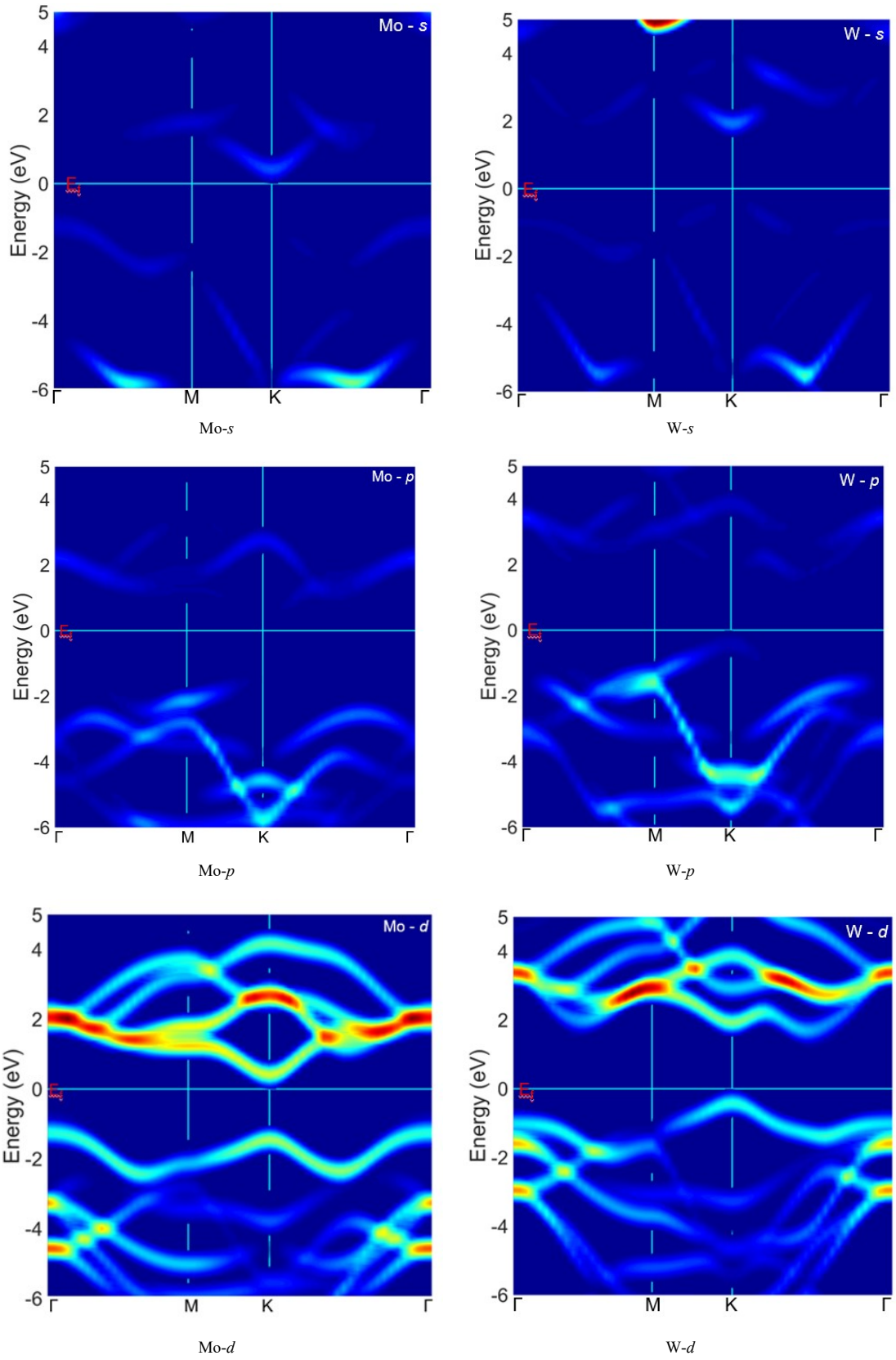


Fig. S 1 Variation of average atomic energy with dynamic steps in AIMD simulations of A2-MWS4 at different temperatures. (a)400K. (b)500K. The inset(right) of the figure showed a snapshot of the structure at the point of maximum energy fluctuation.

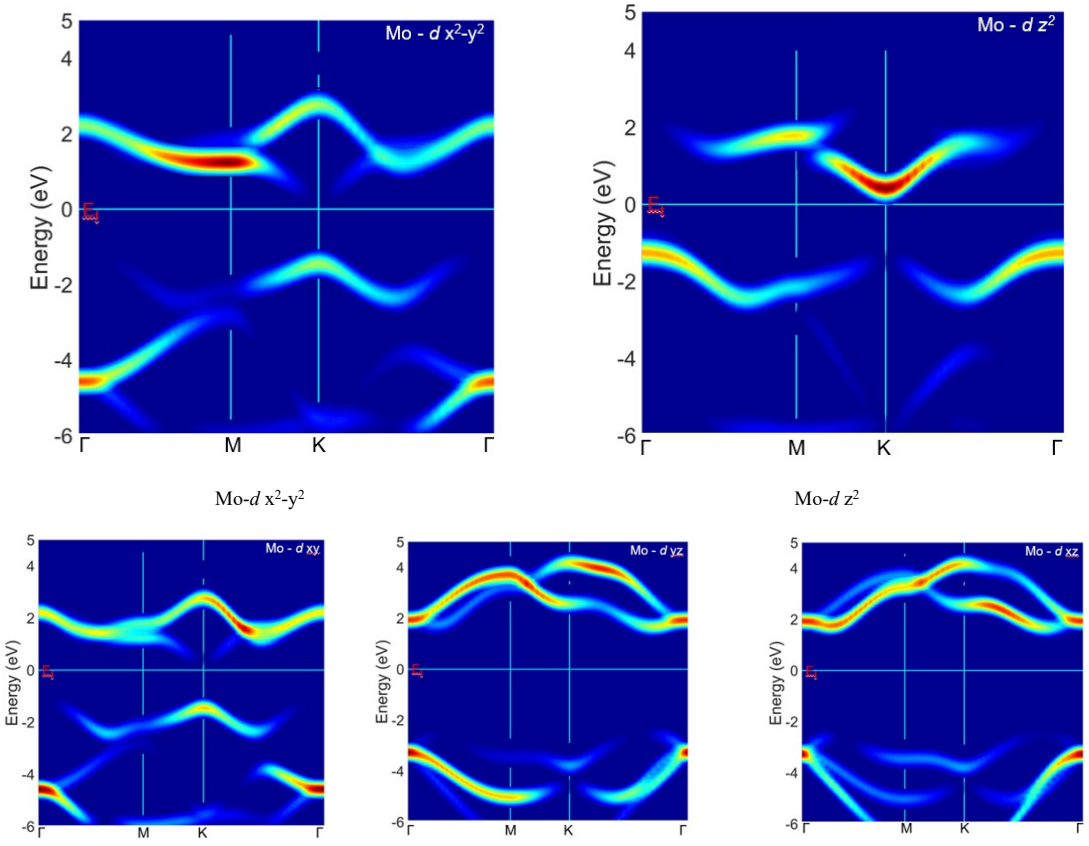


Mo-*s* W-*s*

Mo-*p* W-*p*

Mo-*d* W-*d*

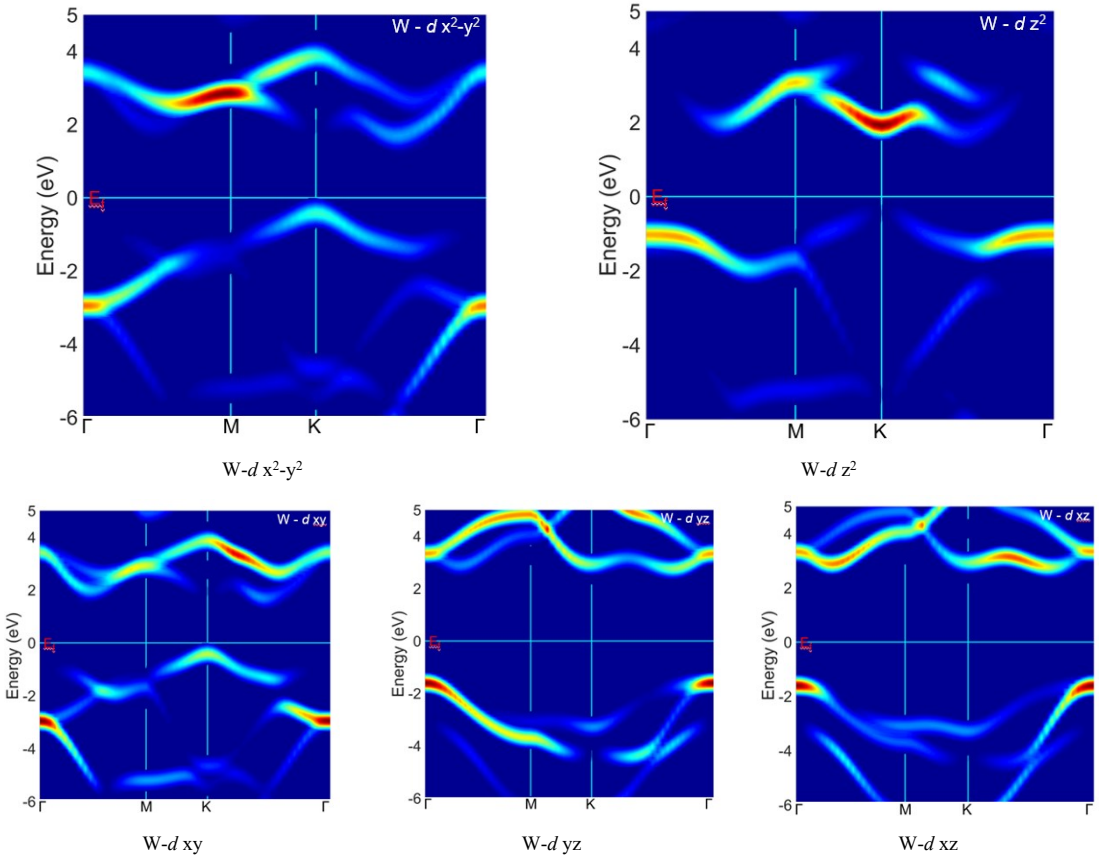
Fig. S2 A2-MWS_4 partial (Mo, and W) band structures.



Mo-*d* x^2-y^2 Mo-*d* z^2

Mo-*d* xy Mo-*d* yz Mo-*d* xz

Fig. S3 A2-MWS_4 partial (Mo) band structures.



W-*d* x^2-y^2 W-*d* z^2

W-*d* xy W-*d* yz W-*d* xz

Fig. S4 A2-MWS4 partial(W) band structures.

As shown in **Fig. S2**, **Fig. S3**, and **Fig. S4**, the VBM of A2-MWS4 is mainly contributed by the *d* (*d* xy, and *d* x^2-y^2) orbitals of W atoms and the CBM is mainly contributed by the d (*d* z^2) orbitals of MoS2.

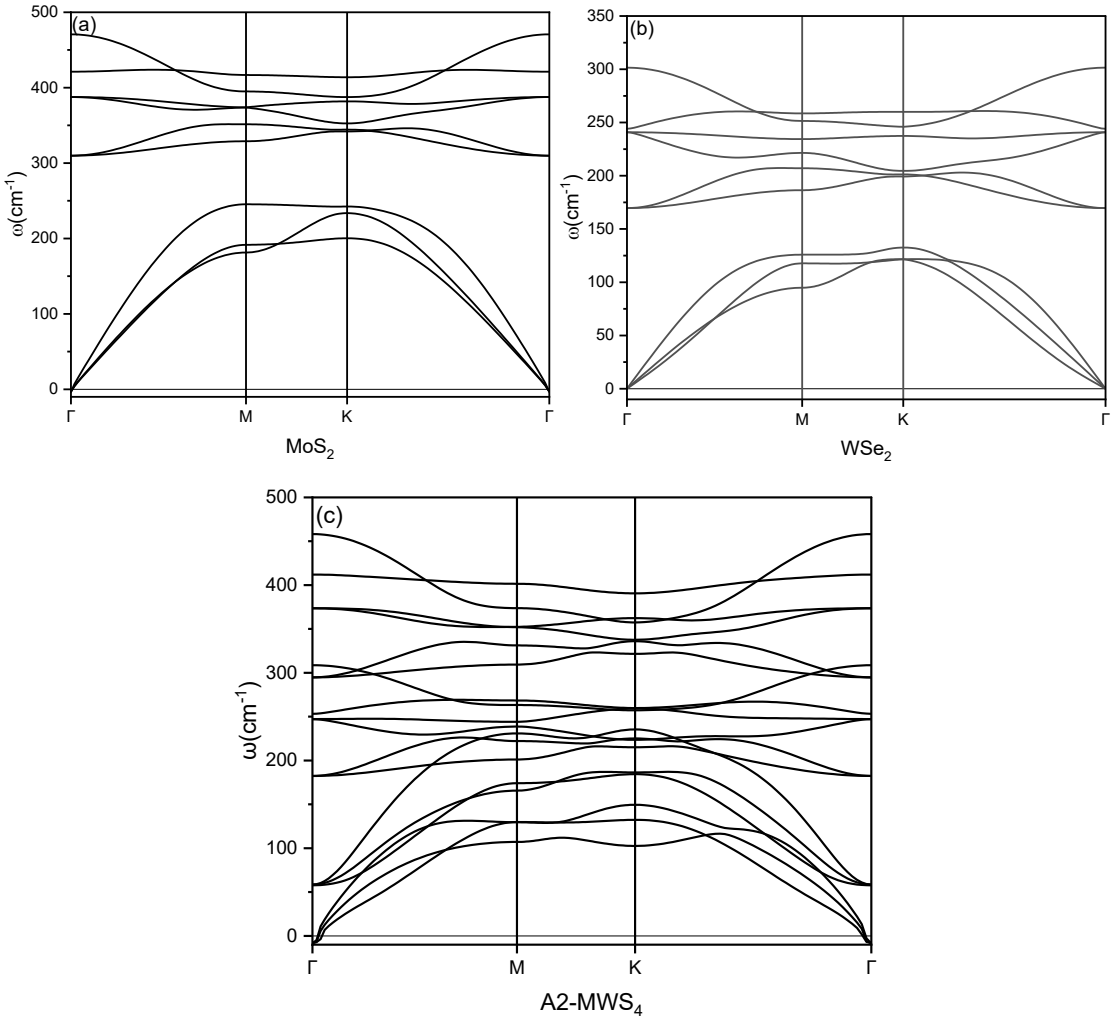


Fig. S5 The phonon dispersion curve of (a) MoS_2, (b) WSe_2, (c)A2-MWS_4.

As seen in **Fig. S5**, the phonon dispersion curve of A2-MWS4, WSe2, and MoS2 had small imaginary frequency (~ 8.42 cm-1, ~ 0.07 cm-1, and ~ 1 cm-1) around the Γ point. These imaginary modes may be caused by the non-optimal lattice constants and the finite supercell size, which indicates that the structures was thermodynamically stable.

References

[1] Hwang YH, Yun WS, Cha G-B, Hong SC, Han SW. Thermally driven homonuclear-stacking phase of MoS2 through desulfurization. Nanoscale. 2019;11:11138-44.

[2] Hsu W-T, Lu L-S, Wang D, Huang J-K, Li M-Y, Chang T-R, et al. Evidence of indirect gap in monolayer WSe2. Nature Communications. 2017;8:929.