

Supplementary Materials

A Broad-band Self-powered Photodetector Based on a MoTe₂/Bi₂Te₃ Heterojunction for Optical Imaging and Bias-controlled Signal Modulation

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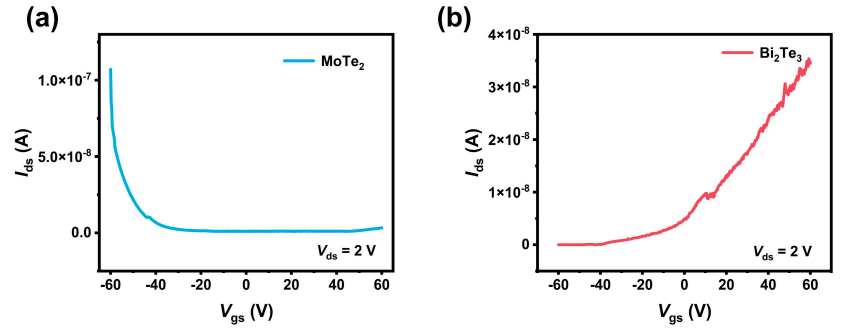


Figure S1. Transfer curves measured at $V_{ds} = 2$ V of (a) MoTe₂ and (b) Bi₂Te₃, respectively.

To demonstrate the conduction type of mechanically exfoliated materials, the wide-range transfer characteristics of the fabricated MoTe₂ and Bi₂Te₃ based devices are presented in Figures S1 (a) and (b), respectively. In Figure S1 (a), at a constant drain bias (V_{ds}) of 2 V, the source-drain current (I_{ds}) of the MoTe₂ device decreases with increasing positive gate voltage (V_{gs}), confirming its p-type behavior. Similarly, as shown in Figure S1(b), with V_{gs} scanned from -60 V to +60 V at a fixed $V_{ds} = 2$ V, the I_{ds} of the Bi₂Te₃ device decreases with increasing negative gate voltage, verifying its n-type nature.

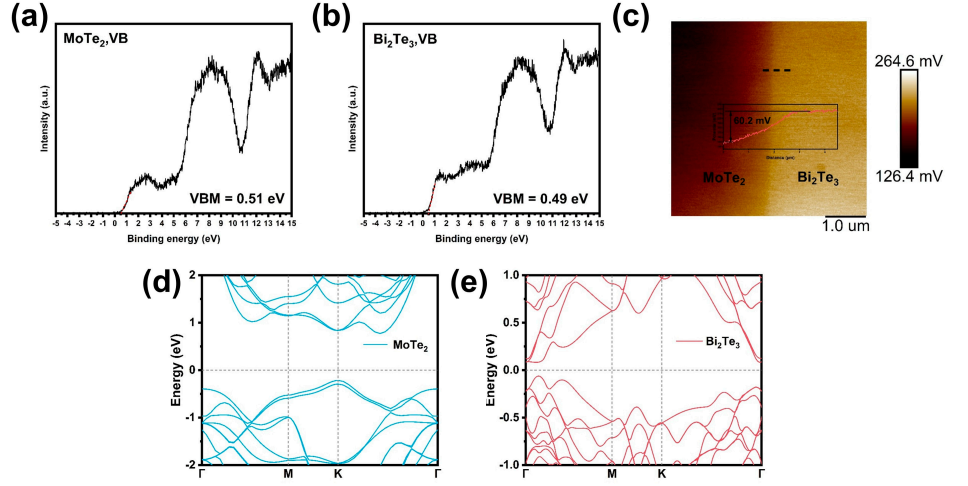


Figure S2. X-ray photoelectron spectroscopy of (a) MoTe₂ and (b) Bi₂Te₃. The intercept of X axis indicates the energy level of valence-band maximum (VBM) with respect to Fermi level of the probe having work functions of 4.56 eV. Therefore, the VBM of MoTe₂ and Bi₂Te₃ is -5.07 and -5.05 eV with respect to vacuum level, respectively. (c) The potential difference between MoTe₂ and Bi₂Te₃ with an inset of the potential height profile. The calculated band structures of (d) bulk MoTe₂ and (e) bulk Bi₂Te₃, respectively.

Kelvin probe force microscopy (KPFM) was used to measure the built-in contact potential difference (CPD) at the interface between the MoTe₂ flake and the Bi₂Te₃ flake, as shown in Figure S2(c). The surface potential distribution (SPD) of the MoTe₂ and Bi₂Te₃ flakes relative to the AFM tip can be expressed by the following equations:

$$eSPD_{MoTe_2} = W_{tip} - W_{MoTe_2} \quad (5)$$

$$eSPD_{Bi_2Te_3} = W_{tip} - W_{Bi_2Te_3} \quad (6)$$

where e is the elementary charge, and W_{MoTe_2} , $W_{Bi_2Te_3}$, and W_{tip} are the work functions of the MoTe₂ flake, Bi₂Te₃ flake, and AFM tip, respectively. The Fermi level difference ΔE_f can be calculated using the following equation.

$$\begin{aligned} \Delta E_f &= W_{Bi_2Te_3} - W_{MoTe_2} \\ &= eSPD_{MoTe_2} - eSPD_{Bi_2Te_3} \end{aligned} \quad (7)$$

As shown in the inset of Figure S2(c), the work function difference between MoTe₂ and Bi₂Te₃ is about 60.2 mV. In this work, first-principles calculations were performed based on density functional

theory (DFT), and all calculations were carried out using the Vienna Ab initio Simulation Package (VASP). The electron-ion interaction was described via the projector augmented wave (PAW) method, while the exchange-correlation energy was treated with the Perdew–Burke–Ernzerhof (PBE) functional within the framework of the generalized gradient approximation (GGA). The cutoff energy (ENCUT) for the plane-wave basis set was set to 400 eV, and the energy convergence criterion for electronic self-consistent iterations was set to 10^{-6} eV. Given that the system contains heavy elements whose relativistic effects exert a significant impact on the electronic structure, spin–orbit coupling (SOC) was further introduced to perform accurate electronic structure calculations after the convergence of the self-consistent computation. As shown in the Figure S2 (d) and (e), the calculated band structures of bulk MoTe₂ and bulk Bi₂Te₃ are presented. It can be concluded from the figure that both materials possess an indirect bandgap, with bandgap values of 0.99 eV and 0.145 eV, respectively, which are in good agreement with the corresponding bandgap values reported in previous studies [1, 2].

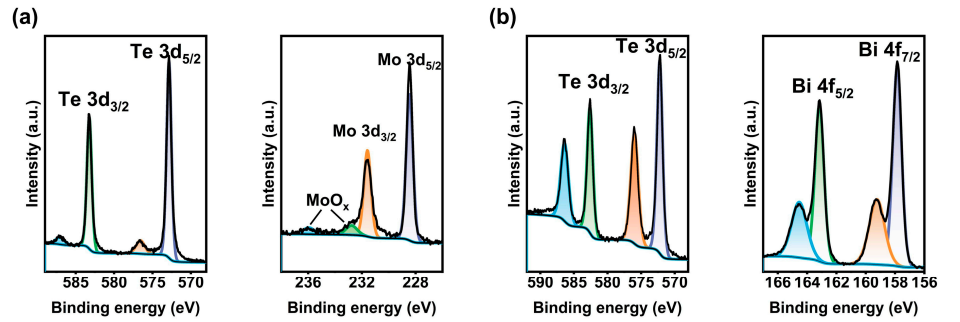


Figure S3. XPS spectrum of (a) MoTe₂ and (b) Bi₂Te₃, respectively.

Table S1. Peak assignment, FWHM and binding energy of each peak.

Name	FWHM fit param (eV)	Peak BE (eV)
Te 3d _{3/2} (Te-O)	1.36	587.01
Te 3d _{3/2} (Mo-Te)	0.78	583.22
Te 3d _{5/2} (Te-O)	1.36	576.57
Te 3d _{5/2} (Mo-Te)	0.78	572.85
Mo 3d _{3/2} (Mo-O)	1.02	235.95
Mo 3d _{5/2} (Mo-O)	1.02	232.77
Mo 3d _{3/2}	0.64	231.58
Mo 3d _{5/2}	0.62	228.45
Te ⁴⁺ (1)	1.41	586.38

Te 3d _{3/2}	1.06	582.60
Te ⁴⁺ (2)	1.39	575.98
Te 3d _{5/2}	1.06	572.22
Bi 4f _{5/2}	0.75	163.16
Bi 4f _{7/2}	0.75	157.85

Figure S3 and Table S1 present the XPS spectra of MoTe₂ and Bi₂Te₃, and the corresponding detailed quantitative data, respectively. Figure S3 (a) shows characteristic peaks of the Mo element near 230 eV in the right panel of the XPS spectrum. Specifically, two dominant peaks at 228.45 eV and 231.58 eV are assigned to Mo⁴⁺ 3d_{5/2} and Mo⁴⁺ 3d_{3/2}, respectively. In addition, a distinct characteristic peak at 235.95 eV corresponds to the Mo⁶⁺ state, forming a characteristic doublet of Mo⁶⁺ 3d_{5/2} and Mo⁶⁺ 3d_{3/2}. Notably, the Mo⁶⁺ 3d_{5/2} peak overlaps with the Mo⁴⁺ 3d_{3/2} peak. Through peak fitting, we resolve the Mo⁶⁺ 3d_{5/2} component at 232.77 eV and the Mo⁴⁺ 3d_{3/2} component at 231.58 eV, respectively. This result confirms the existence of Mo-O bonds. The formation of substitutional oxygen defects in the lattice induces hole doping, resulting in hole-dominated p-type carrier transport in the exposed MoTe₂ layer, which is in good agreement with the transfer curve of MoTe₂ in Figure S1 (a).

In the left panel of Figure S3 (a), the XPS peaks at 572.85 eV and 583.22 eV are attributed to Te 3d_{5/2} and Te 3d_{3/2} of the Mo-Te bond, respectively, while the peaks at 576.57 eV and 587.01 eV correspond to Te 3d_{5/2} and Te 3d_{3/2} of the Te-O bond, respectively [3]. In addition, the XPS characteristic peaks at 572.22 eV and 582.6 eV in Figure S3 (b) are assigned to Te 3d_{5/2} and Te 3d_{3/2}, respectively. The extra peaks at 575.98 eV and 586.38 eV originate from the oxidized Te⁴⁺ species in Bi₂Te₃. The right panel of Figure S3 (b) presents the Bi 4f spectrum of Bi₂Te₃, which exhibits two characteristic peaks at the high-binding-energy end (163.16 eV) and low-binding-energy end (157.85 eV), corresponding to the spin-orbit split Bi 4f_{5/2} and Bi 4f_{7/2} states, respectively. All the measured peak positions are in excellent agreement with previously reported results in the literature [3, 4].

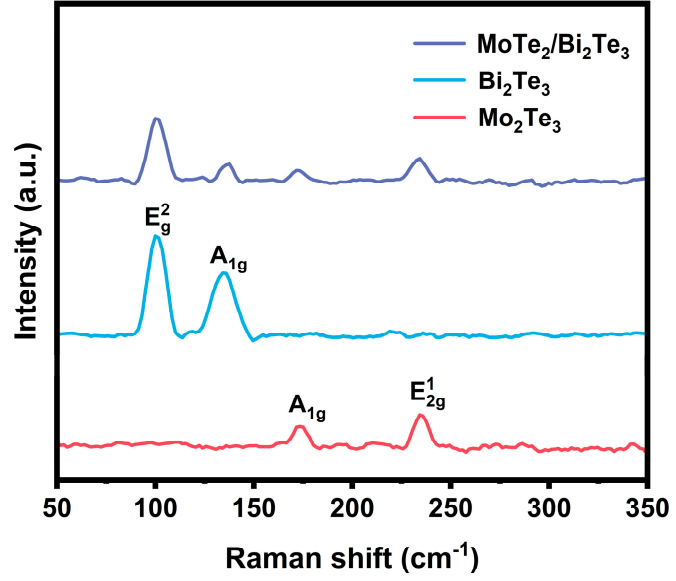


Figure S4. The Raman spectra of MoTe₂, Bi₂Te₃ and MoTe₂/Bi₂Te₃.

As shown in Fig. S4, The Raman spectrum of pristine Bi₂Te₃ (blue curve) exhibits characteristic Raman peaks at 99.93 cm⁻¹ and 134.17 cm⁻¹, corresponding to the E_g^2 and A_{1g} vibrational modes, respectively. Pristine MoTe₂ shows distinct vibrational peaks at 173.35 cm⁻¹ (A_{1g}) and 234.25 cm⁻¹ (E_{2g}^1), respectively. The Raman spectrum of the MoTe₂/Bi₂Te₃ heterostructure retains the phonon modes from both constituent materials. Notably, the reduced peak intensity is attributed to Raman quenching across the heterointerface, which is a signature of strong interlayer coupling effects [5]. Raman analysis confirms the phase composition of the MoTe₂/Bi₂Te₃ heterostructure, and is in excellent agreement with previously reported spectral data for both materials [6, 7]. It is worth noting that a slight red shift of the A_{1g} peak of MoTe₂ is observed. This phenomenon originates from the spontaneous electron transfer from Bi₂Te₃ to MoTe₂, resulting in an electron doping effect in MoTe₂. This indicates that the constituent materials in the heterostructure are free of contamination after the PVA-assisted dry transfer process [8], demonstrating the formation of a well-defined interface in the van der Waals heterostructure.

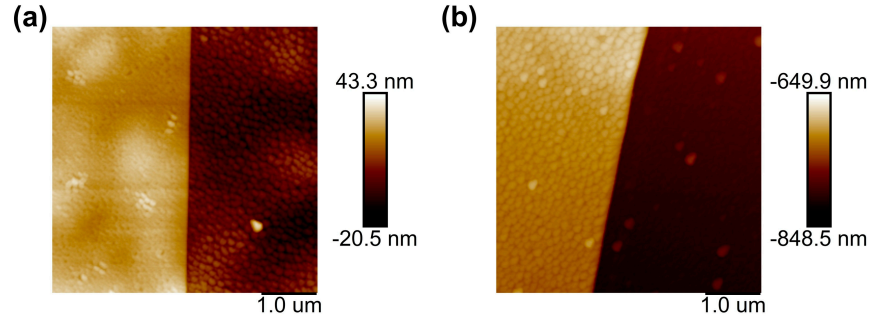


Figure S5. AFM image of (a) MoTe₂ and (b) Bi₂Te₃ interface, respectively.

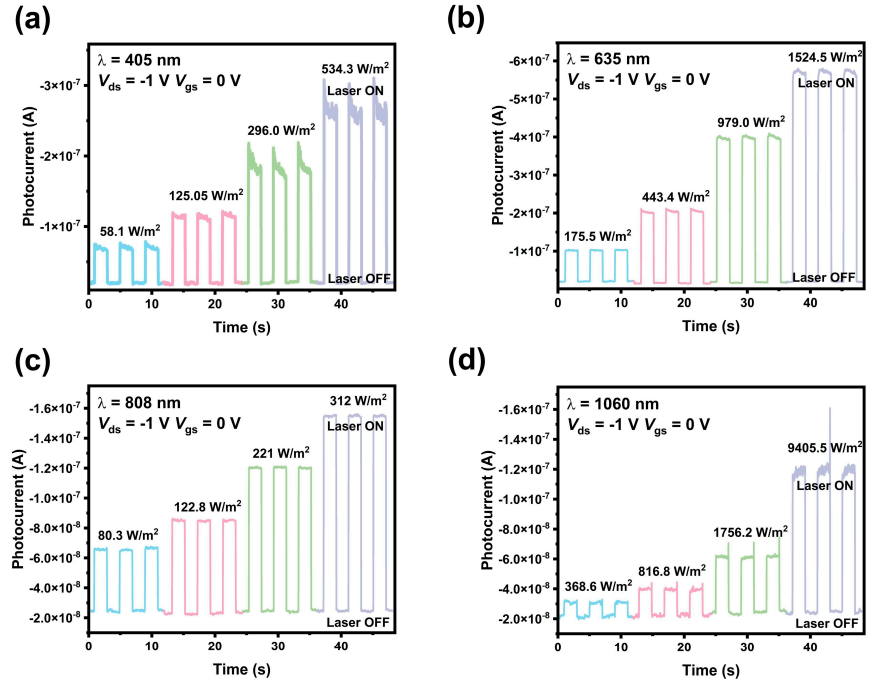


Figure S6. The I_{ds} - t response of the MoTe₂/Bi₂Te₃ heterojunction at $V_{ds} = -1 \text{ V}$ in dark and under the irradiation of (a) 405 nm, (b) 635 nm, (c) 808 nm and (d) 1060 nm laser, respectively, with different light power intensities.

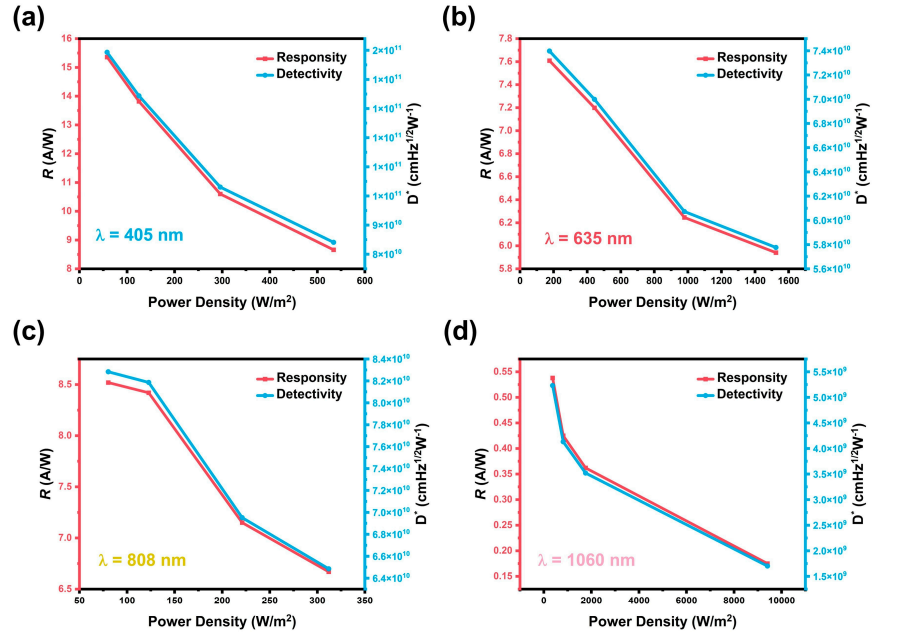


Figure S7. The light-power-dependent responsivity and detectivity under the irradiation of (a) 405 nm, (b) 635 nm, (c) 808 nm and (d) 1060 nm laser, respectively.

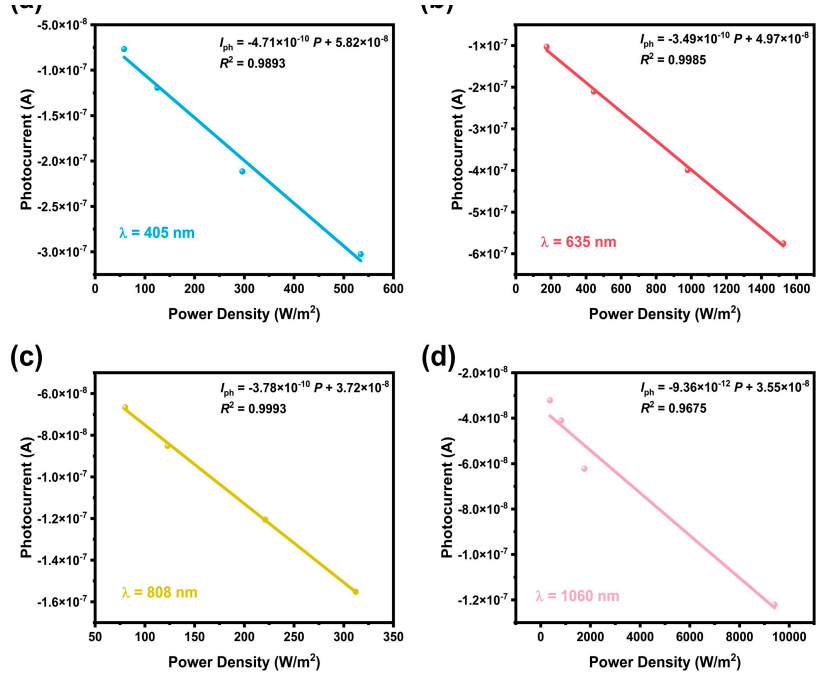


Figure S8. Photocurrent of the heterojunction as a function of incident power density at (a) 405 nm, (b) 635 nm, (c) 808 nm and (d) 1060 nm laser wavelengths, together with the corresponding linear fitting results.

Take 808 nm laser as an example, by fitting the experimental data, the photocurrent and optical power density satisfy the relation $I_{ph}(A) = -3.78 \times 10^{-10} P + 3.72 \times 10^{-8}$ ($R^2=0.999$, P has units of W/m²).

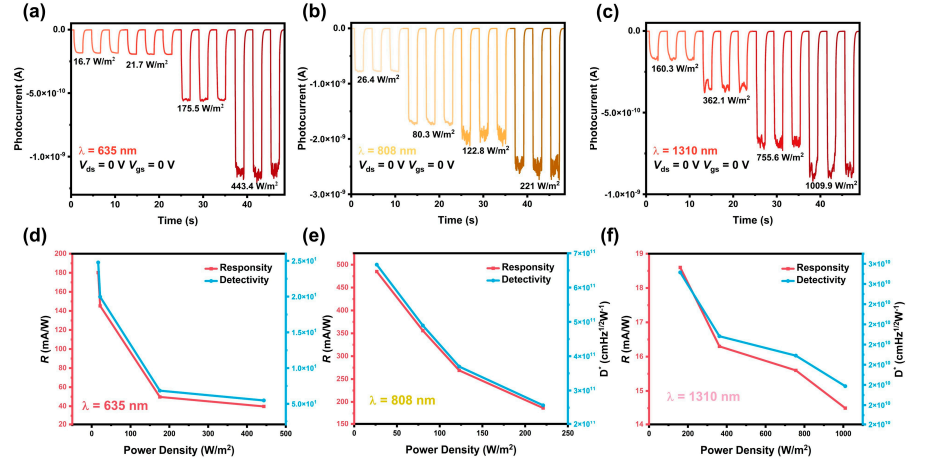


Figure S9. The I_{ds} - t response of the MoTe₂/Bi₂Te₃ heterojunction at $V_{ds} = 0$ V in dark and under the irradiation of (a) 635 nm, (b) 808 nm and (c) 1310 nm laser, respectively, with different light power intensities. The light-power-dependent responsivity and detectivity under the irradiation of (d) 635 nm, (e) 808 nm and (f) 1310 nm laser, respectively.

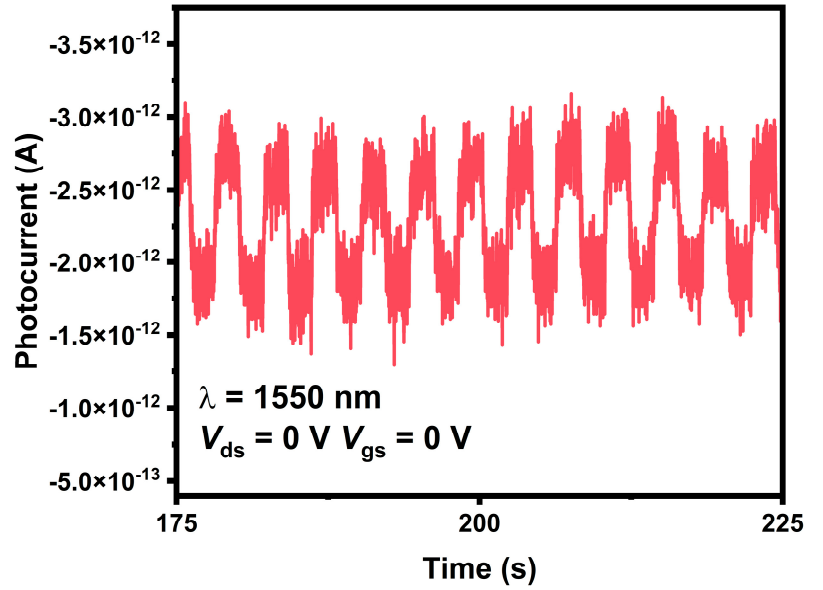


Figure S10. The I_{ds} - t response of the MoTe₂/Bi₂Te₃ heterojunction at $V_{ds} = 0$ V in dark and under the irradiation of 1550 nm.

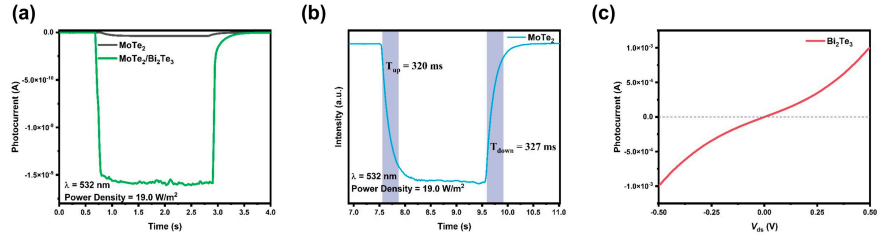


Figure S11. (a) $I_{\text{ds}} - t$ characteristics of MoTe₂/Bi₂Te₃ devices and MoTe₂ devices, measured at $V_{\text{ds}} = 0$ V and $V_{\text{gs}} = 0$ V. (b) Optical response time of MoTe₂ devices. (c) $I_{\text{ds}} - V_{\text{ds}}$ characteristics of Bi₂Te₃ at $V_{\text{gs}} = 0$ V.

To further compare the photoresponse characteristics of the heterojunction device and the pristine single-component MoTe₂ device, Figure S11 presents the photocurrent and response speed of the two devices. The results indicate that the photocurrent of the MoTe₂/Bi₂Te₃ heterojunction device is 43 times higher than that of the pristine MoTe₂ device. As shown in Figure S11 (b), the MoTe₂ device exhibits a rise time of 320 ms and a decay time of 327 ms, which are significantly slower than those of the MoTe₂/Bi₂Te₃ device (rise time: 77 ms, decay time: 75 ms). Furthermore, as shown in Figure S11 (c), the pristine single-component Bi₂Te₃ device exhibits an ultra-high dark current of approximately 1 mA at $V_{\text{ds}} = \pm 0.5$ V under zero gate voltage.

Table S2. Performance of a Self-Powered Photodetector Based on Heterojunctions.

Heterojunction	λ (nm)	R_{λ} (mA/W)	Response / decay time	Spectral response range (nm)	ref
Ga ₂ O ₃ /Bi ₂ Se ₃	365	0.175	28 ms/36 ms	254~1000	[9]
MoTe ₂ /MoS ₂	915	79	180 μ s/202 μ s	520~1550	[10]
Bi ₂ Te ₃ /Sb ₂ O ₃ /p-Si	850	316.5	24.6 ms/25.1 ms	254~1050	[11]
Gr/WSe ₂ /MoTe ₂	785	330	6.49 μ s/6.22 μ s	405~1064	[12]
Te/Bi ₂ Te ₃ /In ₂ O ₃	850	0.4195	52.8 ms/30.6 ms	850~1050	[2]
MoTe ₂ /MoWS ₄	1310	600	21.6 ms/5.8 ms	532~1550	[13]
SnSe ₂ /MoTe ₂	532	459	1.16 ms/1.14 ms	405~635	[14]
GeS ₂ /MoTe ₂	850	1090	56 ms/64 ms	520~1340	[15]
MoTe ₂ /InSe	405	433.88	99 μ s /117 μ s	405~980	[16]
ZnSnO/SnSe	365	895	0.9 ms/3.0 ms	365~670	[17]
SnSe ₂ /MoTe ₂	520	1470	101 μ s /113 μ s	520~1550	[18]
2H-MoTe ₂ /1T'-MoTe ₂	660	80.59	2.4 ms/5.2 ms	405~2200	[19]
MnS/MoS ₂	532	1310	20 ms/30 ms	405~635	[20]

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