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Supporting information for

Diverse Modes Regulated Photoresponse and High-Resolution Imaging based on

van der Waals Semimetal PtTe₂/Semiconductor MoTe₂ Junctions



Figure S1 The AFM image of the PtTe₂ nanosheets prepared by the Au-assisted exfoliation method. a the 2D topological AFM image of PtTe₂ nanosheets on Ti/Au SiO₂/Si substrate, and the height profiles along the white lines, respectively.



Figure S2 The HRTEM-EDS mapping analysis of the $PtTe_2/Au$ nanosheets. a Lowmagnification TEM image of the exfoliated and as-transferred $PtTe_2$ nanosheets with Au particle. b Element mapping of the two detected elements: Pt and c Te. d EDS spectrum extracted from the light blue rectangle in **a**.



Figure S3 The HRTEM image and SAED patterns of (a) individual PtTe₂ nanosheets and (b) individual MoTe₂ nanosheets.



Figure S4 AFM image of PtTe₂ and e MoTe₂ along the red, respectively.



Figure S5 a. I_{ds} - V_{ds} curves of the PtTe₂/MoTe₂ vertical Schottky junction diode with changing the temperature in dark and vacuum condition. **b.** The rectification ratio at different temperatures under the source-drain voltage variation of -2 V - +2 V. **c.** Arrhenius curve. **d.** The slope S- V_{DS} curve fitted by the Arrhenius curve in **Fig. S5c**.



Figure S6 Electrical performance of another ultrathin PtTe_2 FET. a. $\rm I_{ds}\text{-}V_{ds}$ curve.



Figure S7 Electrical performance of another thin MoTe₂ FET. a. I_{ds} - V_{ds} curve. b. Tranfer characteristic at $V_{ds} = 2$ V scanning from -40 V to 40 V. c. Output curves at various gate voltages.



Figure S8 Optoelectrical properties of the PtTe₂/MoTe₂ vertical heterojunction under illumination of different wavelength. a. The normalized I_{ph} as a function of illumination wavelength from 650 to 1100 nm at zero bias and zero source-drain voltages. b. The I_{ph} under the varied illumination wavelength from 650 to 1100 nm at zero gate voltage and zero source-drain voltage. The incident power P = 4.10 mW cm⁻². c. Logarithmic I_{ds} -V_{ds} curves in the dark and under different wavelengths illumination.



FigureS9 a, b I_{sc} and V_{oc} and light on/off ratio of PtTe₂/MoTe₂ heterostructure under different laser powers intensity of 635 nm, respectively. **c, d** I_{sc} and V_{oc} and light on/off ratio of the heterostructure under different laser powers intensity of 1310 nm, respectively.



Figure S10 Photoresponse performance of another PtTe₂/MoTe₂ vertical heterojunction using thin PtTe₂ of the thickness of ~20 nm. a. Optical microscope image of the stacked PtTe₂/MoTe₂ heterostructure. b. I_{ds} - V_{ds} liner curves of the PtTe₂/MoTe₂ vertical Schottky junction diode with changing the incident power density under the light illumination of 1064 nm. c. I_{ds} - V_{ds} log curves of the PtTe₂/MoTe₂ vertical Schottky junction diode with changing the incident power density under the light illumination of 1064 nm. d.Transfer curves relate to light power density at $V_{ds} = +1$ V under 1064 nm illuminations. e. Photocurrent curves relate to light power density at $V_{ds} = +1$ V and 0 V under 1064 nm illuminations. f. Responsivity and detectivity ($V_{ds} = +1$ V and 0 V) relate to light power density under 1064 nm illuminations.



Figure S11 Photoresponse performance of another $PtTe_2/MoTe_2$ vertical heterojunction using thin $PtTe_2$ of the thickness of ~50 nm. a. Optical microscope image of the stacked $PtTe_2/MoTe_2$ heterostructure. b. I_{ds} - V_{ds} curves of the $PtTe_2/MoTe_2$ vertical Schottky junction diode with changing

the incident power density under the light illumination of 980 nm. **c.** Variation of output electrical power with various light illumination power densities under light illumination of 980 nm. **d.** Photoresponse ($V_{ds} \ 0 \ V$) and V_{oc} relate to light power density under 980 nm illuminations. **e.** Responsivity ($V_{ds} = -1 \ V$ and 0 V) and V_{oc} relate to light power density under 980 nm illuminations. **f.** Photocurrent curves relate to light power density at $V_{ds} = -1 \ V$ and 0 V under 980 nm illuminations.



Figure S12. Stability test of photoswitch behavior at $V_{ds} = 0$ V under 532 nm illumination.



Figure S13. The time-resolved photoresponse of the device under 635 nm illumination with various light power densities at the $V_g = -40$ V, 0 V, 40 V, respectively. Figure S14. The time-resolved photoresponse under 1310 nm.