

Supplementary Information

Bias-reconfigurable barrier photodetectors for integrated broadband sensing and neuromorphic vision

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Supplementary Note 1. Comparison of Our Work with 2D Neural Network Sensors, Optoelectronic RRAM, and Silicon-Based Platforms

1. Comparison with 2D neural network sensors

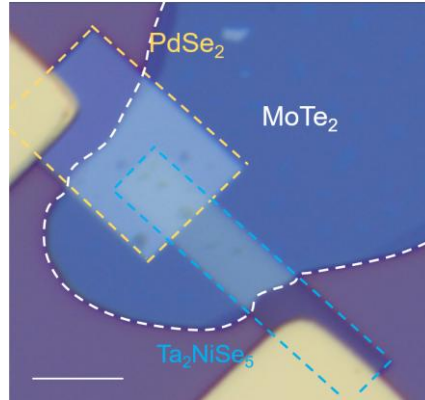
These devices perform high-speed image preprocessing by directly implementing analog convolution at the sensor front end. For instance, Mennel et al. reported a WSe₂ image sensor array in which individual devices exhibit a response time on the microsecond scale, but the photocurrent decays rapidly after light removal and lacks tunable persistent memory¹. Their subsequent 2D material-based classifier shows a response time constant of tens of microseconds, again without demonstrating long-term plasticity. Li et al. demonstrated a MoS₂-based optical synapse that emulates synaptic functions, but its response time is designed above the millisecond range and is incompatible with nanosecond-speed photodetection². Chen et al. reported a graded neuron sensor that realizes motion perception through adjustable photoresponse, yet its speed modulation range is confined to the millisecond-to-second scale³. In contrast, our device achieves a response time as short as 614 ns under low bias, suitable for high-speed image acquisition, while the decay time extends to 8.28 s under higher bias, providing long-term synaptic plasticity. The response time can be continuously tuned over seven orders of magnitude, a degree of continuous modulation not reported in the 2D neural network sensors. Furthermore, our device extends the spectral response to the mid-infrared at 3.8 μm and exhibits intrinsic polarization sensitivity, whereas the sensors mentioned above operate primarily in the visible to near-infrared range.

2. Comparison with optoelectronic RRAM

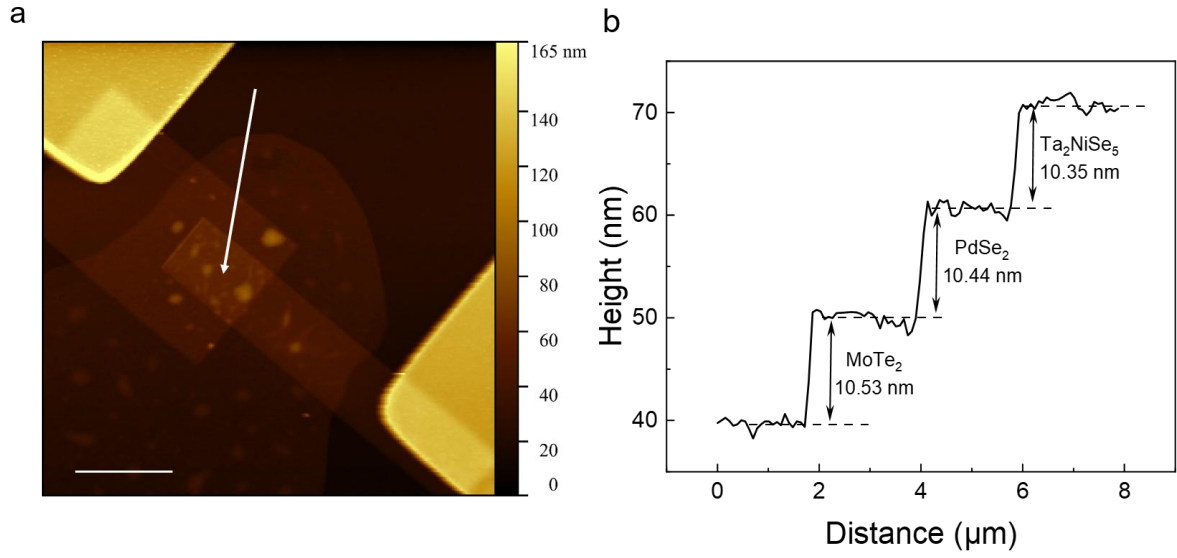
These devices emulate synaptic weights by modulating resistive states through light and electric fields. Representative works include the oxide-based optoelectronic RRAM by Zhou et al., whose conductance update time is in the microsecond-to-millisecond range and cannot achieve nanosecond response⁴. Guo et al. reported a VO₂/WO₃ heterojunction device that switches between photodetection and synaptic functions, but its response time is limited to the microsecond regime, and the two modes require distinct operating conditions rather than being dynamically and continuously tunable⁵. Chen et al. demonstrated a 2D perovskite-based optoelectronic memristor with multistate storage, yet the write/erase speeds are constrained by ionic kinetics, with typical operation times in the sub-millisecond to millisecond range⁶. These devices commonly rely on ion migration or defect trapping, which fundamentally limits their speed and leads to discrete state transitions. In comparison, our device achieves continuously tunable response time from 614 ns to 8.28 s, without the need for an electroforming process, thereby avoiding the initialization steps and uniformity issues commonly associated with RRAM.

3. Comparison with silicon-based platforms

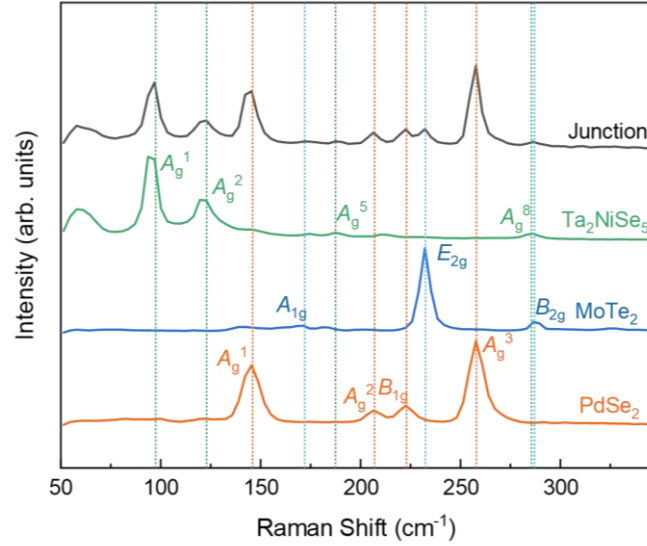
Jang et al. demonstrated in-sensor computing using electrostatically doped silicon transistors; although high speed can be achieved, external gate bias circuitry is required, and the spectral response of silicon cannot reach the mid-infrared⁷. Choi et al. reported a silicon-based floating-gate optoelectronic device capable of emulating synaptic plasticity, but weight updates also rely on external circuitry, and simultaneous broadband detection and polarization sensitivity were not demonstrated⁸. CMOS-based neuromorphic vision sensors typically require multiple transistors to emulate a single synaptic or neuronal function, in contrast to the single-device multimode reconfiguration demonstrated in this work⁹. Our device, by simply varying the drain bias, can dynamically switch between high-speed photodetection and neuromorphic processing without auxiliary gate circuitry. This simplifies pixel-level integration and provides broadband detection from the visible to the mid-infrared together with intrinsic polarization sensitivity, which are difficult to achieve on silicon platforms.



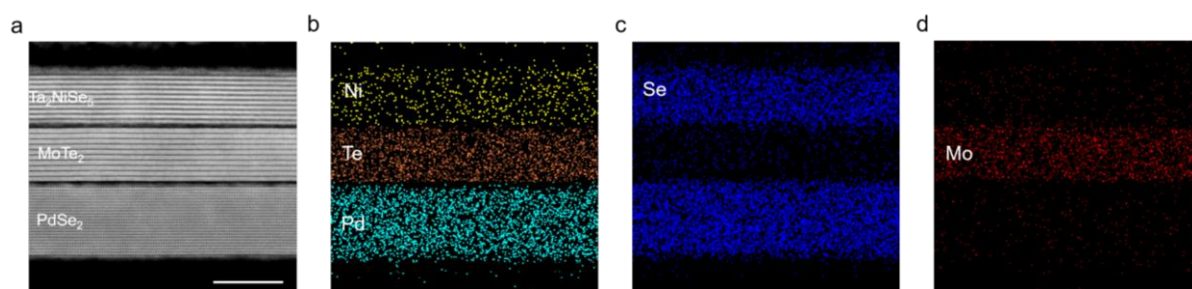
Supplementary Fig. 1 | Optical image of the $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2/\text{PdSe}_2$ vdWH device. Scale bar: 5 μm .



Supplementary Fig. 2 | AFM characterization of the Ta₂NiSe₅/MoTe₂/PdSe₂ vdWH device. AFM image (**a**) and corresponding line profiles (**b**) of the Ta₂NiSe₅/MoTe₂/ PdSe₂ heterostructure. Scale bar: 5 μm .

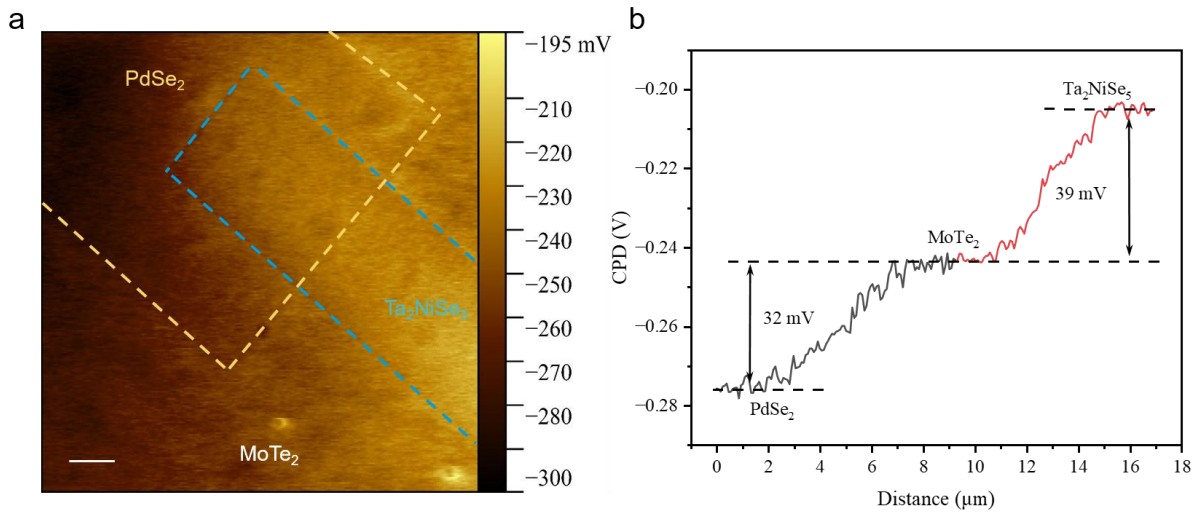


Supplementary Fig. 3 | Raman spectral characterization of Ta₂NiSe₅, MoTe₂ and PdSe₂ individual layers and the Ta₂NiSe₅/MoTe₂/PdSe₂ heterojunction. The peaks located at ~96 cm⁻¹ and ~121 cm⁻¹ correspond to the A_g^1 and A_g^2 phonon modes of Ta₂NiSe₅, respectively. The peaks at ~175 cm⁻¹, ~235 cm⁻¹ and ~291 cm⁻¹ are assigned to the A_{1g} , E_{2g} and B_{2g} phonon modes of MoTe₂, respectively, while the peaks located at ~147 cm⁻¹, ~206 cm⁻¹, ~224 cm⁻¹ and ~258 cm⁻¹ can be assigned to the A_g^1 , A_g^2 , B_{1g} and A_g^3 phonon modes of PdSe₂, respectively, consistent with prior studies¹⁰⁻¹³.

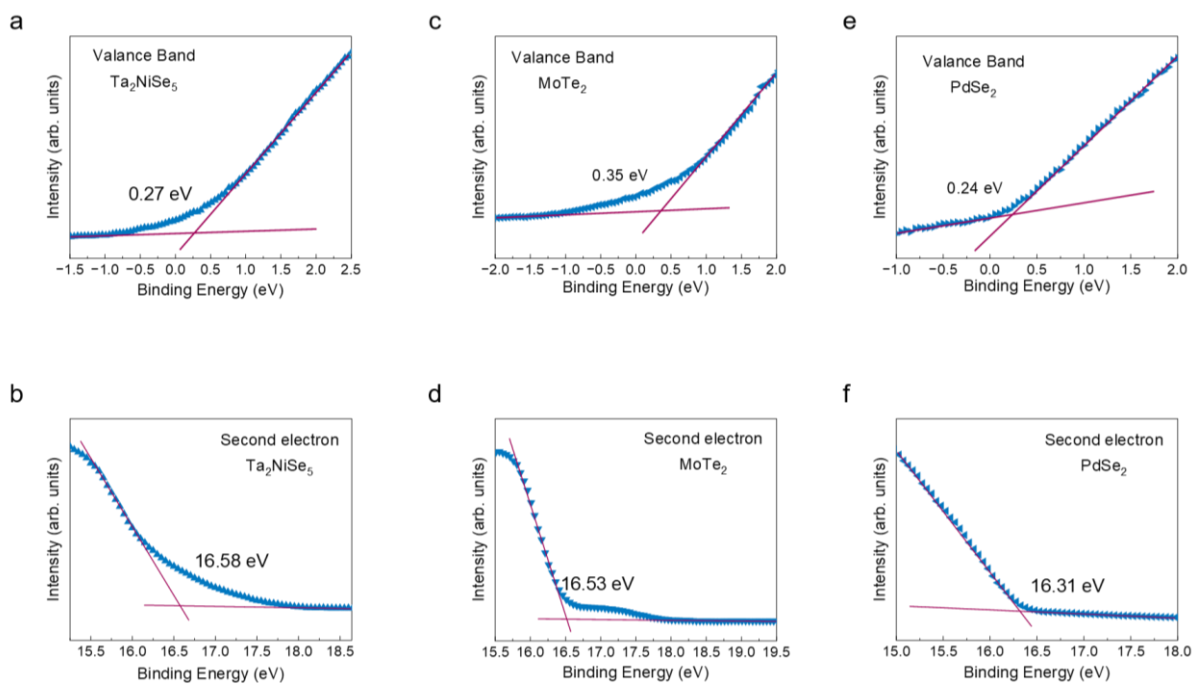


Supplementary Fig. 4 Structural and interfacial characterization of the heterostructure.

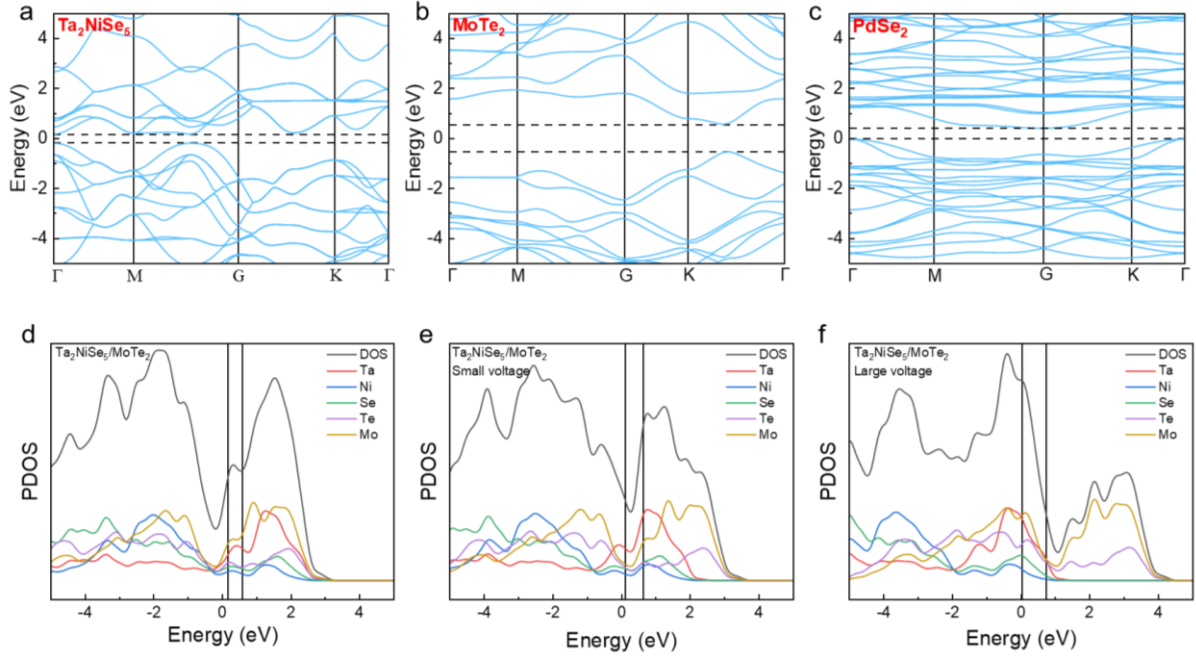
(a) Cross-sectional TEM image of the vertical heterostructure, showing a well-defined and clean interface between the stacked layers. (b-d) EDS elemental mapping of the corresponding region, illustrating the spatial distribution of the constituent elements. The elemental maps confirm a clear separation of each layer, indicating the formation of a sharp and high-quality interface. Scale bar: 10 nm.



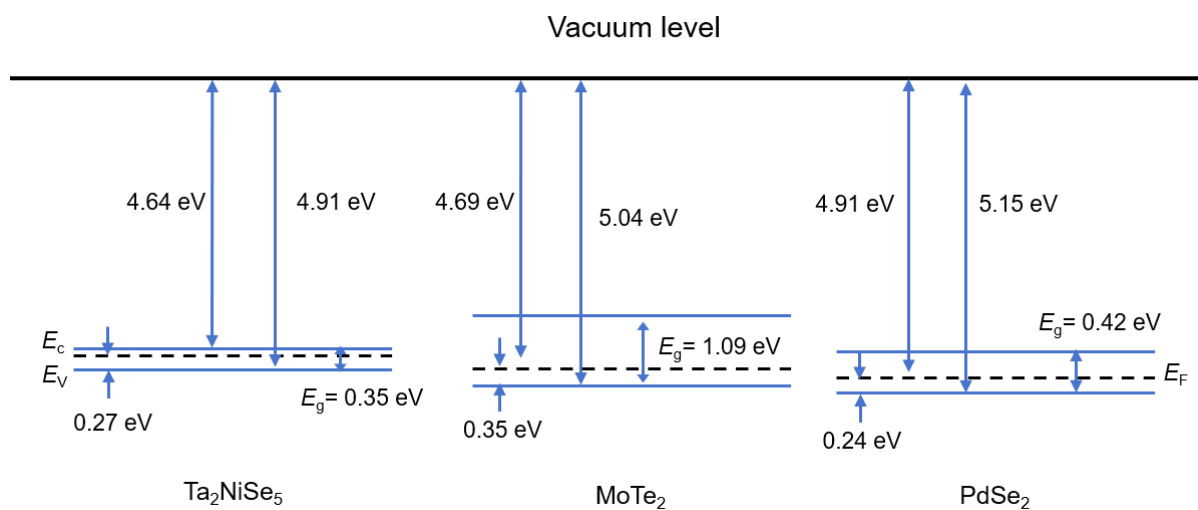
Supplementary Fig. 5 | KPFM characterization of the Ta₂NiSe₅/MoTe₂/PdSe₂ vdWH device. KPFM image (a) and surface potential difference (b) taken along the white line in a. Scale bar: 1 μm.



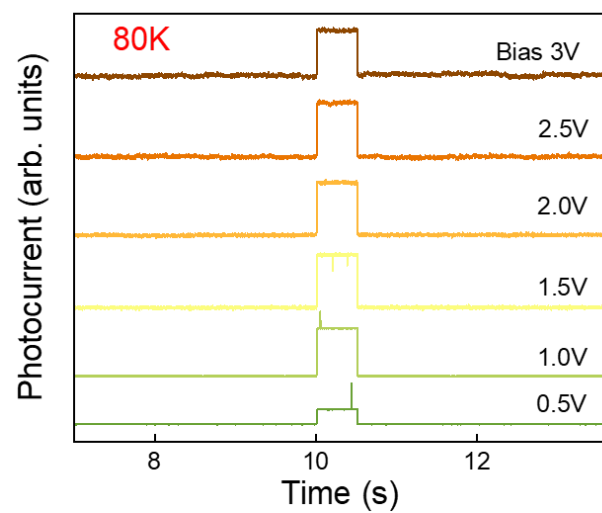
Supplementary Fig. 6 | UPS characterization of Ta_2NiSe_5 , MoTe_2 , and PdSe_2 . (a, b) Valence band spectrum and secondary electron cutoff of Ta_2NiSe_5 . (c, d) Valence band spectrum and secondary electron cutoff of MoTe_2 . (e, f) Valence band spectrum and secondary electron cutoff of PdSe_2 .



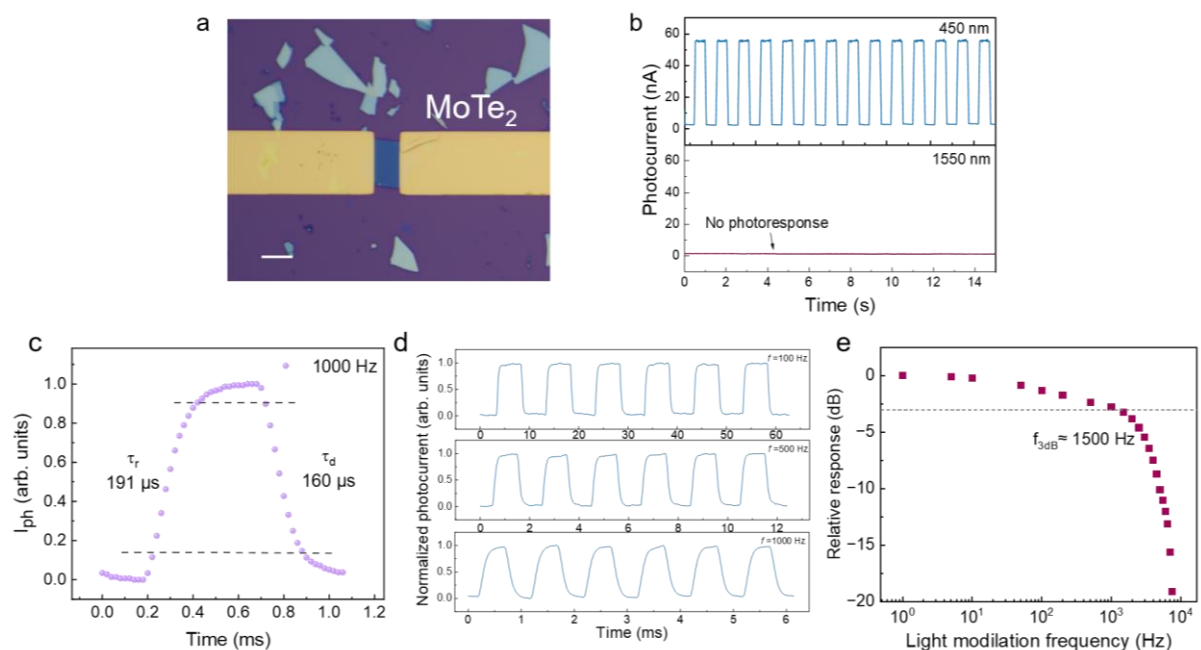
Supplementary Fig. 7 | Band structures and density of states of the heterostructure. (a–c) Band structures of the individual Ta_2NiSe_5 , MoTe_2 , and PdSe_2 , with the valence band maximum and conduction band minimum indicated. (d–f) Projected density of states of the $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2$ heterostructure under zero bias, small bias, and large bias, showing the total DOS and element-projected contributions. Energy is referenced to the Fermi level in all panels.



Supplementary Fig. 8 | Energy band profiles of Ta_2NiSe_5 , MoTe_2 , and PdSe_2 .

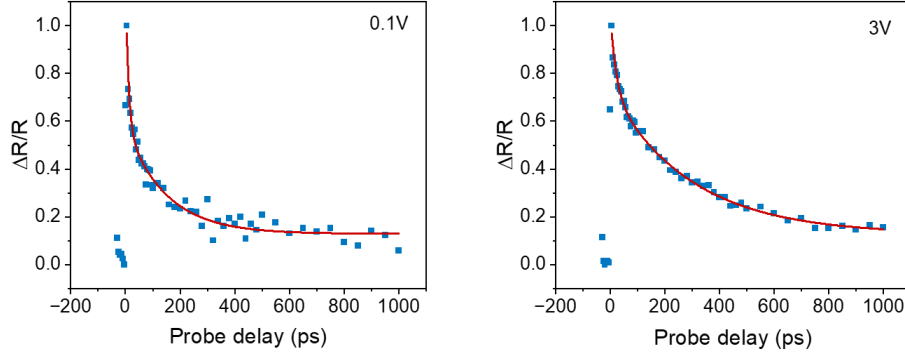


Supplementary Fig. 9 Bias-dependent photocurrent measured at 80 K.



Supplementary Fig. 10 | Photoresponse of the MoTe₂ device. (a) Optical image of the MoTe₂ device. (b) Time-resolved photocurrent measured under 1550 nm and 450 nm illumination. Scale bar: 10 μm . (c) Response time of the device. (d) Photocurrent under different modulation frequencies. (e) -3 dB cutoff frequency.

Here, we fabricated a single-MoTe₂ device and confirmed that it shows no photoresponse at 1550 nm. In stark contrast, our Ta₂NiSe₅/MoTe₂/PdSe₂ heterostructure clearly exhibits bias-dependent switching between fast and slow response at 1550 nm, a wavelength at which MoTe₂ cannot efficiently generate photocarriers. This demonstrates that the tunable response speed does not primarily rely on photogeneration within the MoTe₂ layer. Moreover, we measured the temporal response of a standalone MoTe₂ photodetector under 633 nm illumination. The rise and fall times are 191 μs and 160 μs , respectively, and the -3 dB bandwidth is only ~ 1.5 kHz. These values are approximately three orders of magnitude slower than the 257 ns / 274 ns response of the heterostructure at low bias. If photogenerated carriers in MoTe₂ were dominating the photoresponse of the heterostructure, the overall speed would be limited by the slow MoTe₂ component and would struggle to reach the nanosecond regime.

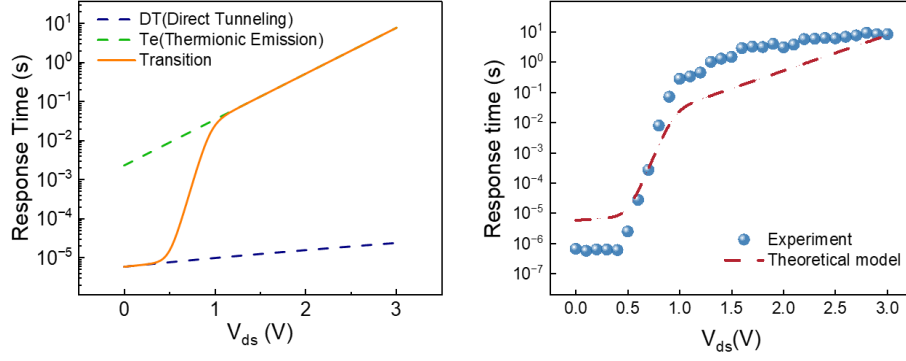


Supplementary Fig. 11 Pump-probe measurements of the device under low bias and high bias.

The decay curves were fitted with a standard bi-exponential function:

$$\frac{\Delta R}{R}(t) = A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right) + y_0 \quad (1)$$

where the fast component (A_1 , τ_1) can be understood as the efficient extraction/escape of photogenerated carriers from the Ta_2NiSe_5 absorption layer across the interface, while the slow component (A_2 , τ_2) corresponds to the trapping of carriers at interface states or barrier-localized states. These trapped carriers may serve as a reservoir for the subsequent PPC. The experimental results show that as the bias increases from 0 V to 3 V, the fast amplitude A_1 decreases markedly from 0.65 to 0.31, while the slow amplitude A_2 increases from 0.45 to 0.60. Accordingly, the relative weight of the slow channel, $W = A_2/(A_1 + A_2)$, rises from ~41% to ~66%. Simultaneously, the slow time constant τ_2 lengthens from 147 ps to 297 ps. The variations of these three parameters provide spectroscopic evidence suggesting that the elevated bias raises the interfacial barrier, which significantly suppresses direct carrier extraction while increasing both the population and the release time of trapped carriers.



Supplementary Fig. 12. Calculated bias-dependent response time from the model.

Based on the above physical picture, we have established a three-channel parallel model comprising direct tunneling, thermal excitation, and trap-assisted processes. The total effective response time τ is determined by the sum of the rates of the three channels:

$$\frac{1}{\tau(V)} = \frac{1}{\tau_{DT}(V)} + \frac{1}{\tau_{TE}(V)} + \frac{1}{\tau_{trap}(V)} \quad (2)$$

When the DT channel is open, its rate far exceeds those of the TE and trap channels, so $\tau \approx \tau_{DT}$; when the DT channel is suppressed by the bias, τ approaches τ_{TE} and τ_{trap} , with both TE and trap-assisted processes jointly contributing to the slow response observed at high bias.

(1) Direct tunneling channel

According to the Simmons approximation, the carrier transmission rate for direct tunneling can be expressed as:

$$R_{DT} \propto \exp\left(-\frac{4\pi d}{h} \sqrt{2m^* \phi(V)}\right) \quad (3)$$

Hence, the DT response time, being inversely proportional to the tunneling probability, is:

$$\tau_{DT}(V) = \tau_0 \cdot \exp\left(\frac{4\pi d}{h} \sqrt{2m^* \phi(V)}\right) \quad (4)$$

Where m^* is the hole effective mass, h is Planck's constant, $\phi(V) = \phi_0 + \alpha V$ is the bias-dependent barrier height, with α extracted from the temperature-dependent measurements. d denotes the effective tunneling distance.

(2) Thermal excitation channel

The carrier transport via the TE channel follows a thermal activation law, and its response time can be written as:

$$\tau_{TE}(V) = \tau_0 \cdot \exp\left(\frac{\phi(V)}{k_B T}\right) \quad (5)$$

where k_B is the Boltzmann constant. The prefactor τ_0 is calibrated from the high-bias PPC decay curve.

(3) Trap-assisted channel and the DT-to-TE transition

In the low-bias region ($V < 0.5$ V), the DT channel dominates and the trap channel is effectively shorted out, because the tunneling time is much shorter than typical trap capture and release times. At high bias ($V > 0.5$ V), the DT channel is strongly suppressed, and the response is governed jointly by TE and trap-assisted processes. Carriers need to slowly overcome the barrier via TE and drift in the widened depletion region, where traps are activated by the strong electric field. The slow release of trapped carriers further prolongs the decay time, giving rise to the observed persistent photoconductivity.

With increasing bias, the dominant carrier transport mechanism gradually transitions from DT to TE. This is a gradual transition rather than an abrupt switch. We use the sigmoidal switching function to describe this transition.

$$f(V) = \frac{1}{1 + \exp(-\gamma(V - V_{th}))} \quad (6)$$

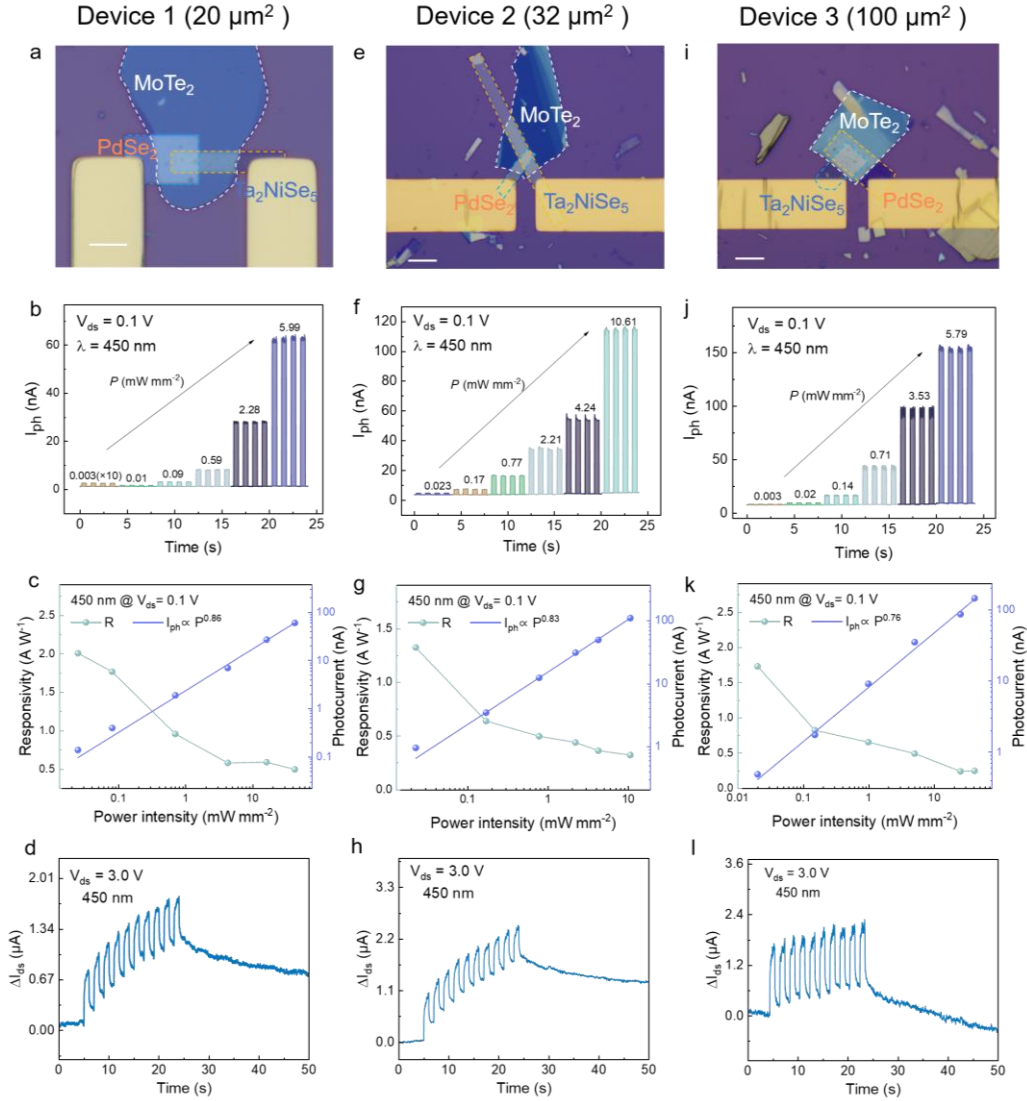
Here, $f(V)$ represents the probability that charge transport occurs via the thermal excitation channel, while $1 - f(V)$ corresponds to the tunneling contribution.

The DT-to-TE transition is essentially a competition between two carrier transport probabilities under continuous bias modulation, and the mathematical characteristic of the sigmoidal function, a smooth transition between two saturation limits, naturally captures this gradual process. Similar approaches have been reported in related device systems. For instance, in ion-migration-based memristors, the thermally activated hopping of ions over a double-well potential gives rise to a switching probability described by a Boltzmann factor, naturally resulting in a sigmoidal macroscopic response¹⁴. In ferroelectric tunnel junctions, the polarization reversal modulates the barrier height, and the resulting tunneling current follows a sigmoidal dependence that can be derived from the Landau-Ginzburg-Devonshire free energy framework¹⁵. In resistive switching devices, conductive filament formation and rupture have also been described using sigmoidal functions¹⁶⁻¹⁷. In our device, the carrier transport probability across the barrier is determined by the competition between field-driven tunneling and thermally activated emission, both of which respond continuously to bias-induced barrier modulation. This physical picture is analogous to those systems. We therefore adopt this approach and use the sigmoidal function to describe the smooth transition between the two transport mechanisms. Its limiting behavior is consistent with experimental observations: DT dominates at low bias, TE dominates at high bias, and both mechanisms coexist in the intermediate region.

By substituting the experimentally determined parameters into the above equation, we calculated the theoretical response time over the full bias range. $\tau_0 = 257$ ns, which is the measured response time at 0.1 V bias. The effective mass m^* was taken as $0.6 m_0$ from reported values for few-layer MoTe₂. The tunneling distance d was set to 10.5 nm, the physical thickness of the MoTe₂ layer determined by AFM. The zero-bias barrier height Φ_0

was determined to be 0.18 eV from the Arrhenius fit of the dark current. The modulation coefficient α was determined to be 0.07 eV/V from the Arrhenius analysis of the photocurrent as a function of bias. The transition voltage V_{th} and the steepness factor γ were determined to be 0.5 V and 18 V⁻¹, respectively, from the sigmoidal fit to the transition region.

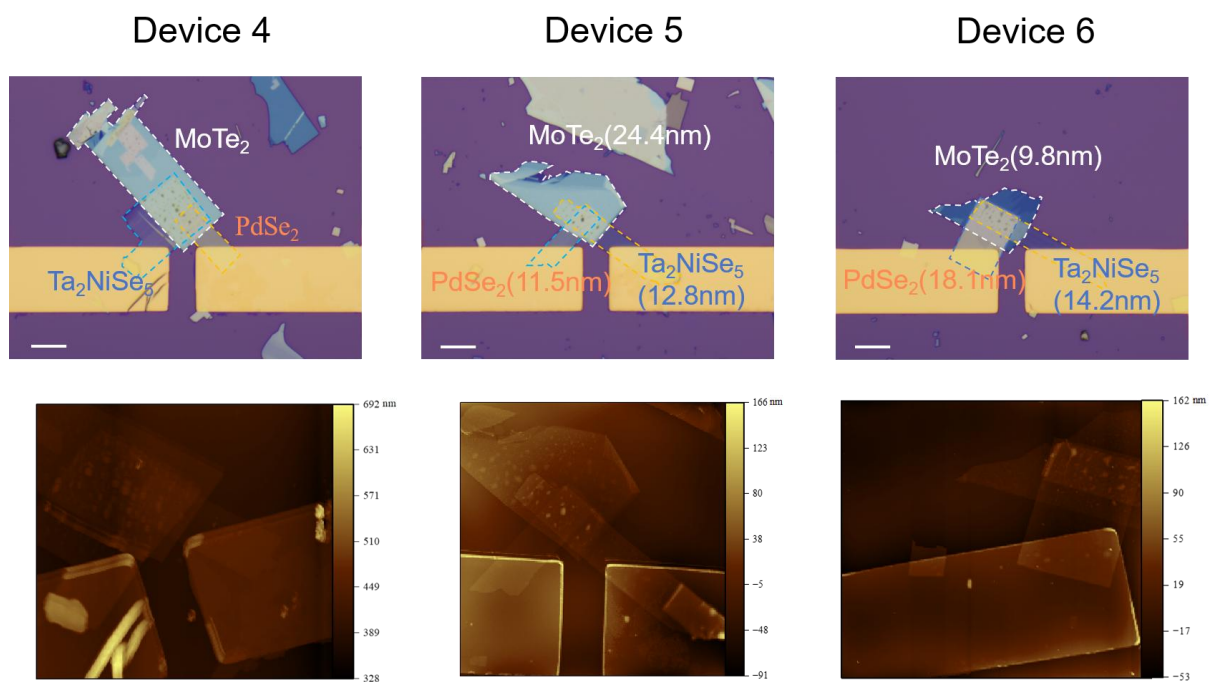
The theoretical curves and experimental data are shown together in Supplementary Fig. 12. Overall, the model captures the general trend of the bias-dependent response time. In the low-bias region, the theoretical values are slightly higher than the experimental data, i.e., the model predicts a longer response time. This is mainly because the tunneling distance d was taken as the physical thickness of the MoTe₂ layer, 10.5 nm. However, in an vdW heterostructure, carrier tunneling preferentially occurs at the narrowest point of the interfacial barrier, and the effective tunneling width is jointly determined by the van der Waals gap and near-interface band bending, which may be considerably smaller than the overall physical thickness of the MoTe₂ layer. Using a larger d in the model leads to an underestimation of the DT tunneling probability, which in turn overestimates the response time in this bias region. In the high-bias region, the experimental response time is longer than the theoretical prediction, exhibiting a slower decay process. This is mainly because the current model considers only two transport channels, pure DT and pure TE, whereas in the actual device, defect states present in the MoTe₂ layer and at the heterointerfaces can contribute to carrier transport, introducing additional time delays that prolong the response. These defect-assisted processes are not captured by the current model, which likely accounts for the deviation in the high-bias regime.



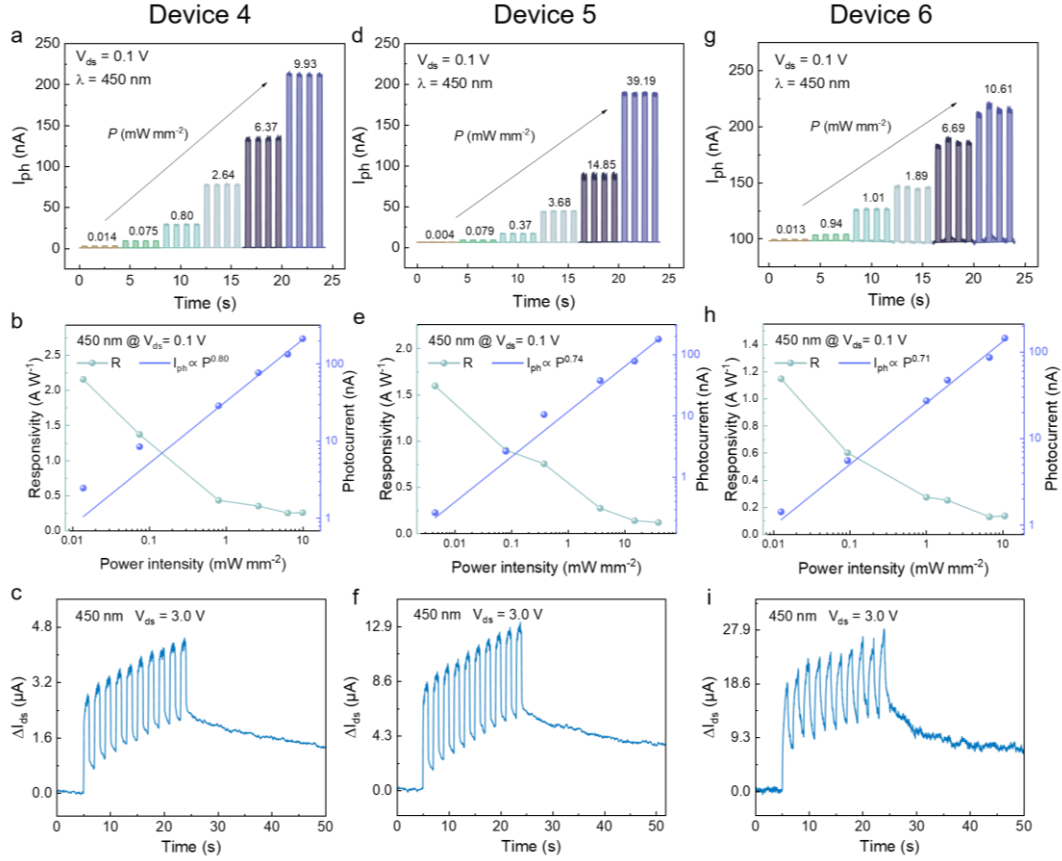
Supplementary Fig. 13 | Photoresponse characteristics of the device in different areas.

(a, e, i) Optical microscopy images of the 3 devices. Scale bars: 5 μm (a); 10 μm (e, i). (b, f, j) Low-bias time-resolved switching response under 450 nm illumination of the 3 devices, showing stable and repeatable photocurrent modulation. (c, g, k) Photocurrent and responsivity as a function of incident optical power density under 450 nm illumination of the 3 devices. (d, h, l) Photocurrent response under higher bias, revealing a persistent photocurrent (PPC) effect with slow decay behavior.

Here, the dark current at $V_{ds} = 0.1$ V increases with increasing area, from ~ 1.32 nA at $20 \mu\text{m}^2$ to ~ 3.8 nA at $32 \mu\text{m}^2$ and further to ~ 8 nA at $100 \mu\text{m}^2$, which is consistent with previous reports on area-dependent dark current scaling in photodetectors. The corresponding responsivities at 450 nm are 2.0 A/W, 1.3 A/W, and 1.7 A/W, respectively, which are overall close to each other. Importantly, all devices with different junction areas exhibit the same bias-dependent fast-to-slow switching behavior, confirming that the bias-controlled behavior is an intrinsic property of the heterojunction rather than being affected by device area.



Supplementary Fig. 14 | Optical microscopy images and corresponding atomic force microscopy (AFM) images of three Ta₂NiSe₅/MoTe₂/PdSe₂ heterostructure devices with different layer thicknesses. Scale bars: 10 μ m.

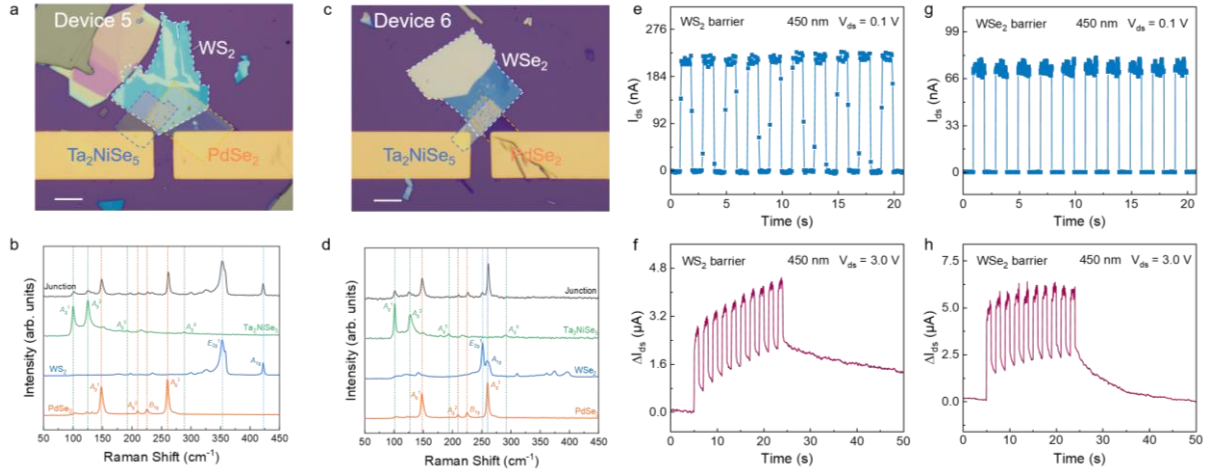


Supplementary Fig. 15 | Photoresponse characteristics of devices with different thicknesses. (a, d, g) Low-bias time-resolved switching response under 450 nm illumination of the 3 devices, showing stable and repeatable photocurrent modulation. (b, e, h) Photocurrent and responsivity as a function of incident optical power density under 450 nm illumination of the 3 devices. (c, f, i) Photocurrent response under higher bias, revealing a pronounced PPC effect with slow decay behavior.

Here, the thickness of the Ta₂NiSe₅ absorption layer and PdSe₂ contact layer were kept approximately constant across all devices, while the thickness of the MoTe₂ barrier layer was systematically varied. The dark current at $V_{ds} = 0.1$ V decreases with increasing MoTe₂ thickness: from ~98 nA at 8.96 nm to ~9.1 nA at 17.08 nm, and further to ~3.12 nA at 23.32 nm. This suppression is consistent with the exponential reduction of tunneling probability in thicker barriers, as described by:

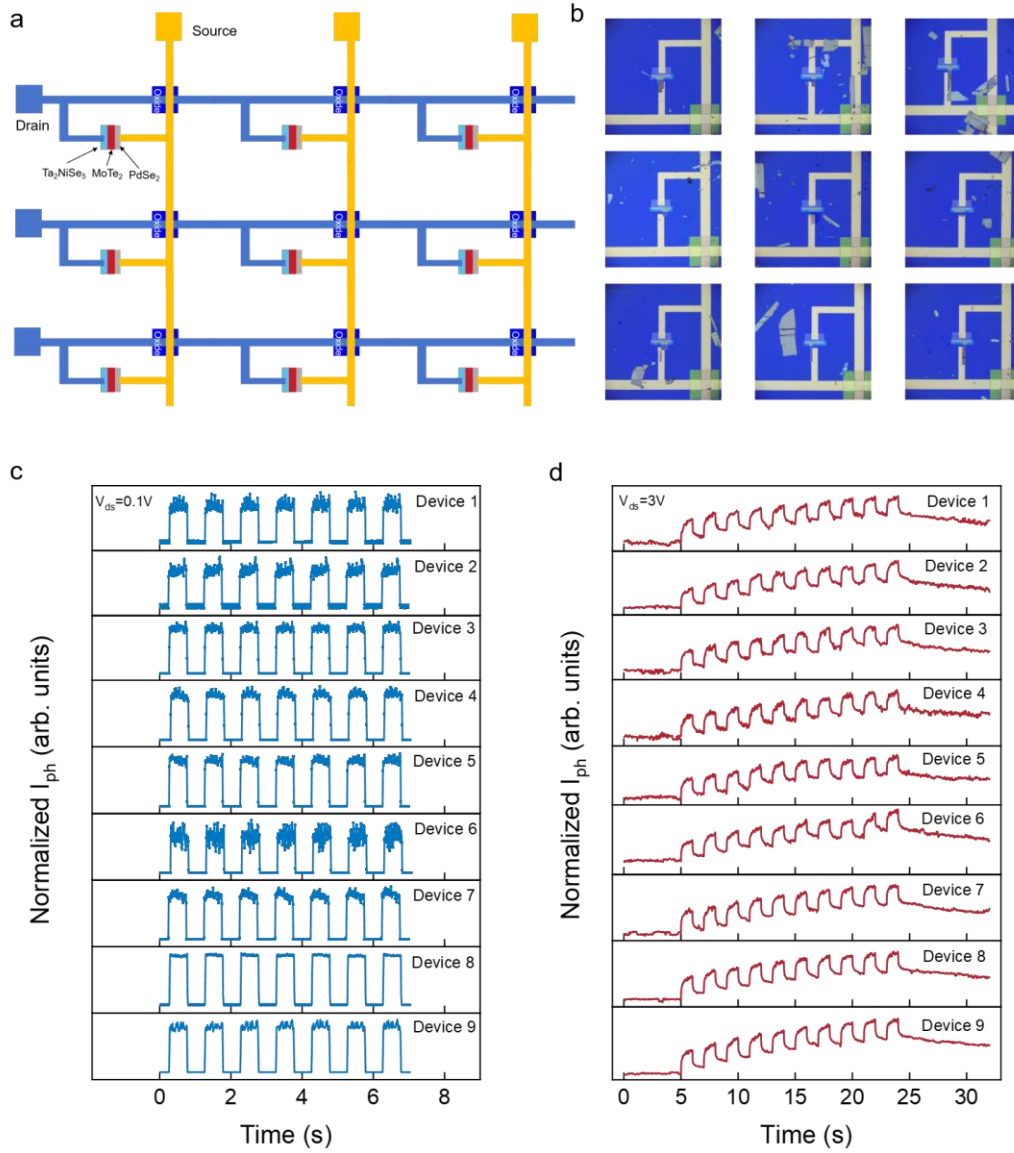
$$I_{DT} \propto \exp\left(-\frac{4\pi d\sqrt{2m^*\phi}}{h}\right) \quad (7)$$

where d is the barrier thickness, m^* is the effective mass, ϕ is the barrier height, and h is Planck's constant. As the MoTe₂ thickness increases, the tunneling distance increases, leading to an exponential decrease in the direct tunneling current. The corresponding responsivities at 450 nm are 1.1 A/W, 1.6 A/W, and 2.1 A/W, respectively, which are overall close to each other. Importantly, all devices with different MoTe₂ thicknesses exhibit the same bias-dependent fast-to-slow switching behavior.

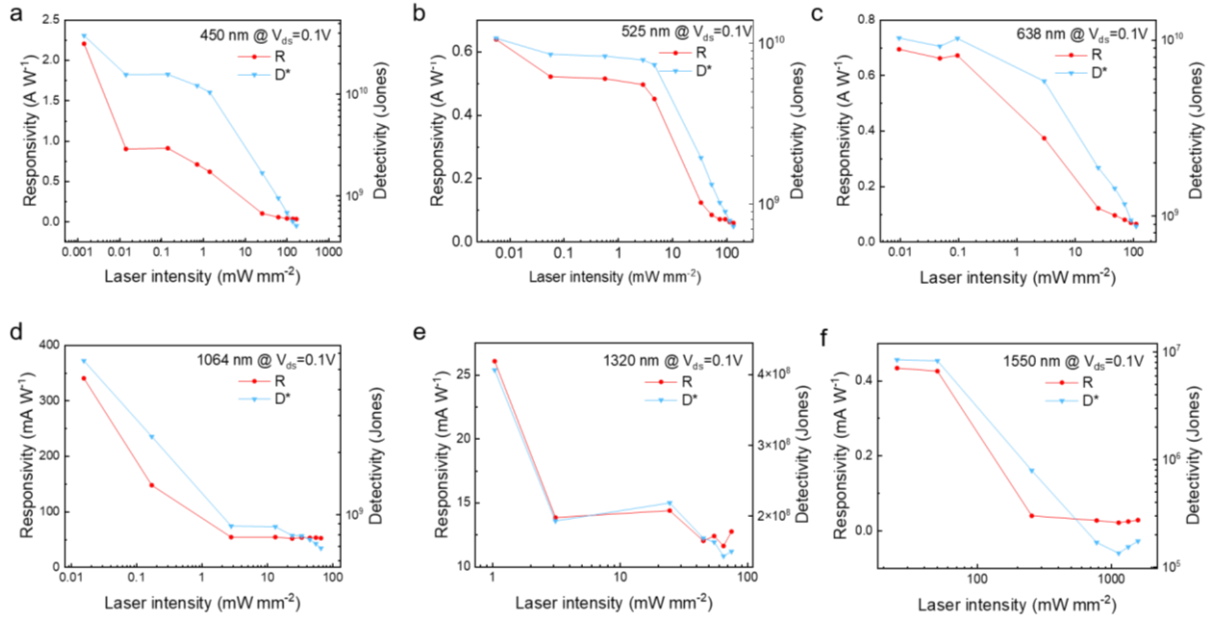


Supplementary Fig. 16 | Photoresponse characteristics of the Ta₂NiSe₅/WS₂/PdSe₂ and Ta₂NiSe₅/WSe₂/PdSe₂ devices. (a-d) Optical image and Raman spectrum of the Ta₂NiSe₅/WS₂/PdSe₂ and Ta₂NiSe₅/WSe₂/PdSe₂ device. (e, g) Low-bias time-resolved switching response under 450 nm illumination of the 2 devices, showing stable and repeatable photocurrent modulation. (f, g) Photocurrent response under higher bias, showing a pronounced PPC effect with slow decay behavior. Scale bars: 10 μm.

Here, we replaced the MoTe₂ barrier with alternative 2D materials, specifically WS₂ and WSe₂, while keeping the Ta₂NiSe₅ absorption layer and the PdSe₂ contact layer unchanged. Remarkably, these heterostructures also exhibit the same bias-dependent transition: fast photoresponse under low bias and slow, persistent response under high bias. The observation of similar phenomena in devices with chemically distinct barrier materials, including MoTe₂, WS₂, and WSe₂, indicates that the underlying mechanism is not limited to a specific material combination but rather arises from a more general band engineering principle applicable to a broad class of vdW heterostructures with suitable band alignment.

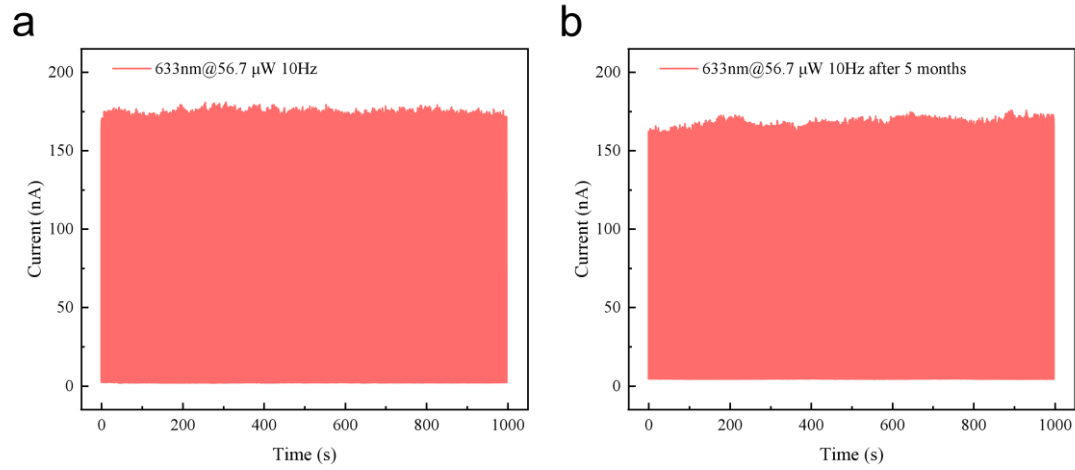


Supplementary Fig. 17 | 3×3 device array and bias-dependent photoresponse. (a) Schematic illustration of the 3×3 device array based on the $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2/\text{PdSe}_2$ heterostructure. (b) Optical image of the 3×3 device array. (c-d) The photoresponse under small bias and large bias.

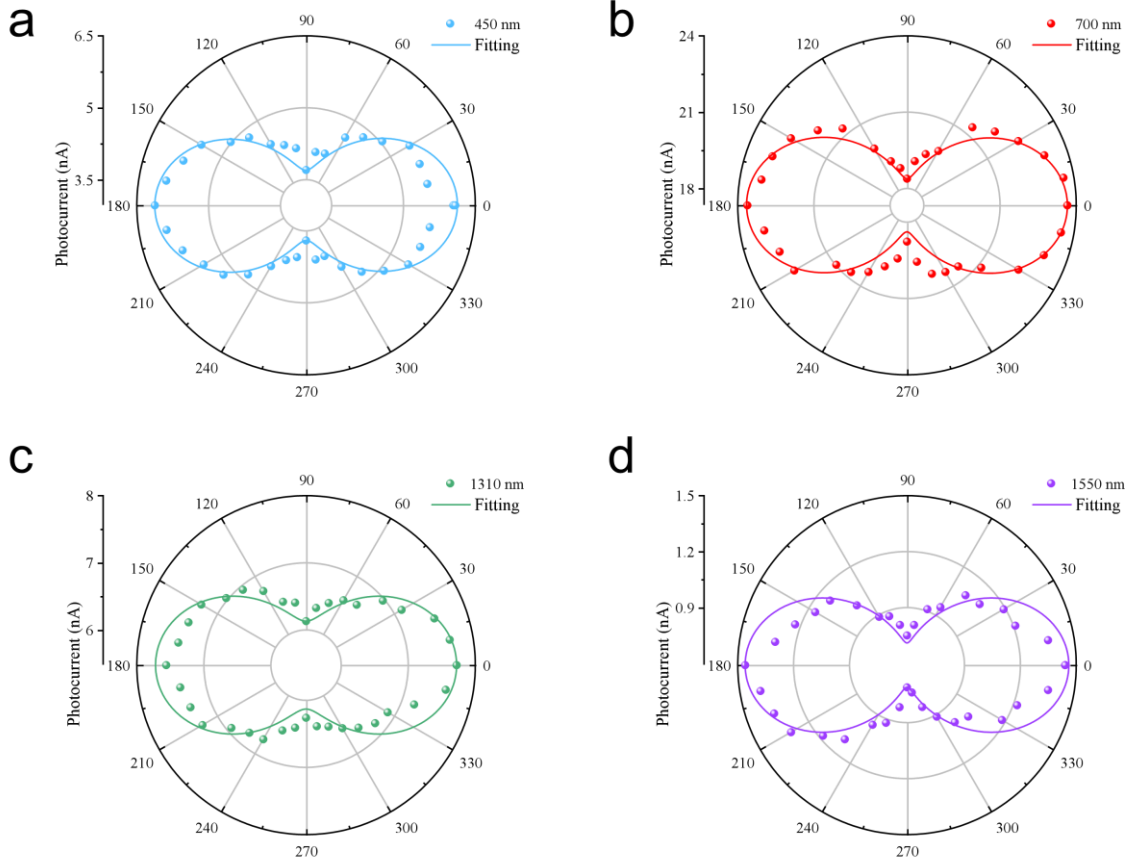


Supplementary Fig. 18 | Device characterization under different light illumination at $V_{ds} = 0.1$ V. physical thickness, responsivity and detectivity at (a) 450 nm, (b) 525 nm, (c) 638 nm, (d) 1064 nm, (e) 1320 nm, (f) 1550 nm.

Here, we have extracted the key figures of merit at $3.8 \mu m$ from the measurements: a responsivity of approximately $0.3 mA/W$, a specific detectivity of approximately 3.9×10^6 Jones, and a response time of approximately $39.7 ms$. Because $3.8 \mu m$ lies near the absorption edge of Ta_2NiSe_5 , the photoresponse at this wavelength is relatively weak under low bias.



Supplementary Fig. 19 | Stability of the Ta₂NiSeS/MoTe₂/PdSe₂ vdWH device. Time-resolved photoresponse of (a) as-fabricated device and the same device (b) after five-month storage. Measured at a wavelength of 633 nm with incident light power of 56.7 μW at $V_{ds} = 0.1$ V. All the measurements were conducted and ambient pressure at room temperature.

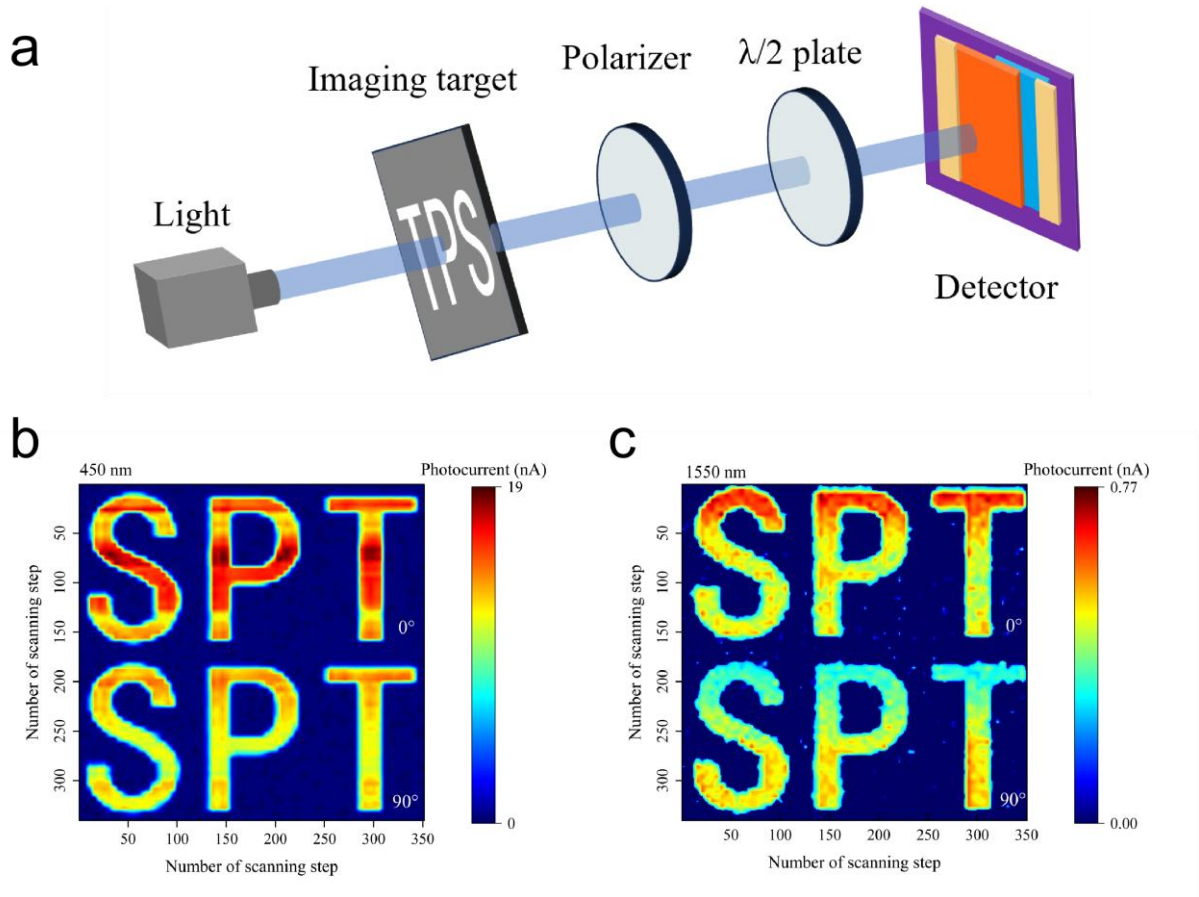


Supplementary Fig. 20 | Photoresponse of the vertical channel device as a function of polarization angle under different light illumination. The polar plots of polarized photocurrents under (a) 450 nm, (b) 700nm, (c) 1310nm and (d) 1550 nm at $V_{ds} = 0.1$ V.

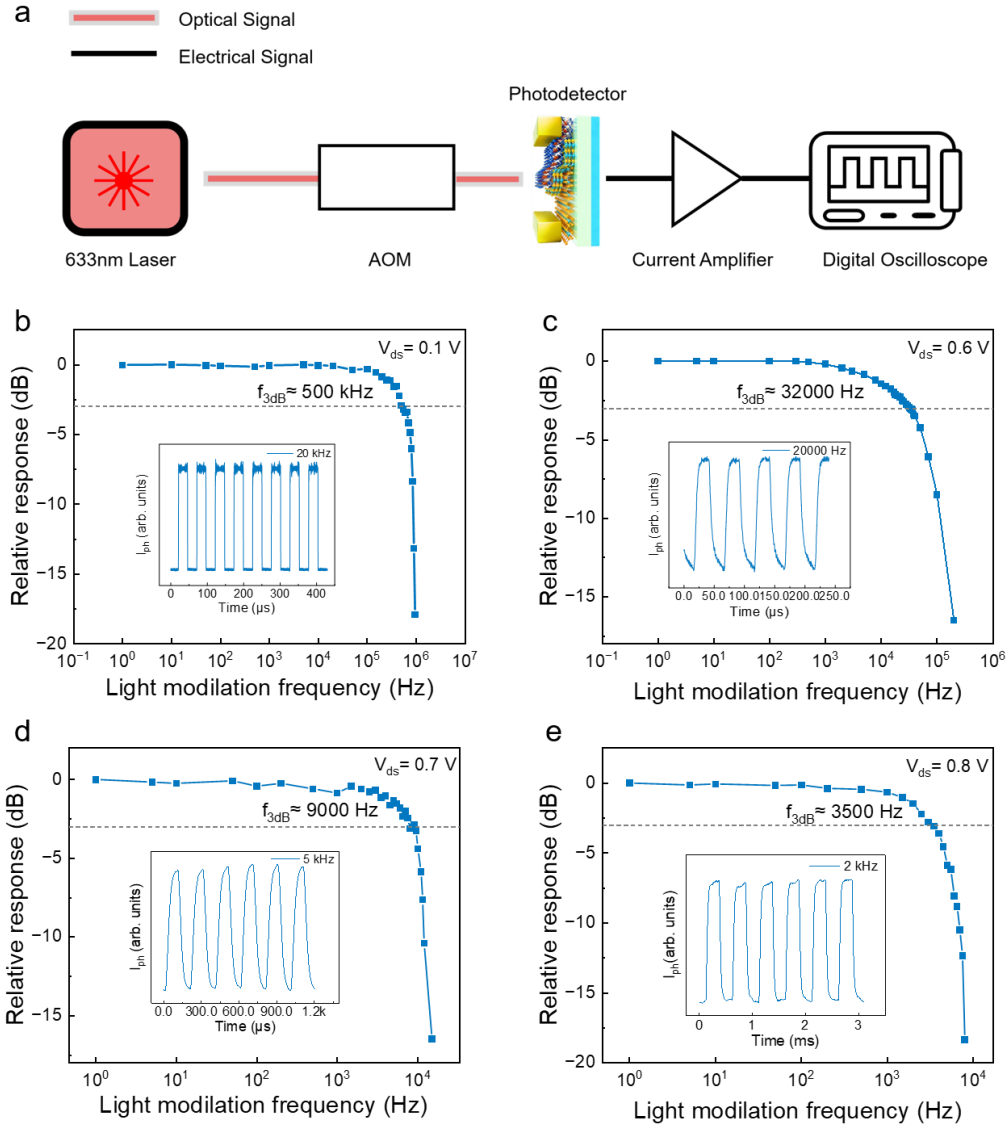
The fitting curve function can be expressed as¹⁸:

$$I_{ph}(\theta) = I_{phmax} \cos^2(\theta + \varphi) + I_{phmin} \sin^2(\theta + \varphi) \quad \text{where } I_{phmax} \text{ represent the maximum}$$

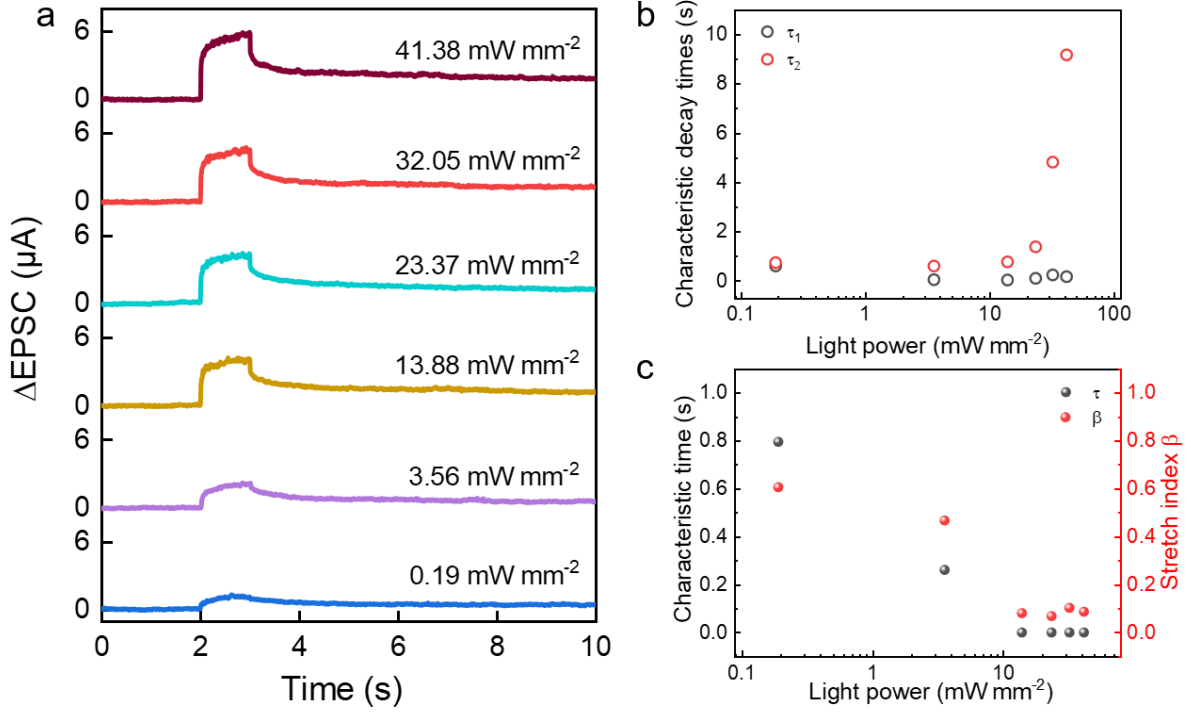
photocurrent and I_{phmin} represent the minimum. φ is the fitting parameter. The wavelength-dependent polarization ratio reflects the combined effects of the wavelength-dependent absorption distribution among the layers and the photon-energy-dependent linear dichroism of each anisotropic material.



Supplementary Fig. 21 | Imaging sensing capability of the photodetectors under polarization conditions. (a) Schematic diagram of the polarization imaging measurement system. Imaging results of the SPT letters under illumination at (b) 450 nm and (c) 1550 nm for polarization angles of 0 ° and 90 °.



Supplementary Fig. 22 | High-frequency response and cut-off frequency. (a) Experimental set-up for the high-frequency measurement. (b-e) Relative response as a function of modulation frequency of the device under different V_{ds} . The dashed line represents the relative response at -3 dB cutoff frequency.



Supplementary Fig. 23 | Light-intensity-dependent optoelectronic synaptic response and decay dynamics analysis. (a) Time-resolved EPSC responses under different light power densities. (b) Extracted characteristic decay time constants at different light intensities obtained from bi-exponential fitting. (c) Light-intensity dependence of the characteristic relaxation time (τ) and stretching exponent (β) extracted from the stretched exponential fitting.

Here, the excitatory postsynaptic current (EPSC) as a function of time under various light power densities. The decay curves were fitted using a bi-exponential function:

$$\Delta EPSC = y_0 + C_1 \exp\left(-\frac{\Delta t}{\tau_1}\right) + C_2 \exp\left(-\frac{\Delta t}{\tau_2}\right) \quad (8)$$

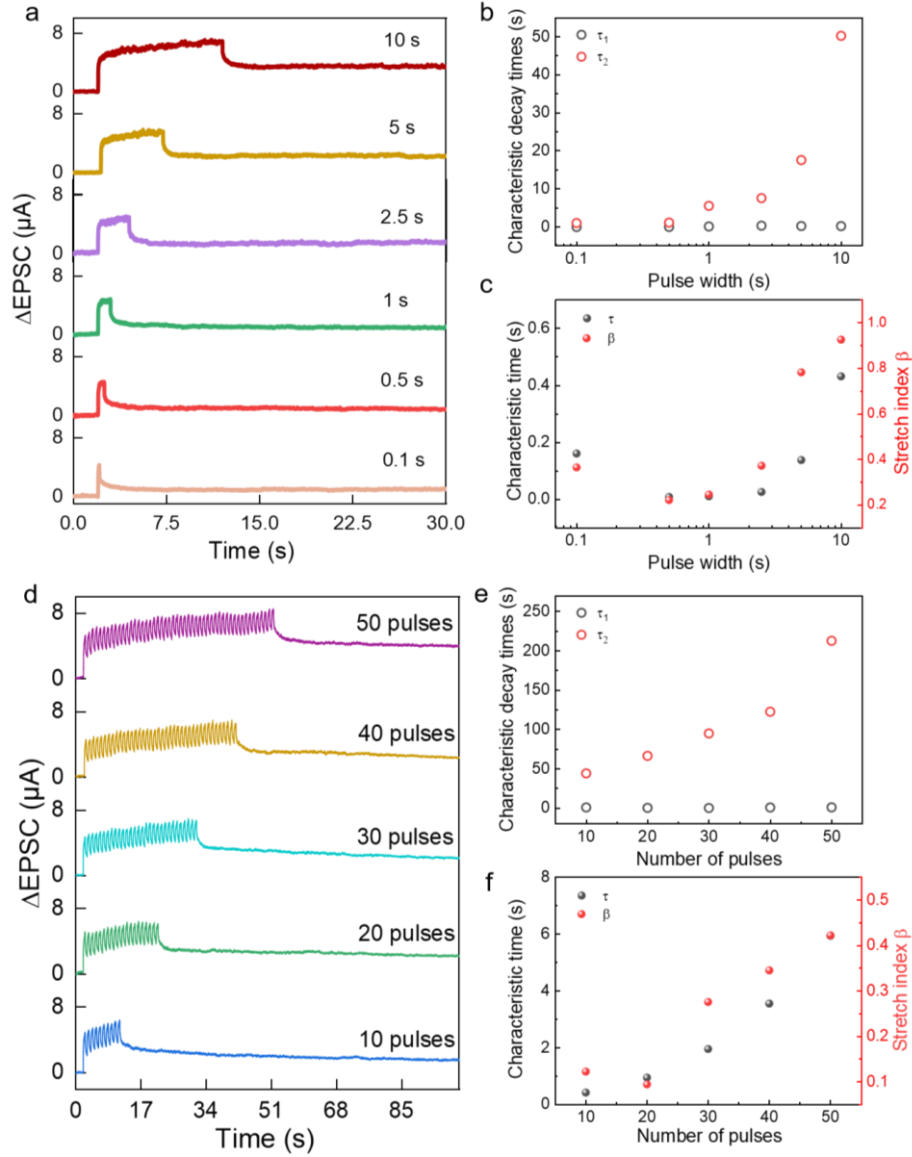
where C_1 and C_2 are the coefficients of the fast and slow components, and τ_1 and τ_2 are the corresponding relaxation time constants. As the light power density increases, τ_1 remains nearly constant, while the slow time constant τ_2 gradually becomes longer. In addition, we fitted the overall decay using the Kohlrausch stretched-exponential function

$$\Delta EPSC \propto \exp\left[-\left(\frac{t}{\tau}\right)^\beta\right] \quad (9)$$

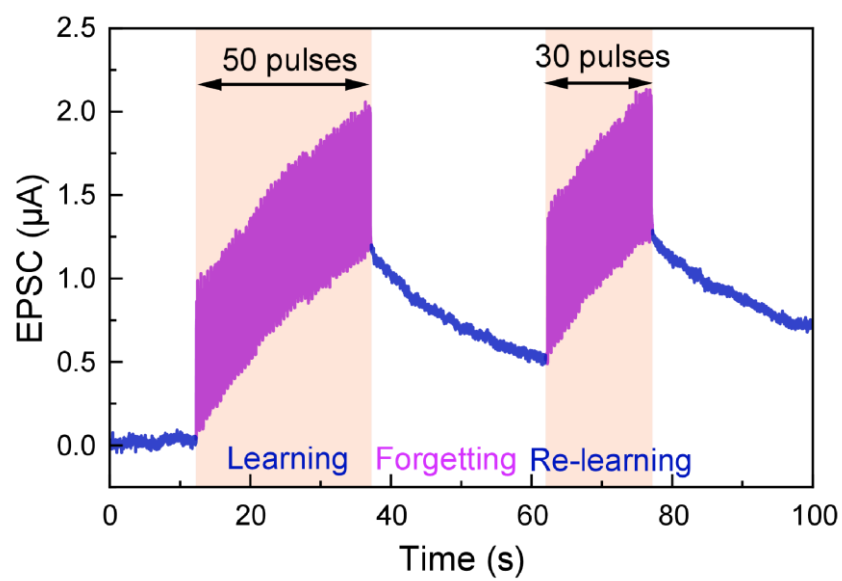
where τ is the characteristic relaxation time and β is the stretching exponent. The parameter β reflects the degree of dispersiveness in the relaxation process: a β close to 1 indicates a more single-exponential decay, whereas a smaller β suggests a broader distribution of relaxation pathways. As the power intensity increases, both τ and β decrease,

indicating that the overall decay becomes faster yet more dispersive. The increase in τ_2 with increasing power intensity shows that the slow process is influenced by excitation intensity; this could be related to enhanced barrier effects under higher light intensity. The decreases in both β and τ indicate more heterogeneous relaxation, consistent with multiple transport pathways. In contrast, when we increase the illumination duration or pulse number, we observe a different behavior: τ_1 again remains essentially unchanged, τ_2 also increases, but β increases, and the Kohlrausch τ increases correspondingly. This opposite evolution of β and τ between the power-dependent and history-dependent experiments suggests that light intensity and illumination history affect the decay dynamics through distinct mechanisms. Higher power intensity yields faster but more dispersive decay, whereas longer illumination or more pulses result in slower but more uniform decay. The latter trend is compatible with progressive filling of reversible shallow traps, where the release process becomes more homogeneous as more traps are occupied. This scenario appears less likely to be dominated by deep traps, which would typically broaden the distribution and reduce the β .

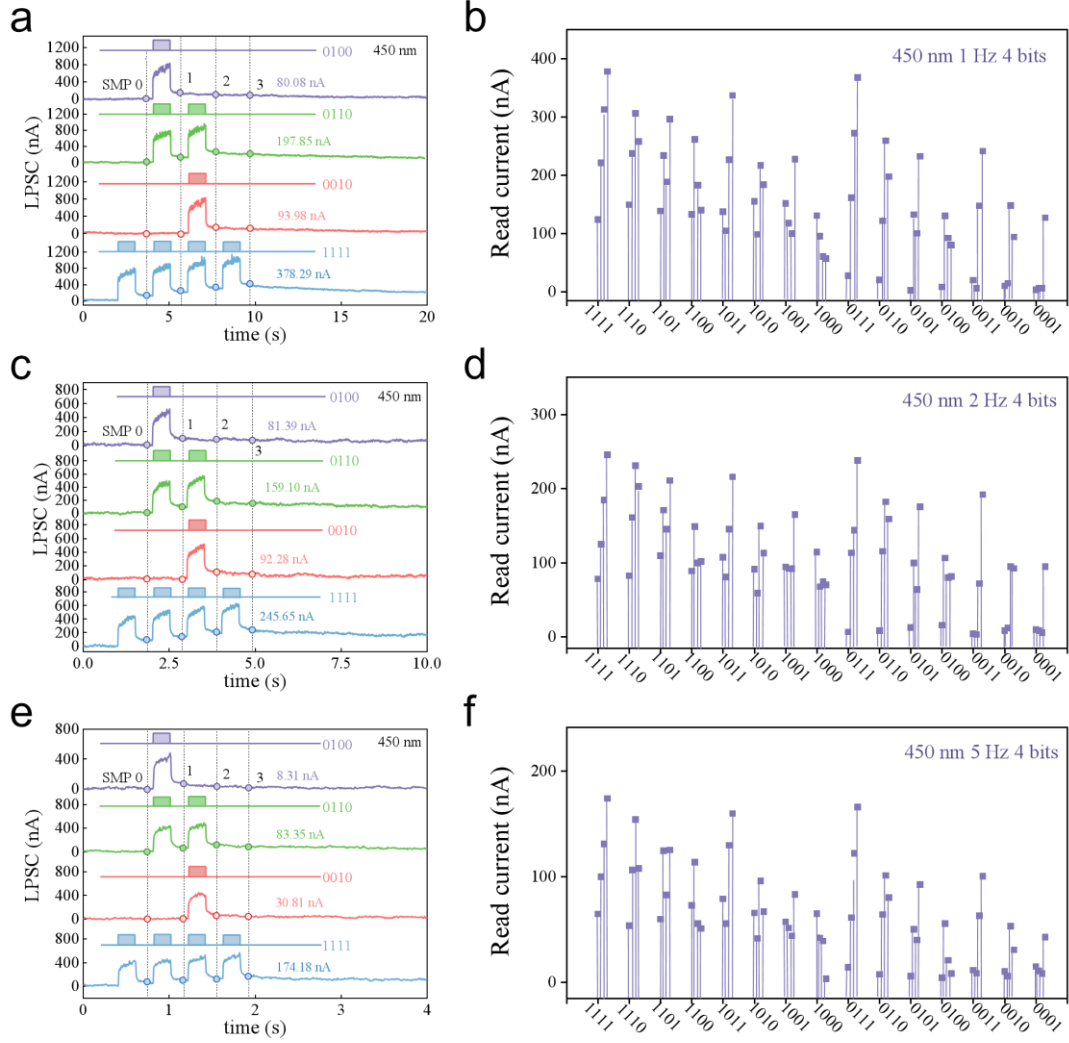
Taken together, these observations indicate that the slow decay is governed by two coexisting factors: barrier-limited transport and shallow-trap-assisted processes. The observed long-term decay does not originate from a conventional deep-trap-dominated PPC, but rather from the interplay between barrier-limited transport and reversible shallow-trap dynamics.



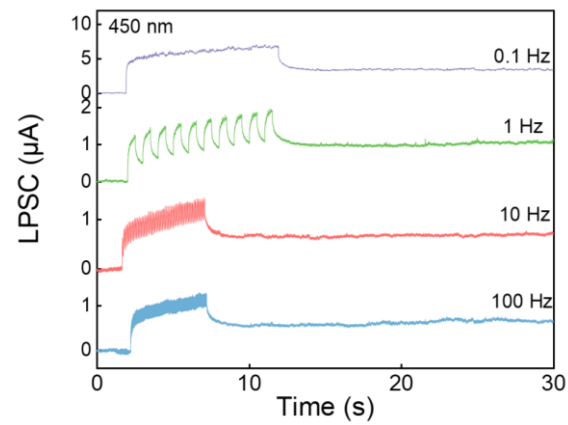
Supplementary Fig. 24 | Illumination-duration-dependent and pulse-number-dependent decay dynamics. (a) Time-resolved EPSC responses under different illumination durations. (b) Extracted characteristic decay time constants at illumination durations. (c) Illumination duration dependence of the τ and β . (d) Time-resolved EPSC responses under different pulse numbers. (e) Extracted characteristic decay time constants at different pulse numbers. (f) Pulse-number-dependent dependence of the τ and β .



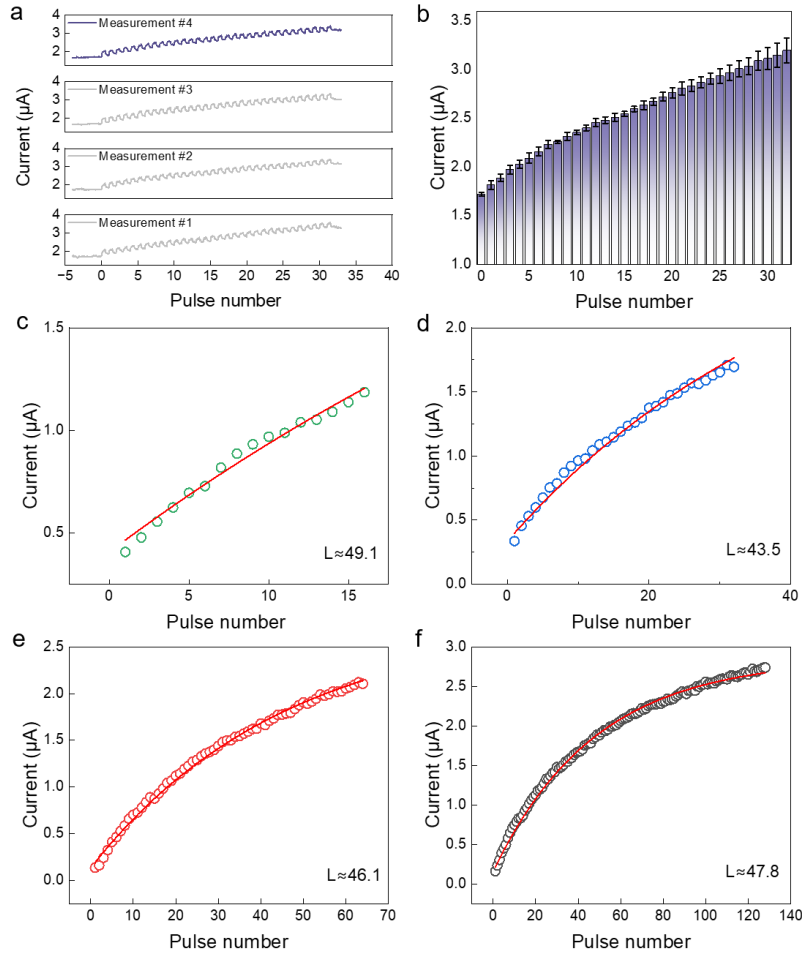
Supplementary Fig. 25 | Learning-experience behavior under pulse stimulation.



Supplementary Fig. 26 | Optoelectronic performance of the optoelectronic neurons. The encoding process under (a) 1 Hz, (c) 2 Hz and (e) 5 Hz light frequency modulation. For 0110 stimulation, where 1 and 0 represents the light illumination and the dark condition, respectively. The photoelectric responses to four-bit light stimulation under (b) 1 Hz, (d) 2 Hz and (f) 5 Hz light modulation irradiation. The characterizations were conducted at $V_g = 0$ V and $V_{ds} = 3$ V.



Supplementary Fig. 27 | Frequency-dependent synaptic response of the device across a wide operating range from 0.1 Hz to 100 Hz.



Supplementary Fig. 28 | Quantitative evaluation of key reservoir computing metrics under pulsed operation. (a, b) Repeated LTP measurements under identical stimulation conditions. The error bars represent the standard deviation of 4 repeated measurements. (c-f) Conductance evolution as a function of pulse number under 16-pulse, 32-pulse, 64-pulse, and 128-pulse conditions, respectively, with the corresponding fitted curves.

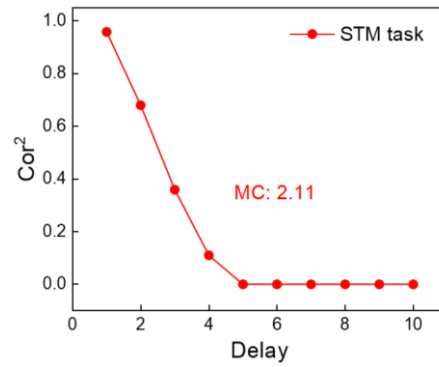
Here, to quantitatively characterize the linearity of the LTP response, the conductance evolution with pulse number was fitted using the saturation model

$$G_{LTP} = G_c(1 - e^{-\frac{P}{L}}) + G_{\min} \quad (10)$$

where G_c is the theoretical maximum conductance swing, $G_{\min}$ is the minimum conductance, and P is the pulse number. In this model, L serves as a linearity index that reflects how quickly the conductance approaches saturation. The local curvature of the normalized conductance trace, given by

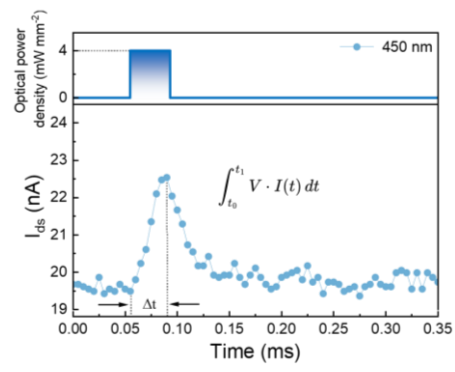
$$\frac{dG_{\text{normalised}}}{dP^2} = -\frac{1}{L^2} e^{-\frac{P}{L}} \quad (11)$$

is largest during the initial pulses and decreases as the conductance approaches saturation. By fixing G_c as a global constant and fitting all non-normalized conductance traces with L as the only free parameter.

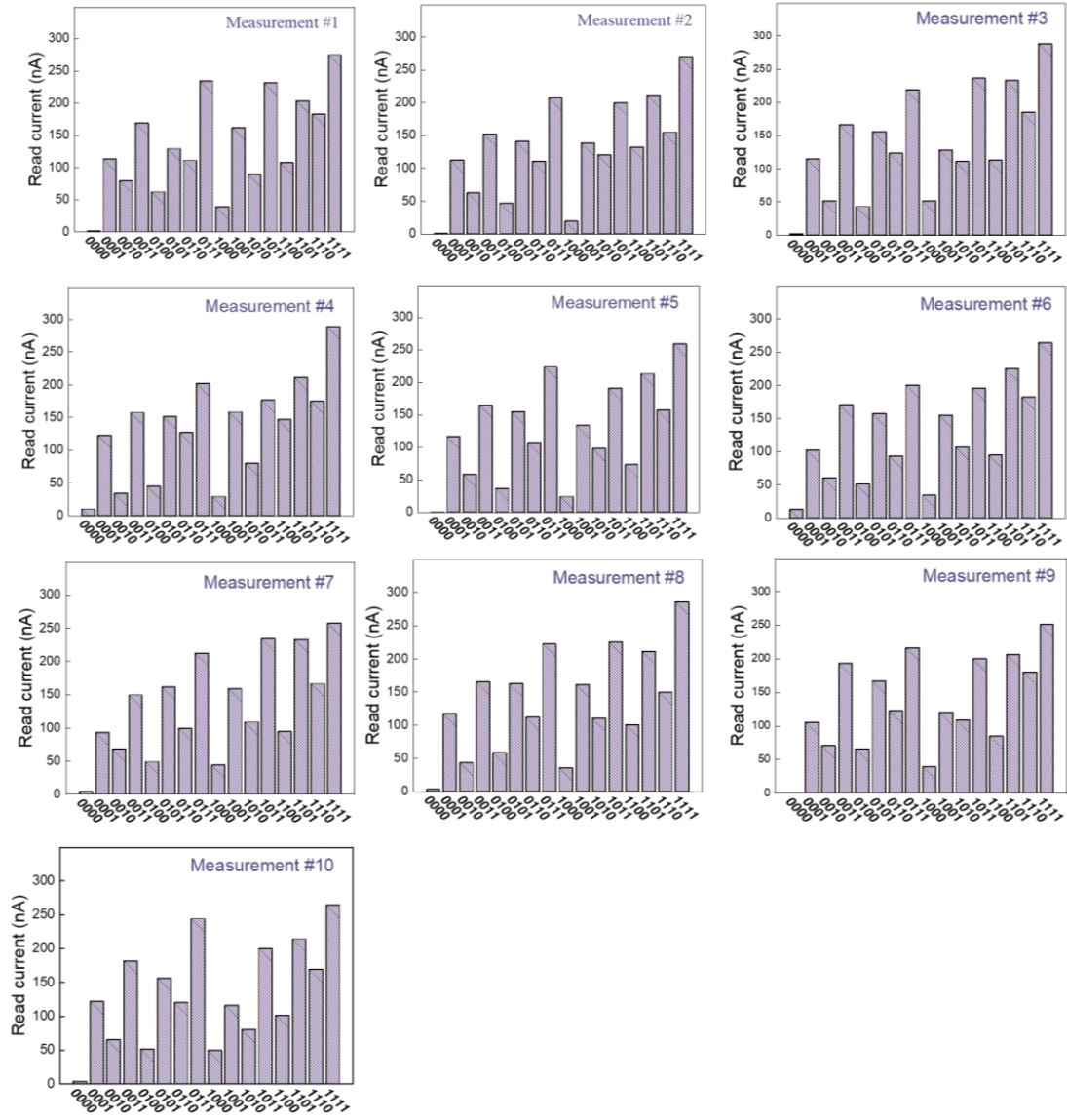


Supplementary Fig. 29 | Memory capacity evaluation based on a STM task, showing the squared correlation coefficient (Cor^2) as a function of delay.

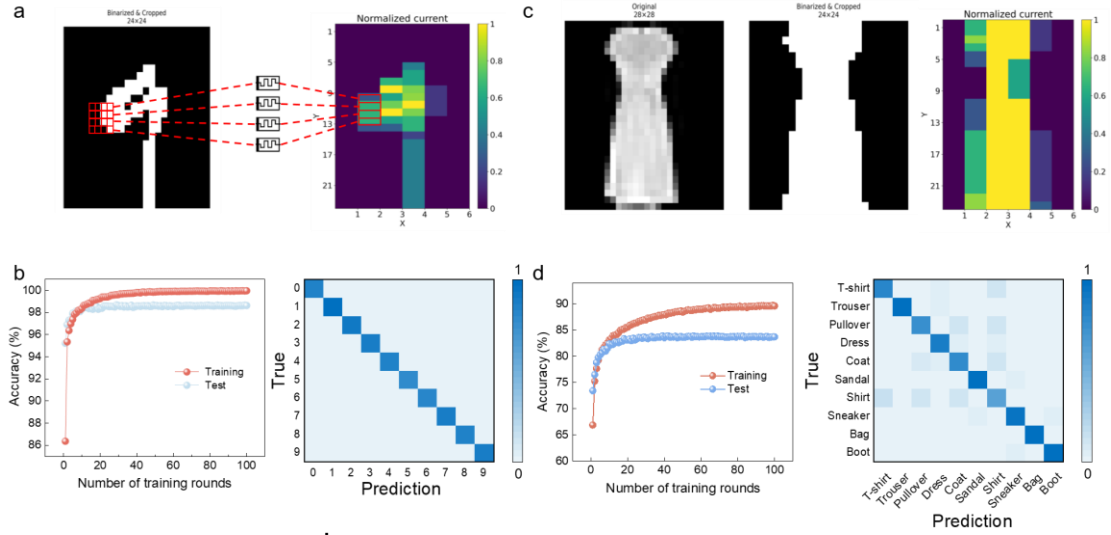
Here, the memory capacity (MC) is evaluated using a standard short-term memory task. Specifically, we calculate the squared correlation coefficient between the device output and time-delayed input signals, and integrate over different delay steps to obtain the total memory capacity. The correlation decays rapidly with increasing delay and approaches zero beyond a delay of 6, yielding an integrated memory capacity of $MC = 2.11$. This value indicates that the device possesses a finite but effective short-term memory window, comparable to other conductance-modulation-based reservoir computing systems, thereby confirming its capability for temporal information processing.



Supplementary Fig. 30 | Diagram of energy consumption measurement methods for a single synaptic event.



Supplementary Fig. 31 | Statistical and temporal characteristics of multi-level photoresponse states.



Supplementary Fig. 32 | Recognition of MNIST and Fashion-MNIST images.

(a) Recognition of the handwritten digit 9 from the MNIST dataset, showing the reservoir output for digit 9. b) Recognition of a dress image from the Fashion-MNIST dataset, showing the reservoir output for dress. c) Training accuracy after 100 epochs for digit pattern recognition and the confusion matrix for all 10 classes of the MNIST dataset. d) Training accuracy after 100 epochs for image recognition on the Fashion-MNIST dataset and the confusion matrix for all 10 classes.

Here, the MNIST dataset consists of 10,000 training images and 7250 test images with a resolution of 28×28 pixels. To adapt the input to the proposed 4-bit RC system, the images were preprocessed through cropping and binarization before being converted into temporal input sequences. Representative simulated outputs for a typical handwritten digit 9 and a dress image from the f-MNIST dataset are presented in Supplementary Fig. 32 a and c, respectively, demonstrating the capability of the device to encode and process distinct visual patterns. Furthermore, to evaluate performance on a more challenging task, the f-MNIST dataset was employed, where the categories exhibit higher inter-class similarity.

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