Supporting information

The role of trap state in MoS_2-based photodetector

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MoS_2 device:

Figure S1. AFM image of MoS_2. The thickness of MoS_2 film is 60 nm, which is characterized by atomic force microscope (AFM). All samples in this paper have approximately the same thickness.

Under the bidirectional sweep of the transfer curve, the curve produces a clear window of hysteresis. Changing the starting voltage of the scanning gate voltage or the magnitude of the optical power density result in a corresponding alteration to the magnitude of the hysteresis window. The hysteresis window is defined as the difference between the two voltages when Ids is equal to 10^{-7}A. The hysteresis window during bidirectional scanning of transfer curves was previously reported to be caused by electron trapping of electrons.[1] The subthreshold slope (SS) is calculated from the up-sweep characteristics of the transfer characteristics to shed light on both the deep and shallow electron trap density, and the respective trap density is obtained from the equation.[1]

$$SS = \frac{mk_BT}{q} ln10; m = 1 + \frac{C_s}{C_{ox}} + \frac{C_{it}}{Cox}; C_{it} = qD_{it}$$

Where C_s is the semiconductor capacitance, C_{ox} is the oxide capacitance, C_{it} is the interface trap capacitance, and D_{it} is the interface trap density.[2] For an ultra-thin body like monolayer it MoS_2, C_s is assumed to be zero. Figure S2c and Figure S2d illustrate the surface trap state density at different starting scan voltages and different optical power densities.

Figure S2. (a)-(d) The subthreshold slope (SS) and surface trapped state density of sample 1 at different starting scan gate voltages and different optical powers. (e)-(h) ΔV_{th} and density of trapped electrons at different starting scanning gate voltages and different optical powers.

Figure S2g and Figure S2h depict the trapped electron density at different starting scan voltages and different optical power densities. To determine trapped electron density, the threshold voltage is determined from both the up-sweep and down-sweep of the transfer characteristics by the equation,

$$\Delta n = \frac{\Delta V_{th} \times C_{ox}}{q}$$

where C_{ox} is the oxide capacitance and q is the elementary charge.

The I–V characteristic curve for Sample 2 is shown in Figure S2. The sample has a Schottky barrier at the metal-material contact, the I–V curve represented the response of two back-to-back series diodes, in which the current showed a symmetric reverse-bias-dominated response with external voltage.[3] Schottky barrier height calculated from hot electron emission theory:[4]

$$\emptyset = \frac{k_B T}{q}(lnA^* T^2 - ln\frac{I_0}{S})$$

The Schottky barrier heights calculated to be $\emptyset_B$ = 0.337 eV.

Figure S3. (a) I–V characteristic curve for sample 2. (b) The transfer characteristics at different optical powers at 635 nm of sample 1.

Figure S3b shows the transfer characteristic curve of sample 1 under 635 nm laser, which is consistent with the results of Figure 1f, the hysteresis window of the transfer characteristic decreases with the increase of optical power. Since MoS_2 is more sensitive to 635 nm laser light, the hysteresis window changes more rapidly.

Mechanism of trap state action under light:

Figure S4. Optical ON/OFF switching cycles under positive gate voltage.

Optical ON/OFF switching cycles with positive gate voltage modulation have faster speed of response The slow change phase is no longer evident, and this is attributed to the Fermi energy levels approaching the conduction band under positive gate voltage. Under these conditions, the energy levels of the trap states become occupied, thus resulting in an absence of the slow change phase. Increases in gate voltage have been shown to increase both the concentration of free electrons and the dark current and noise. This results in a lower I_{on}/I_{off}, which is not favourable for the application of MoS_2-based photodetectors.

Figure S5. Sample 3 of (a) transfer curve, (b) variation of photoresponsivity with gate voltage, and (c) variation of gain with optical power.

The threshold voltage of the amount of to sample 3 by the linear extraction method is about -18 V in the dark state, as shown in Figure S5(a). The variation of responsivity with threshold voltage under laser irradiation at different optical power densities at 635 nm is shown in Figure S5(b). The decrease in responsivity gate voltage shows a trend of increasing and then decreasing. At a given optical power density, the photoresponsivity is greatest near the threshold voltage.

The variation of the gain of the device with optical power at different gate voltages is shown in Figure S5(c). Data extraction using G~P$^{\alpha}$ fitting yielded $\alpha_{(V_G=-15V)}$ = - 0.59， $\alpha_{(V_G=-18V)}$ = - 0.66， $\alpha_{(V_G=-20V)}$ = - 0.59. V_G has a minimum α near the threshold voltage, which corresponds to the largest trap contribution, in agreement with the results for Sample 2 in Figure 2(f). When V_G is reduced to - 25 V, G increases with increasing optical power density at low optical power. This is due to the fact that the presence of a large number of recombination centers at smaller gate voltages weakens the gain of the trap state. Trap-state gain-induced electrons preferentially populate the recombination center at low optical power. As the recombination center is continuously filled, these electrons begin to jump to the conduction band to participate in the conductivity, resulting in an increasing gain effect. As the number of recombination centers declines to a specific threshold, the influence of the trap state action becomes predominant, resulting in a reduction in gain as the optical power density is increased. The change in gain at V_G = - 28 V, - 30 V can verify this theory. And as the gate voltage decreases, the inflection point at which the gain begins to fall is shifted to the right, as a result of the increasing effect of the recombination center with decreasing gate voltage.

Light pulse analysis:

A biexponential relaxation equation[5] be used to fit the ON/OFF switching cycles, and the extracted four phase action times are shown in Figure S6.

Figure S6. Fast rise time (a), slow rise time (b), fast descent time (c) and descent rise time (d) for the same optical power with different gate voltage. Fast rise time (e), slow rise time (f), fast descent time (g) and slow descent time (h) for the same gate voltage with different optical power.

Figure S7. Response speed for (a) the same gate voltage with different optical power versus (b) the same optical power with different gate voltage.

As Figure S7a, the speed of response first increases and then decreases with decreasing gate voltage. This phenomenon is consistent with the responsivity. With small gate voltage traps are filled and photoconductive effect is the main physical mechanism leading to fast speed of response. Decreasing with gate voltage makes the trap energy level bare and starts trapping holes to generate gain, leading to slow speed of response. Further decrease in gate voltage results in the appearance of the recombination center again speeding up the speed of response. At V_G=-45 V, at which point V_G<V_{th}. The recombination center exists in abundance and the photogenerated carriers fill the recombination center first. This results in a slowing down of the speed of response with increasing optical power, as shown in Figure S7b.

Figure S8. Sample 4 Measurement of ON/OFF switching cycles at different source-drain bias voltages at gate voltages V_G of (a) - 15 V, (b) - 20 V, (c) - 25 V, (d) - 30 V, and (e) - 35 V.

The ON/OFF switching cycles measured at the same gate voltage and different source-drain voltages, respectively, are shown in Figure S8. As the source drain voltage increases, the corresponding pulsed photocurrent increases. The current density equation $J = nq\mu\varepsilon$, where n is the electron concentration, q is the electron charge, μ is the electron mobility, and ε is the electric field strength between the source and drain electrodes. Thus as the source drain voltage increases, it leads to an increase in the corresponding pulsed photocurrent. In addition, when the gate voltage V_G > -25 V, the magnitude of the photocurrent decreases with the number of pulses at the same source-drain voltage. This is due to the fact that under each optical pulse there are holes trapped by the trap, and when the second pulse arrives, some of the holes do not return to the ground state in time, resulting in a decrease in the density of the hole-trapped state, and a consequent decrease in the photocurrent gain and photocurrent. And when V_G < -25 V, the photocurrent increases with the number of pulses. At smaller gate voltages, the recombination center action is greater than the gain action of the trap state. Thus, with the first few light pulses, the electrons preferentially filled the recombination center to weaken the effect of the recombination center. When the number of recombination centers is reduced to a certain level, its effect is balanced with the effect of the trap state action, and the photocurrent is not changing with the increase of the pulse, as shown by V_{ds} = 0.1 V in Figure S8(d).

Figure S9. Four-stage currents under the first optical pulse at different source-drain bias voltages for the sample 4 at gate voltages V_G of (a) - 15 V, (b) - 20 V, (c) - 25 V, (d) - 30 V, and (e) - 35 V, respectively.

The photocurrents of the four phases of the first pulse of the ON/OFF switching cycles under different conditions in Figure S8 are extracted as shown in Figure S9. Except for V_G = - 35V, at the same gate voltage, the current in all stages increases with the source-drain voltage. Where $|I_{DE}|$ at V_G = - 35 V shows a decreasing trend with increasing source-drain voltage. The presence of a considerable number of recombination centers at relatively low gate voltages may be a contributing factor. As the source-drain voltage rises, the kinetic energy of the free carriers increases, resulting in greater motion and a higher level of electron susceptibility to recombination. This leads to a significant number of free carriers being recombined in the CD section, while the current change in the DE section is reduced.

Figure S10. α - V_G curve of different samples.

The values of α for different devices demonstrate an initial decrease, followed by an increase, with decreasing gate voltage.

Figure S11. I_{ph}~P and G~P

Table S1. Comparison of photoresponsivity of MoS_2-based photodetectors in recent researches.

Structure	Wavelength	Photoresponsivity ($A W^{-1}$)	Mechanism	Ref.
2L MoS_2	532 nm	1.1×10^5	hot electron-based photogain	6
MoS_2/SiC	325 nm	1.02×10^4	dual-photogating effect	7
1L MoS_2/1L graphene	532 nm	1.2×10^7	heterostructure-based photogain	8
FL MoS_2/CdSe	405 nm	2.5×10^5	n–n type heterojunctions	9
ML MoS_2/PPh3doping	520 nm	3.92×10^5	triphenylphosphine (PPh3) doping	10
Graphene/MoS_2/graphene	633 nm	1.14×10^5	band alignment by graphene	11
1L/6L MoS_2	520 nm	2.67×10^6	multi-heterojunction	12
ML MoS_2	460 nm	1.10×10^6	trap based-photogain	13

The high responsivity of MoS_2 comes from the photo gain and the built-in electric field induced by the homojunction (heterojunction). Figuring out the physical mechanism of photo gain plays an important role in the future development and application of MoS_2-based photodetectors.

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