Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Suppressing photoexcited electron-hole recombination in $MoSe_2/WSe_2$ lateral heterostructures via interface-coupled states engineering: a time-domain *ab initio* study

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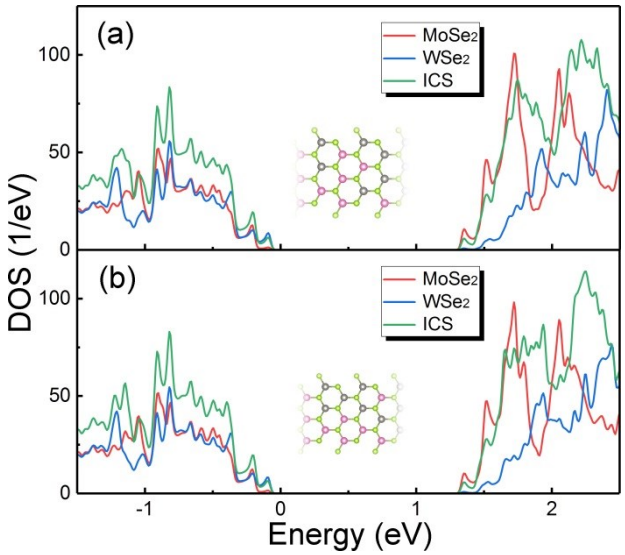

Fig. S1. LDOS of alloy interface models with (a) maximum and (b) medium relative formation energy. The inset is the interface configuration.

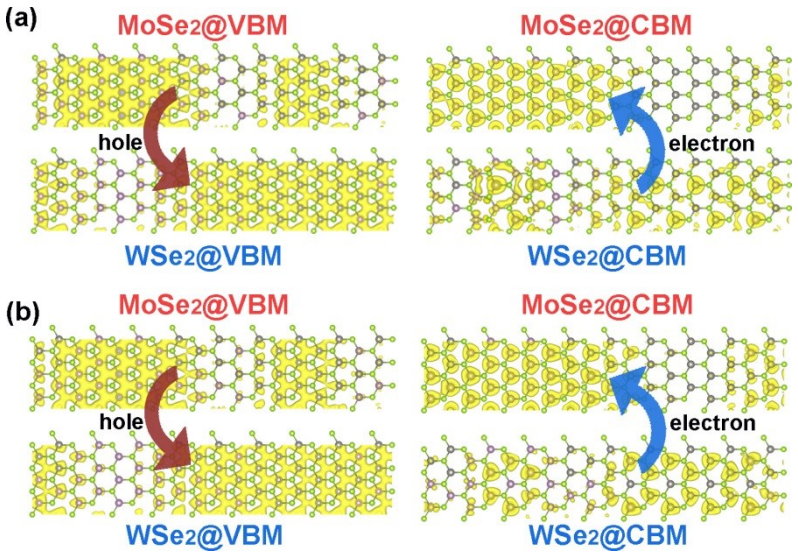

Fig. S2. Charge densities of the band-edge states of alloy interface models with (a) maximum and (b) medium relative formation energy. The red and blue arrows represent hole and electron transfer process, respectively. The isosurface value is set as 0.0012 e/Bohr^3.

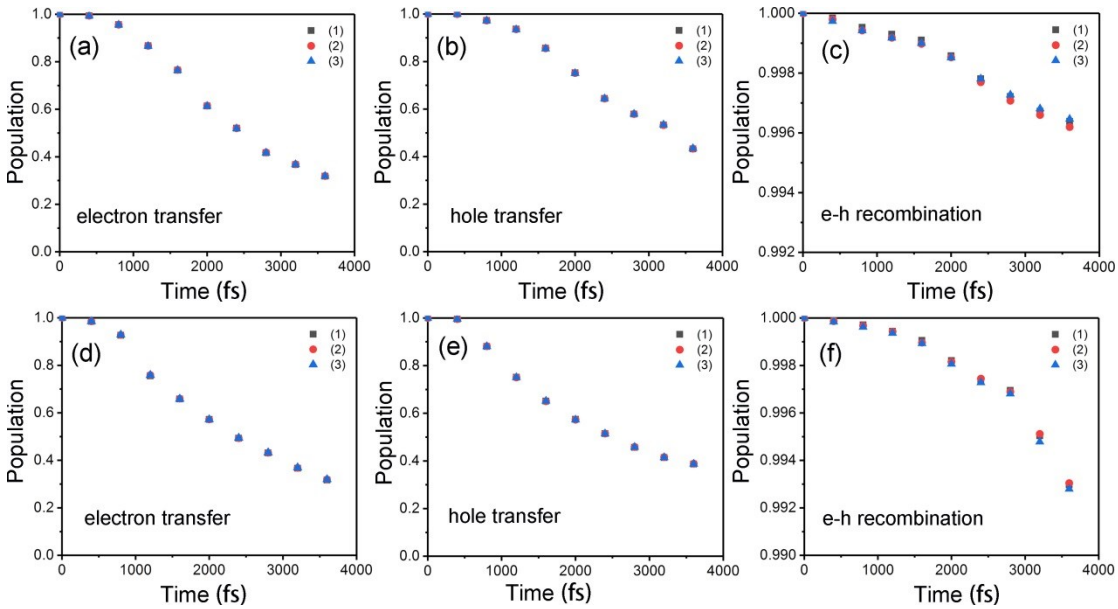

Fig. S3. Charge transfer and e-h recombination dynamics of $MoSe_2/WSe_2$ lateral heterostructures with (a-c) sharp and (d-f) alloy interface. The NAMD calculations are repeated three times, which represented using (1) (2) and (3).

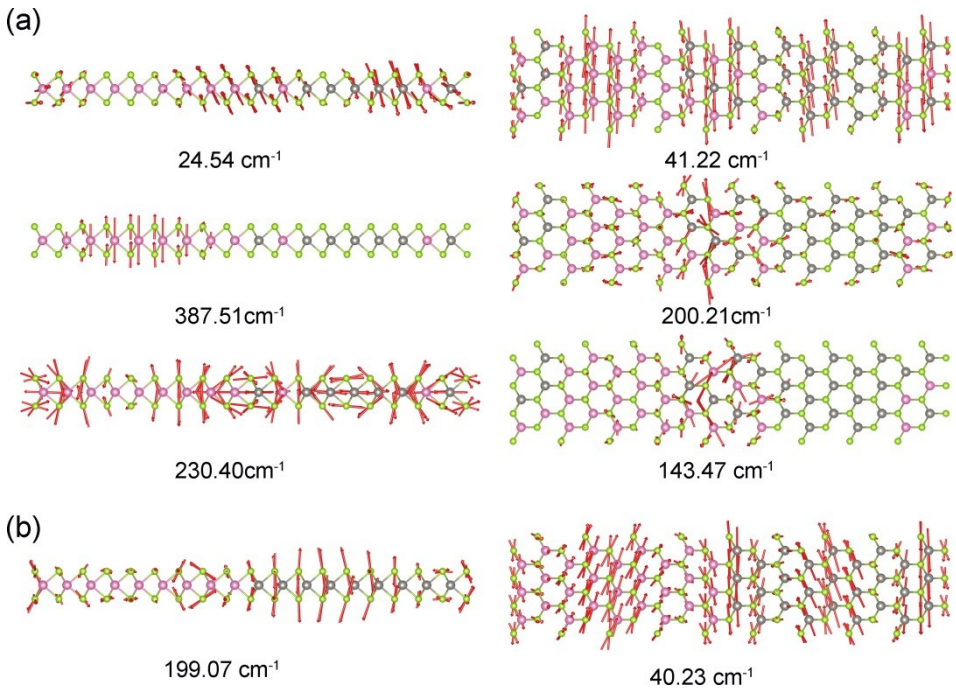

Fig. S4. Vibrational modes for $MoSe_2/WSe_2$ lateral heterostructures with (a) alloy and (b) sharp interface corresponding to the main peaks in Fig. 5. The vibrational modes in left panel are out-of-plane modes and in right panel are in-plane modes. Red arrows represent vibration vector.

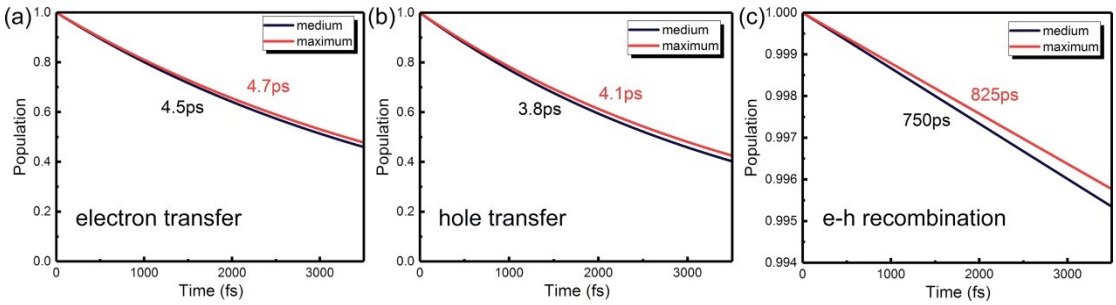

Fig. S5. (a) Electron transfer, (b) hole transfer and (c) e-h recombination dynamics of two alloy interface configurations with medium and maximum relative formation energy. The data are fitted by $f(t) = exp(-t/\tau)$.

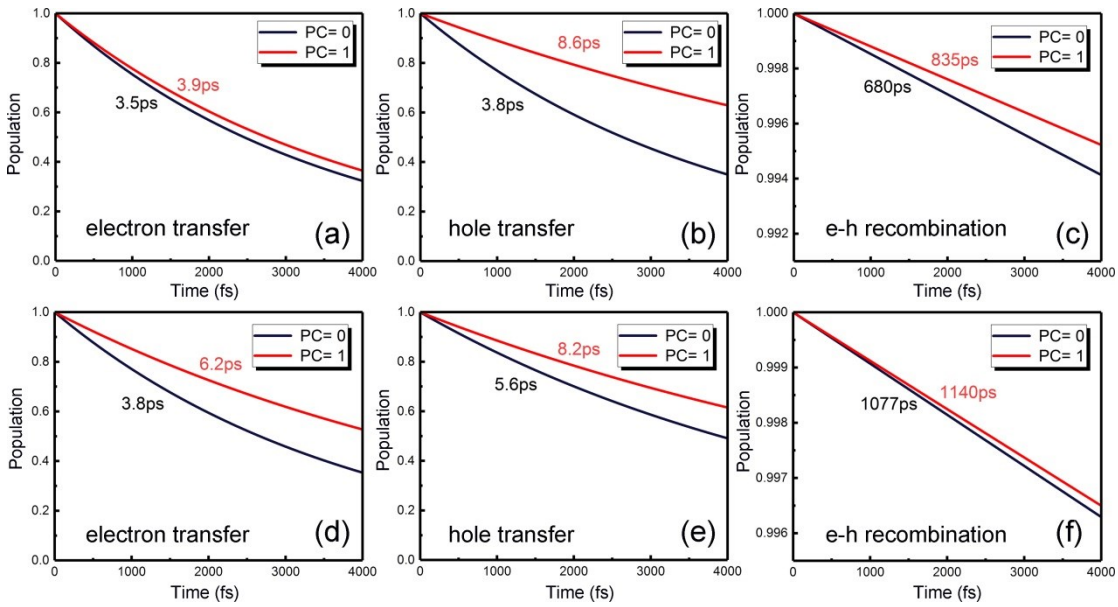

Fig. S6. The dynamics of carrier transfer and e-h recombination of $MoSe_2/WSe_2$ lateral heterostructures with (a-c) alloy interface and (d-f) sharp interface considering the phase correction. PC= 1 and 0 represent the dynamics with and without the phase correction of the adiabatic states. The data are fitted by $f(t) = exp(-t/\tau)$.

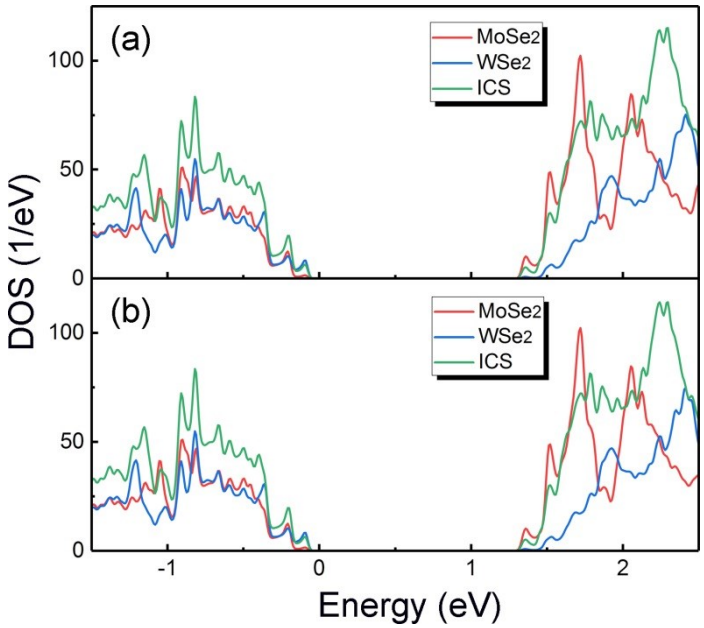

Fig. S7. LDOS of alloy interface models (a) without dipole and dispersion corrections and (b) with dipole and dispersion corrections.

In TMDs, defects will normally introduce defect trap states in band gap, which would significantly accelerate the electron-hole recombination.[1-4] Here we calculated the TDOS of $MoSe_2/WSe_2$ lateral heterostructures with interface vacancy defect. As shown in Fig. S8, the deep trap states appear in the band gap, which would potentially accelerate the electron-hole recombination.

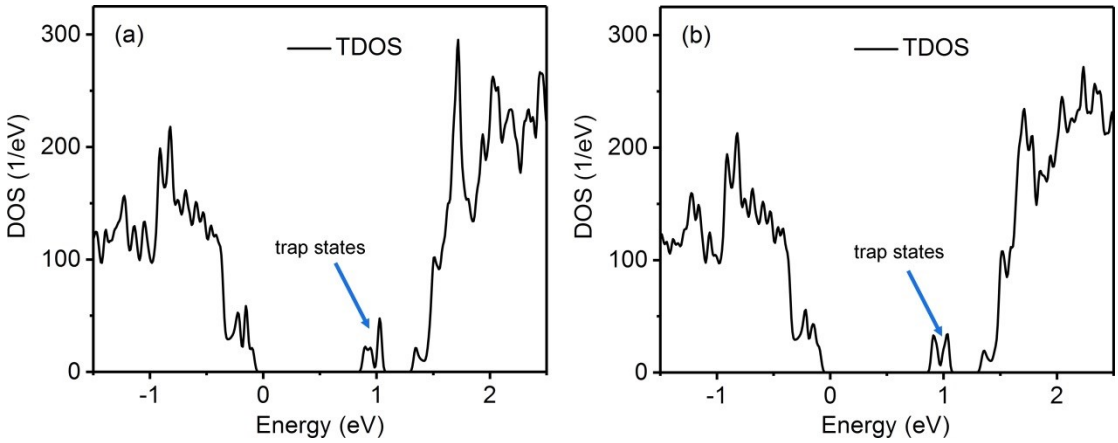

Fig. S8. TDOS of $MoSe_2/WSe_2$ lateral heterostructures with interface vacancy defect: (a) sharp interface and (b) alloy interface.

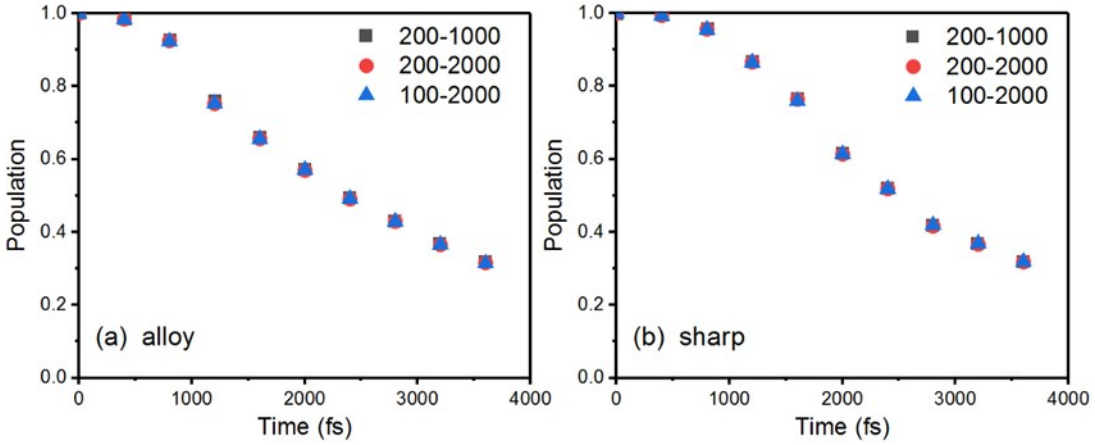

Fig. S9. Charge transfer of $MoSe_2/WSe_2$ lateral heterostructures with (a) alloy and (b) sharp interface. Square, circular and triangle represent the different sets of parameters. 200-1000: 200 initial configurations and 1000 surface hopping trajectories; 200-2000: 200 initial configurations and 2000 surface hopping trajectories; 100-2000: 100 initial configurations and 2000 surface hopping trajectories.

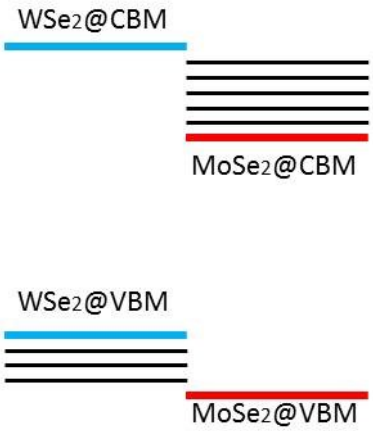

Fig. S10. Schematic diagram of energy states of $MoSe_2/WSe_2$ lateral heterostructures. Black lines represent the deep band states of $MoSe_2$ and WSe_2

Table S1 Relative formation energies of $MoSe_2/WSe_2$ lateral heterostructures with alloy interfaces. The minimum energy of -0.0195 eV is marked as bold.

	Relative formation energy (eV/unit cell)							
79 alloy interfaces	-0.0092	-0.0171	-0.0089	-0.0092	-0.0144	-0.0121	-0.0145	-0.0064
	-0.0064	**-0.0195**	-0.0064	-0.0121	-0.0090	-0.0091	-0.0171	-0.0090
	-0.0065	-0.0060	-0.0093	-0.0122	-0.0119	-0.0157	-0.0146	-0.0153
	-0.0176	-0.0094	-0.0121	-0.0123	-0.0121	-0.0091	-0.0127	-0.0120
	-0.0122	-0.0171	-0.0152	-0.0095	-0.0058	-0.0061	-0.0117	-0.0058
	-0.0092	-0.0120	-0.0058	-0.0142	-0.0119	-0.0126	-0.0120	-0.0093
	-0.0119	-0.0122	-0.0127	-0.0152	-0.0090	-0.0121	-0.0119	-0.0122
	-0.0119	-0.0120	-0.0174	-0.0119	-0.0142	-0.0142	-0.0090	-0.0123
	-0.0171	-0.0122	-0.0092	-0.0120	-0.0151	-0.0154	-0.0154	-0.0121
	-0.0156	-0.0062	-0.0063	-0.0124	-0.0124	-0.0123	-0.0089	

Table S2 Total energies and entropies of alloy interface configurations with minimum and the second minimum energy. $E_{tot+dipole+dispersion}$ and E_{tot} represents total energy with and without dipole and dispersion corrections respectively, and E_{TS} represents entropy.

	E_{tot} (eV)	$E_{tot+dipole+dispersion}$ (eV)	E_{TS} (eV)
Minimum energy model	-1124.744	-1179.182	-15.428
Second minimum energy model	-1124.732	-1179.170	-15.425

The input file of the NAMD calculations

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elec_dt = 0.001
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diconds = 5

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2. The POSCAR of $MoSe_2/WSe_2$ lateral heterostructures with alloy and sharp interface model

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0.500079989	0.499810010	0.442790002
0.500079989	0.499810010	0.576259971
0.555639982	0.666480005	0.442790002
0.555639982	0.666480005	0.576259971
0.611190021	0.499810010	0.442790002
0.611190021	0.499810010	0.576259971
0.666750014	0.666480005	0.442790002
0.666750014	0.666480005	0.576259971
0.722299993	0.499810010	0.442790002
0.722299993	0.499810010	0.576259971
0.777859986	0.666480005	0.442790002
0.777859986	0.666480005	0.576259971
0.833410025	0.499810010	0.442790002
0.833410025	0.499810010	0.576259971
0.888970017	0.666480005	0.442790002
0.888970017	0.666480005	0.576259971
0.944530010	0.499810010	0.442790002
0.944530010	0.499810010	0.576259971
0.000079989	0.999809980	0.442790002
0.000079989	0.999809980	0.576259971
0.055639982	0.833150029	0.442790002
0.055639982	0.833150029	0.576259971
0.111189999	0.999809980	0.442790002
0.111189999	0.999809980	0.576259971
0.166749999	0.833150029	0.442790002
0.166749999	0.833150029	0.576259971
0.222299993	0.999809980	0.442790002
0.222299993	0.999809980	0.576259971
0.277859986	0.833150029	0.442790002

0.277859986	0.833150029	0.576259971
0.333420008	0.999809980	0.442790002
0.333420008	0.999809980	0.576259971
0.388969988	0.833150029	0.442790002
0.388969988	0.833150029	0.576259971
0.444530010	0.999809980	0.442790002
0.444530010	0.999809980	0.576259971
0.500079989	0.833150029	0.442790002
0.500079989	0.833150029	0.576259971
0.555639982	0.999809980	0.442790002
0.555639982	0.999809980	0.576259971
0.611190021	0.833150029	0.442790002
0.611190021	0.833150029	0.576259971
0.666750014	0.999809980	0.442790002
0.666750014	0.999809980	0.576259971
0.722299993	0.833150029	0.442790002
0.722299993	0.833150029	0.576259971
0.777859986	0.999809980	0.442790002
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0.833410025	0.833150029	0.442790002
0.833410025	0.833150029	0.576259971
0.888970017	0.999809980	0.442790002
0.888970017	0.999809980	0.576259971
0.944530010	0.833150029	0.442790002
0.944530010	0.833150029	0.576259971
0.518599987	0.333149999	0.509530008
0.574159980	0.166480005	0.509530008
0.629710019	0.333149999	0.509530008
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0.796379983	0.166480005	0.509530008
0.851930022	0.333149999	0.509530008
0.907490015	0.166480005	0.509530008
0.963039994	0.333149999	0.509530008
0.518599987	0.666480005	0.509530008
0.574159980	0.499810010	0.509530008
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0.740819991	0.666480005	0.509530008
0.796379983	0.499810010	0.509530008
0.851930022	0.666480005	0.509530008
0.907490015	0.499810010	0.509530008
0.963039994	0.666480005	0.509530008
0.518599987	0.999809980	0.509530008

0.574159980	0.833150029	0.509530008
0.629710019	0.999809980	0.509530008
0.685270011	0.833150029	0.509530008
0.740819991	0.999809980	0.509530008
0.796379983	0.833150029	0.509530008
0.851930022	0.999809980	0.509530008
0.907490015	0.833150029	0.509530008
0.963039994	0.999809980	0.509530008

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