Supporting Information

Electrically Tunable Interlayer Recombination and Tunneling Behavior in WSe₂/MoS₂ Heterostructure for Broadband Photodetector

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As shown in Figure S1, the top-gate voltage (x-axis) and bias voltage (y-axis) divide the conductivity of the device into four parts, which has the potential of multidimensional tunability [1, 2]. As shown in (e), zone A, B belong the state IV (the division of the state is shown in Figure 3), where both forward and reverse bias voltages drive a diffusion of electron carriers between WSe₂ and MoS₂ to generate the drain current. Zone C, D belongs the state II, where the heterostructure is a typical p-n junction. Hence, at zone C the forward bias leads to a diffusion current contributed to the diffusion of majority carriers, while the reverse bias generates a drift current attributed to the minority carriers at zone D [3, 4]. Moreover, it was observed that the depletion zone (state III) gets wider significantly with higher positive back-gate voltage. Wherein, the state IV remains unmoved, and the state II shifts towards left. However, with higher negative back-gate voltage, the depletion zone slightly widens, where the state IV and II shift to right.



Figure S1 Transfer characteristics at varying bias voltages under different back-gate voltages. All the black dash lines are copied from that of (e), which presents the change of the transfer's contour color under different back-gate voltages.

At the p-i state (see Figure 3), the Fermi level starts to shift toward the conduction band in MoS₂ and the accumulated holes in WSe₂ diffuses into MoS₂, resulting in a diffusion current. With more positive V_{tg} , the carrier concentrations in WSe₂ and MoS₂ reach an equilibrium state. In the process, the variation of V_{tg} impedes the diffusion of majority carriers between WSe₂ and MoS₂. With further more positive V_{tg} , MoS₂ turns into the electron accumulation state and the electron concentration in MoS₂ becomes higher than that of WSe₂. Thus, the increase of V_{tg} benefits the diffusion current in the device until the state of i-n.



Figure S2 (a, b) Transfer and (c, d) transconductance characteristics under different back-gate voltages at fixed bias voltage of 1 V.



Figure S3 Output curves under different top- and back-gate voltages.



Figure S4 Transfer curves under illumination of 400 nm wavelength with different light intensity at different back-gate voltages and fixed bias voltage of 1 V. The marked

regions (II, III and IV) in the figures represent different electrical states of the device at different top-gate voltages. It shows that the state II widens and the state IV shifts to left with increasing light intensity.



Figure S5 (a) Transfer characteristics under illumination of 400 nm wavelength with different light intensity at fixed back-gate voltage of 0 V and fixed bias voltage of 1 V. (b) Output characteristics under illumination of 400 nm wavelength with different light intensity at fixed top- and back-gate voltages of 0 V and -40 V, respectively. (c) Transfer characteristics under different temperatures at fixed top-gate voltage of 0 V and fixed bias voltage of 0 V and fixed bias voltage of 5 V. (d) Output characteristics under different temperatures at fixed top-gate voltages at fixed top- and back-gate voltages of 0 V and -60 V, respectively.



Figure S6 The responsivity curves under illumination of 400, 700, 900 and 1010 nm wavelengths with different light intensity at different top- and back-gate voltages and fixed bias voltage of 1 V.



Figure S7 The specific detectivity curves under illumination of 400, 700, 900 and 1010 nm wavelengths with different light intensity at different top- and back-gate voltages and fixed bias voltage of 1 V.

Materials	Wavelength	Responsivity	Specific Detectivity	Daf
Configuration	nm	A/W	Jones	Kel.
WSe ₂ /CuO	530	0.28×10 ⁻³	0.19×10 ¹⁰	[5]
WSe ₂ /MoS ₂	532	1.392	7.78×10^{12}	[6]
b-AsP/WSe ₂	275	244	2.27×10^{12}	[7]

Table S1 Performance comparison of 2D heterostructure-based photodetectors.

WSe ₂ /hBN	532	170	1.1×10^{12}	[8]
WSe_2/MoS_2	532	2700	5×10 ¹¹	[9]
MoS ₂ /WSe ₂	633	8	2.93×10 ¹¹	[10]
WS ₂ /WSe ₂ /p-Si	405	3.72	2.39×10 ¹²	[11]
Ta2NiSe5/WSe2	638	0.04×10 ⁻³	3.6×10 ⁵	[12]
InSe/WSe ₂	633	5300	1011	[13]
PtS ₂ /WSe ₂	635	0.42	1.37×10^{12}	[14]
WSe_2/MoS_2	400	27445	10 ¹³	This work
WSe ₂ /MoS ₂	1010	2827	10 ¹⁰	This work

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