Supplementary material

A new charge transfer pathway in $MoSe_2$-WSe_2 heterostructure under the condition of B-excitons resonantly pumped

Zifan Hu, Hai Wang, Lei Wang, and Haiyu Wang*
State Key Laboratory of Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun 130012, China. E-mail: Haiyu_wang@jlu.edu.cn

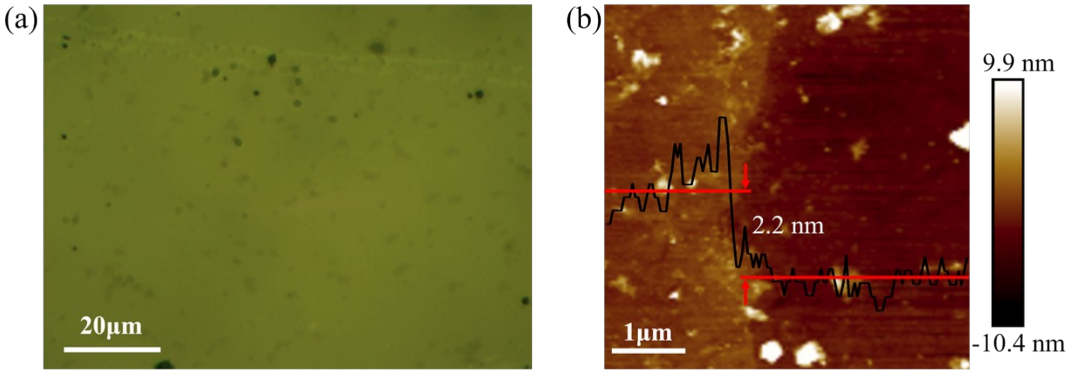

Figure S1 a) The optical images of $MoSe_2$–WSe_2 heterostructure. b) AFM characterization of the $MoSe_2$–WSe_2 heterostructure.

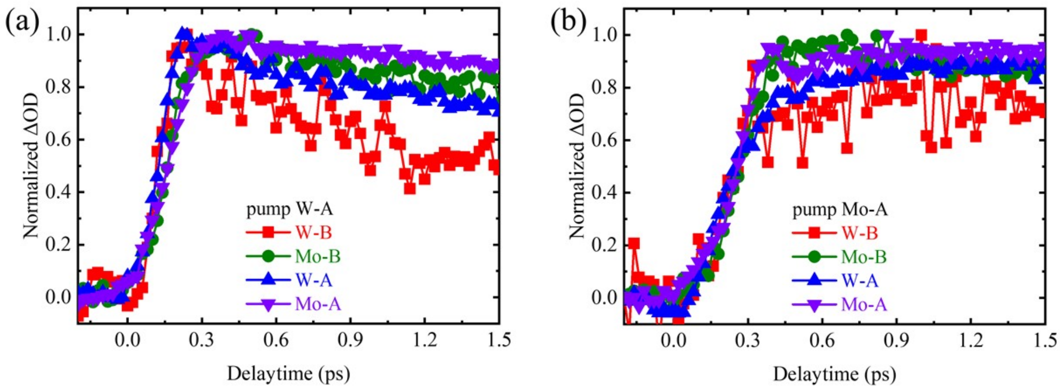

Figure S2 Dynamics of A- and B-excitons of $MoSe_2$–WSe_2 HS in the initial stage under conditions of a) W-A and b) Mo-A resonantly pumped.

Under conditions of W-A and Mo-A resonantly pumped, A- and B-excitons in the WSe_2 layer of $MoSe_2$–WSe_2 HS had synchronous build-up dynamics, as same as A- and B-excitons in the $MoSe_2$ layer. Because only A-excitons in the WSe_2 or $MoSe_2$ layer were pumped, the charge transfer process occurs through the energy state with lower energy corresponding to A-excitons.

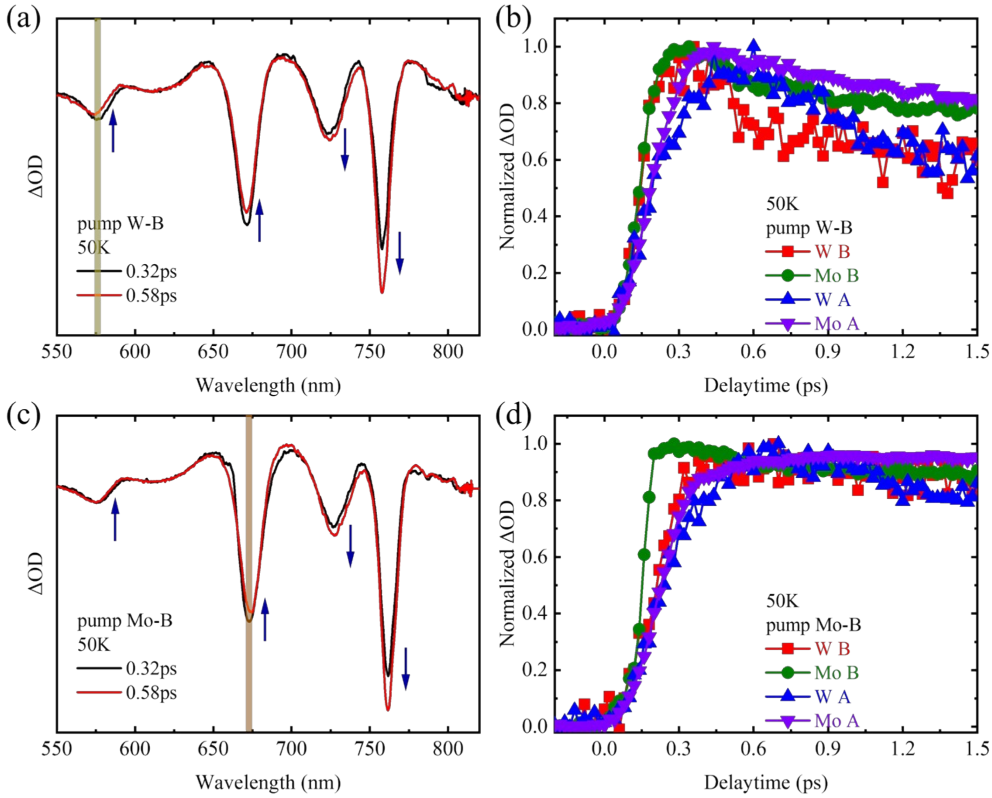

Figure S3 The TA spectra of $MoSe_2$–WSe_2 at time delays of 0.32 and 0.58 ps at a temperature of 50 K upon being pumped by 580 nm and 670 nm light are shown in a) and c). The dynamics of A and B-excitons in MoSe2–WSe2 at a temperature of 50 K during the initial stage upon being pumped by 580 nm and 670 nm light are shown in b) and d).

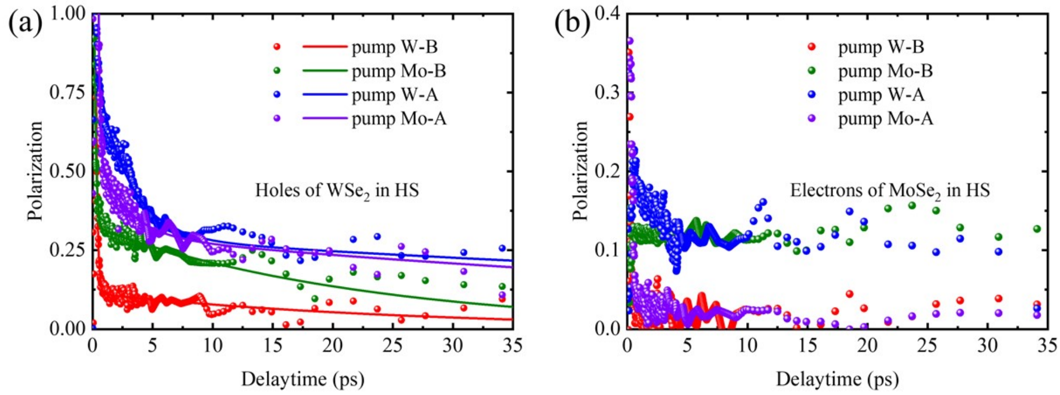

Figure S4 Polarization dynamics of a) holes of WSe_2 and b) electrons of $MoSe_2$ in the heterostructure under conditions of W-B (red), Mo-B (olive), W-A (blue), and Mo-A (violet) resonantly pumped. The solid lines are the fitted curves.

Due to a poor signal-to-noise ratio near 800 nm of probe light and small spin-valley polarization of the electrons of $MoSe_2$ in the heterostructure, it is difficult to fit the depolarization dynamics of the electrons with an exponential function. However, we still could conclude that the depolarization of electrons and holes is no longer synchronous and the polarization of holes is larger than that of electrons under various conditions thanks to the obvious spin-valley polarization of holes.

Table S1 Best fitting parameters of holes of WSe_2 in $MoSe_2$-WSe_2 HS with a Function of $I(t) \propto \Sigma_i A_i \exp(-t/\tau_i)$.

The wavelength of pump light (nm)	τ_1 (ps)	τ_2 (ps)	τ_3 (ps)
590 (pump W-B)	0.13 (84.5%)	26.46 (15.5%)	
700 (pump Mo-B)	0.14 (66.1%)	22.85 (33.9%)	
750 (pump W-A)	2.6 (64.0%)	117.04 (36.0%)	
800 (pump Mo-A)	0.13 (47.3%)	2.45 (23.2%)	82.25 (29.5%)

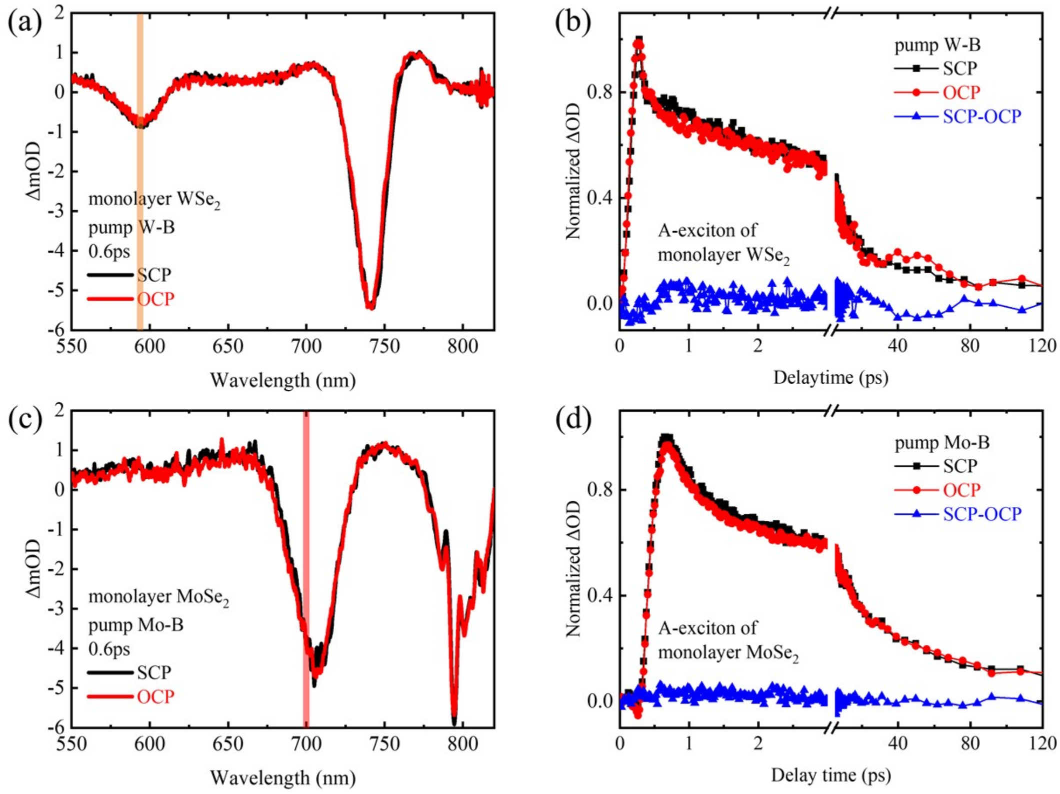

Figure S5 Spin-valley polarization in monolayer WSe_2 and $MoSe_2$. The TA spectra of SCP and OCP detection of a) WSe_2 and c) $MoSe_2$ at 0.6ps under the condition of B-exciton resonantly pumped. SCP and OCP dynamics of A-exciton of monolayer b) WSe_2 and d) $MoSe_2$. The blue trace is the evolution of the difference between SCP and OCP.