

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

Corresponding Author: Dr Fang Yang

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

In this manuscript, Wang et al. report a photodetector based on a Ta₂NiSe₅/MoTe₂/PdSe₂ vertical heterostructure, in which the response speed can be tuned by varying the applied bias. The device achieves a response time of ~600 ns under low-voltage operation and exhibits broadband photoresponse, indicating promising performance. The bias-dependent modulation of the photoresponse dynamics is of potential interest. However, in its current form, the manuscript remains insufficiently developed for publication in Nature Communications. Several key conclusions are not adequately supported by experimental evidence, and the physical mechanisms are not rigorously analyzed. More importantly, although the device is claimed to bridge high-speed photodetection and neuromorphic computing, the presented application does not convincingly demonstrate this capability and instead relies on conventional, previously reported behavior. Overall, substantial additional work is required, and the manuscript in its current form does not meet the standard for publication in Nature Communications.

Specific comments are as follows:

1. The use of the term “unipolar barrier” in the title is not well justified. The manuscript primarily describes two different operating regimes under low and high bias, but does not provide a clear physical basis or discussion that supports the designation of a unipolar barrier. As such, the terminology appears misleading and not fully consistent with the content of the work.
2. The analysis of the band structure is not sufficiently convincing. The KPFM measurements presented in Supplementary Fig. 3 show noticeable drift, which raises concerns regarding their reliability for quantitatively determining work-function differences among the constituent materials. Moreover, KPFM can only provide information about the Fermi level (work function), and cannot directly determine the conduction-band and valence-band positions. Therefore, the band diagrams shown in Supplementary Fig. 5 are not adequately supported by the presented characterization. In addition, the relative valence-band alignment between Ta₂NiSe₅ and MoTe₂ appears inconsistent between Fig. 2a and Fig. 2b, which contradicts basic semiconductor physics, as the relative band offsets at a heterointerface should remain invariant despite band bending.
3. The manuscript lacks sufficient structural and interfacial characterization of the heterostructure. Raman spectroscopy alone is not sufficient to demonstrate interface quality, particularly for vertical heterostructures where interfacial properties critically affect device performance and transport behavior. Cross-sectional STEM characterization is required.
4. There is no discussion of how structural parameters such as layer thickness, lateral size, or contact area influence device performance. These factors are important for understanding both the transport mechanism and the scalability of the device. The authors should investigate and discuss how these parameters affect device behavior and the underlying mechanisms.
5. Although the device exhibits attractive photodetection performance, including sub-microsecond response time and mA/W-level responsivity, the manuscript does not provide sufficient physical insight into the origin of these properties. The underlying mechanisms responsible for the high performance are not adequately analyzed. A more comprehensive physical model, supported by experimental data, is required to explain the observed behavior.
6. The synaptic characterization is relatively conventional and does not provide clear novelty. In addition, the physical origin of the strong bias dependence of the response speed is not discussed in sufficient depth. The explanation based solely on thermal emission appears oversimplified and does not fully account for the observed behavior.
7. The energy-consumption analysis is unclear. In Fig. 4e, the authors present energy consumption values at different wavelengths, but do not explain how these values are calculated. It is also unclear whether the reported values include only

optical energy or also account for electrical energy consumption during device operation.

8. The presentation of Fig. 4h is not sufficiently clear. Although 16 different output states are shown, the current format does not allow straightforward comparison. In addition, given the inherent variability in temporal decay processes, the absence of repeated measurements and statistical analysis raises concerns regarding the robustness of the results.

9. The connection between the tunable response dynamics and neuromorphic vision is not convincingly demonstrated. While the manuscript emphasizes the capability of integrating high-speed sensing and neuromorphic computation, the application demonstration does not utilize the fast response regime, nor does it experimentally validate reservoir computing functionality. Instead, the demonstration relies on the slow, volatile photoresponse, which has been widely reported in previous studies. As a result, this part lacks novelty and does not substantiate the claims made in the Introduction. Furthermore, no device array is fabricated or experimentally characterized, and the application is supported only by simulations, which significantly weakens the overall impact. The authors should design application scenarios that explicitly leverage the tunable response dynamics, and experimentally validate these using device arrays. Discussions on scalability, device uniformity, and cycle-to-cycle stability are also necessary.

10. Finally, there is a textual error at line 158, where “Under no illumination” should be corrected to “Under illumination.”

Reviewer #2

(Remarks to the Author)

This paper presents a bias-reconfigurable photodetector based on a Ta₂NiSe₅, MoTe₂, PdSe₂ van der Waals heterostructure. The work moves toward the realization of compact neuromorphic systems by enabling a single photodetector to switch between sensing and memory functions. This integration reduces the need for multiple hardware components and simplifies system architecture. The device exhibits multifunctionality, combining photodetection, memory effects, and neuromorphic behavior, along with broadband and polarization-sensitive detection capabilities. Overall, the manuscript presents a promising step toward hardware-efficient intelligent vision systems.

While this concept is widely discussed at present, the core aspects of physical interpretation and experimental verification remain weak. I cannot support publication now because much improvement remains in terms of analysis, performance comparison, and technical details.

1. The proposed mechanism relies critically on the modulation of interfacial barrier height under applied bias. However, the claimed evolution of band alignment lacks simulation supporting and direct experimental evidence. Key metrics such as memory capacity and nonlinearity are not quantitatively evaluated. In addition, the demonstrated results rely on a relatively small, custom dataset without validation on standard benchmark tasks.

2. The manuscript employs a Ta₂NiSe₅, MoTe₂, PdSe₂ heterostructure; however, its advantages over existing systems such as 2D neural network sensors, optoelectronic RRAM, and silicon-based platforms are not clearly articulated.

3. The reported modulation of response time from nanosecond level (0.1 V) to second level (3 V) spans over seven orders of magnitude. Attributing this dramatic change solely to barrier-height-induced transition from direct tunneling to thermal emission is not fully convincing, particularly given the observed long retention behavior. The authors should clarify how they exclude the possibility that high bias induced defects (e.g., due to the damaging in material under strong electric field) contribute to the super slow dynamics.

4. The time-domain photoresponse at 0.1 V shows a rise time of ~257 ns, which corresponds to an estimated bandwidth of ~1.36 MHz ($f=0.35/t$). However, the frequency-domain measurement presented in Supplementary Fig. 11 shows a bandwidth of only ~0.11 MHz, which is over one order of magnitude lower. Please estimate this discrepancy.

5. The barrier layer of the proposed unipolar photodetector is MoTe₂. However, MoTe₂ has a relative low bandgap (~1 eV), which means MoTe₂ is sensitive to most of the wavelengths that the authors used (e.g. 450 nm – 1320 nm). So, MoTe₂ cannot really prevent electron transport, because it can generate photocarriers itself. This further leads to the incomplete of the bias-related response speed mechanism, as the response speed (e.g. Fig. 3e) is measured under 633nm, which can cause the photoresponse in MoTe₂ “barrier” layer. Please evaluate contribution of photogenerated carriers in MoTe₂ and clarify how this affects the interpretation of the barrier-controlled response speed mechanism.

6. The central claim is based on barrier modulation within a specific Ta₂NiSe₅/MoTe₂/PdSe₂ heterostructure. However, no experiments are provided to demonstrate the generality or reproducibility of this mechanism. For instance, it would be valuable to examine whether similar behavior persists under variations, such as different material thicknesses and alternative barrier materials.

7. The manuscript presents polarization-sensitive photodetection characteristics (Fig. 3d). However, this functionality seems disconnected from the core device concept and is not utilized in the demonstrated applications (Fig. 5). The authors should clarify the role and significance of polarization sensitivity in the context of the proposed system.

8. The author claims broadband photoresponse from the visible (450nm) to mid-infrared (3.8um). However, the presented photodetector performance data (e.g., I-V characteristics, responsivity, and response time) are only experimentally demonstrated up to approximately 3.1um. The evidence at 3.8um appears to be limited to synaptic (EPSC) measurements, without corresponding photodetector performance. Therefore, the claim of operation up to 3.8um is not sufficiently supported by the current data.

9. The simulation task remains unclear how such functionality can be practically realized at the hardware level. In particular, the manuscript appears to rely on a single device. Could the authors clarify how this approach enables scalable on-chip integration? How would multiple devices be interconnected to support real-time processing, and what are the limitations for large-scale system implementation?

10. In Supplementary Fig. 12, the transition from short-term memory (STM) to long-term memory (LTM) is demonstrated. However, the analysis remains largely qualitative. Quantitative metrics such as retention time constants and decay rates are not provided. Could the authors include a more rigorous and quantitative characterization of the synaptic dynamics to better substantiate the claimed memory behavior?

Reviewer #3

(Remarks to the Author)

This manuscript could become acceptable for publication in Nature COmmunications after major revisions. For details, see attached report.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I am very pleased to see the extensive experiments and analysis added by the authors, which makes the manuscript more substantial and reliable. I believe the authors has provided sufficient responses to the reviewer's comments, so I suggest publishing it in Nature Communications.

Reviewer #2

(Remarks to the Author)

Reviewer #3

(Remarks to the Author)

The authors have expanded their already impressive wealth of experimental data to support their claims. While not always easy to read and follow, after full inspection of the various new datasets provided, I find the additional work mostly adequate and suitable to sufficiently support the main conclusions of this manuscript. This was my main concern with the previous version, and this has been removed. Therefore, I can now recommend the acceptance of this work for publication in Nature Communications.

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The revised manuscript adequately addresses my previous concerns and provides sufficient additional evidence to support its main conclusions.

I have no further major objections and recommend the manuscript for publication in Nature Communications.

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REVIEWER COMMENTS

Dear editor,

Thank you very much for providing us with the constructive comments from the reviewers. We would like to thank all the referees for their critical comments that allowed us to further improve our manuscript. The manuscript has been revised according to the reviewer's comments, and a point-by-point response is given below.

Reviewer #1:

In this manuscript, Wang et al. report a photodetector based on a Ta₂NiSe₅/MoTe₂/PdSe₂ vertical heterostructure, in which the response speed can be tuned by varying the applied bias. The device achieves a response time of ~600 ns under low-voltage operation and exhibits broadband photoresponse, indicating promising performance. The bias-dependent modulation of the photoresponse dynamics is of potential interest. However, in its current form, the manuscript remains insufficiently developed for publication in Nature Communications. Several key conclusions are not adequately supported by experimental evidence, and the physical mechanisms are not rigorously analyzed. More importantly, although the device is claimed to bridge high-speed photodetection and neuromorphic computing, the presented application does not convincingly demonstrate this capability and instead relies on conventional, previously reported behavior. Overall, substantial additional work is required, and the manuscript in its current form does not meet the standard for publication in Nature Communications.

[Response]:

We sincerely thank the reviewer for the summary of our work and for highlighting the promising performance and potential interest of the bias-dependent photoresponse modulation, as well as the valuable suggestions, which help us to further improve the quality of this manuscript. We have carefully addressed the concerns point-by-point as follows.

[Comments 1]: *The use of the term “unipolar barrier” in the title is not well justified. The manuscript primarily describes two different operating regimes under low and high bias, but does not provide a clear physical basis or discussion that supports the designation of a unipolar barrier. As such, the terminology appears misleading and not fully consistent with the content of the work.*

[Response]:

We thank the reviewer for this insightful comment. We have carefully considered the term “unipolar barrier” and would like to clarify our rationale and the revision we have made.

In the original manuscript, we adopted the term “unipolar barrier” primarily to emphasize the **carrier-selective transport characteristics** of our heterojunction. As shown in the band alignment of the heterojunction, the large conduction-band offset and small valence-band offset allow photogenerated holes to pass through the interface while effectively blocking electrons. This functional behavior, which involves preferential transport of one carrier type, resembles the conceptual essence of a “unipolar” barrier.

Nevertheless, upon the reviewer’s suggestion, we recognize that the term “unipolar barrier photodetector” carries a well-established and specific meaning in the literature, particularly associated with III–V infrared detector architectures such as nBn and pBn structures.^[1, 2] In that context, the unipolar barrier is deliberately designed to block majority carriers for dark current suppression while maintaining photocurrent collection. Although our device exhibits analogous carrier selectivity, its operating mechanism involves a combination of tunneling, barrier modulation under bias, and trap-assisted processes, which extends beyond the classical definition and application scope of unipolar barrier photodetectors. To avoid potential misunderstanding by readers familiar with that established concept, and to ensure terminological precision, we have replaced **“unipolar barrier photodetector”** with **“barrier photodetector”** throughout the revised manuscript.

[1] *Applied Physics Letters*, 89, 2006.

[2] *Applied Physics Letters*, 95, 2009.

[Comments 2]: *The analysis of the band structure is not sufficiently convincing. The KPFM measurements presented in Supplementary Fig. 3 show noticeable drift, which raises concerns regarding their reliability for quantitatively determining work-function differences among the constituent materials. Moreover, KPFM can only provide information about the Fermi level (work function), and cannot directly determine the conduction-band and valence-band positions. Therefore, the band diagrams shown in Supplementary Fig. 5 are not adequately supported by the presented characterization. In addition, the relative valence-band alignment between Ta₂NiSe₅ and*

MoTe₂ appears inconsistent between Fig. 2a and Fig. 2b, which contradicts basic semiconductor physics, as the relative band offsets at a heterointerface should remain invariant despite band bending.

[Response]:

We sincerely thank the reviewer for the valuable comments. We fully agree with the reviewer that KPFM measurements can only provide information about the Fermi level of the materials and cannot directly determine the positions of the conduction-band minimum (CBM) and valence-band maximum (VBM). To address this, we have combined KPFM with ultraviolet photoelectron spectroscopy (UPS) measurements and first-principles calculations to determine the band alignment. The details are provided below.

First, we performed UPS measurements using a He I excitation source to determine the work functions and valence-band positions of the materials. The work functions were obtained from the secondary-electron cutoff using $W = h\nu - E_{\text{cut}}$, where $h\nu = 21.22$ eV is the photon energy of the He I source. For Ta₂NiSe₅, MoTe₂, and PdSe₂, the work functions are 4.64 eV, 4.69 eV, and 4.91 eV, respectively, and the VBM are located at 0.27 eV, 0.35 eV, and 0.24 eV below the Fermi level.

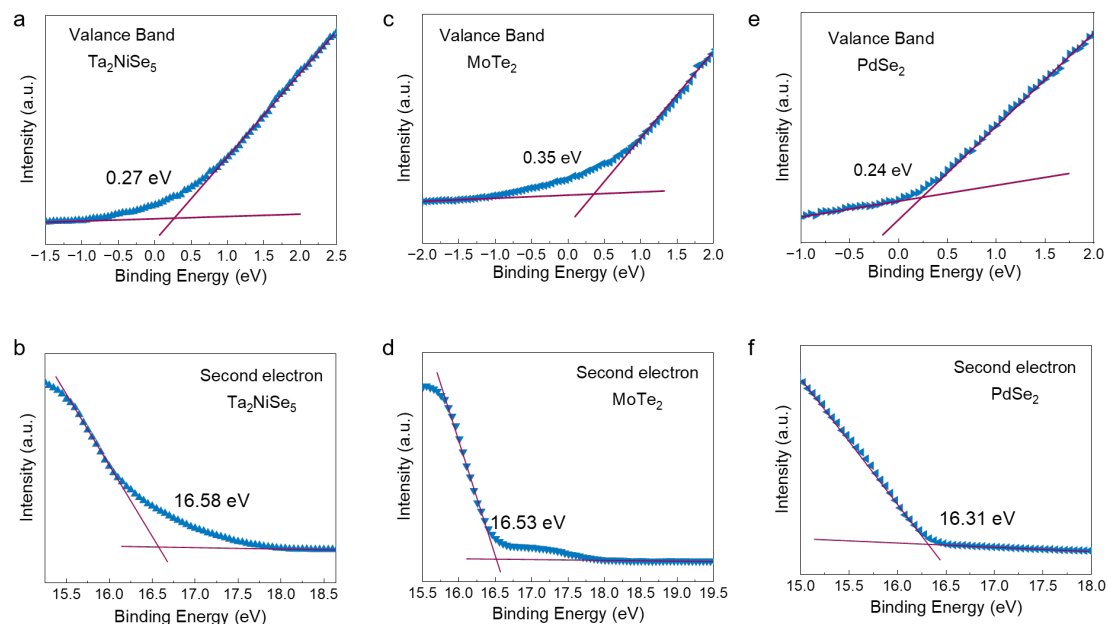


Figure R1 UPS characterization of Ta₂NiSe₅, MoTe₂, and PdSe₂. (a, b) Valence band spectrum and secondary electron cutoff of Ta₂NiSe₅. (c, d) Valence band spectrum and secondary electron cutoff of MoTe₂. (e, f) Valence band spectrum and secondary electron cutoff of PdSe₂.

Second, we carried out first-principles density functional theory calculations using VASP

method. The calculated band structures of the individual materials yield the following bandgap values: approximately 0.35 eV for Ta_2NiSe_5 , 1.09 eV for MoTe_2 , and 0.42 eV for PdSe_2 .

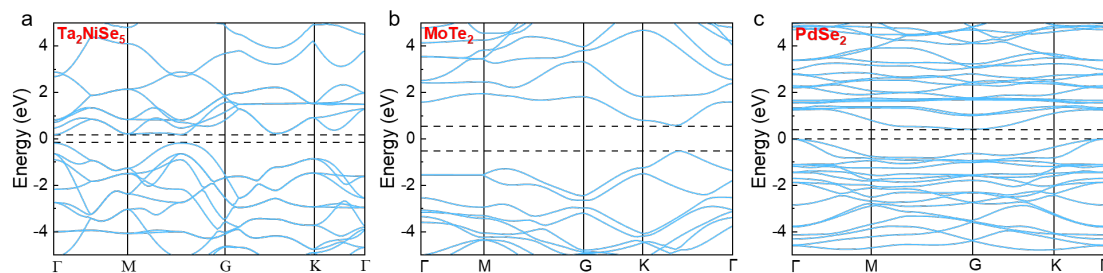


Figure R2 DFT calculations of the electronic structure of Ta_2NiSe_5 , MoTe_2 , and PdSe_2 .

Hence, by combining the VBM positions and work functions obtained from UPS with the bandgap values from DFT, we reconstructed the band diagrams of the heterojunction.

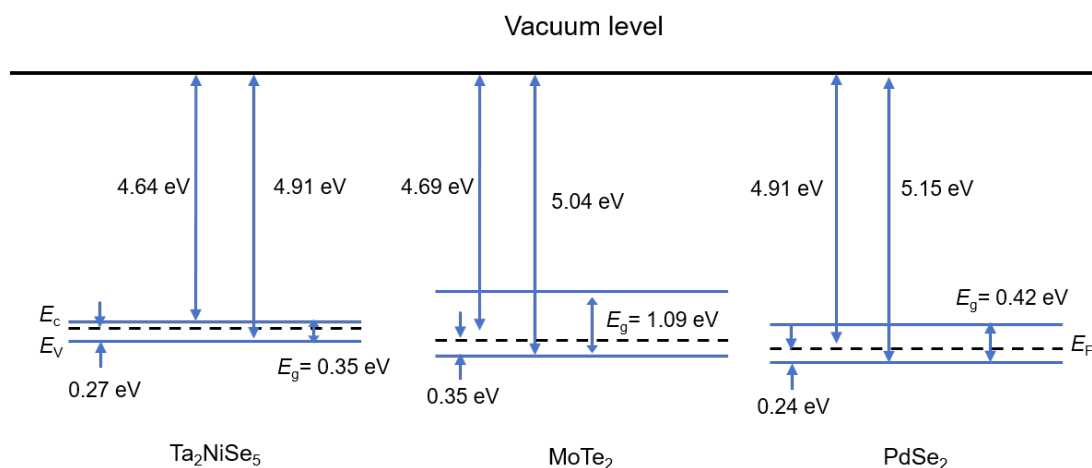


Figure R3 Energy band profiles of Ta_2NiSe_5 , MoTe_2 , and PdSe_2 .

Third, the relative valence-band alignment between the heterojunction shown in the original Fig. 2a and Fig. 2b was intended as a schematic illustration to qualitatively demonstrate that the applied bias can effectively modulate the interfacial barrier height and carrier transport behavior in the heterostructure. Such modulation of interfacial band profiles induced by applied voltages has been widely discussed in previous studies on semiconductors and van der Waals heterostructures.^[1,2] However, we acknowledge that the original representation may have caused ambiguity regarding the invariance of the intrinsic band alignment. Following the reviewer's suggestion, we have carefully redrawn the energy-band diagrams under different bias voltages, explicitly distinguishing the intrinsic band offsets from the bias-dependent band bending (Figure R4).

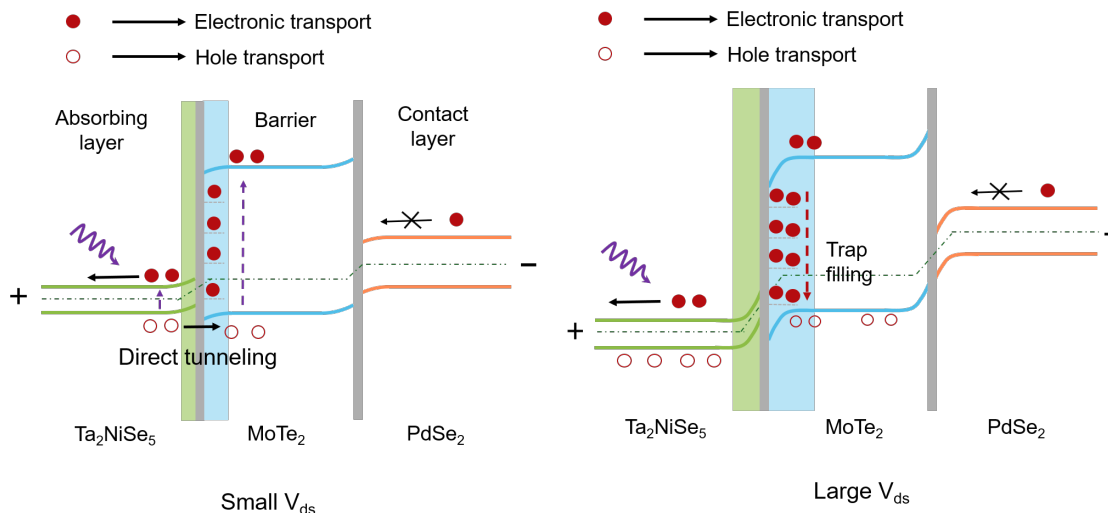


Figure R4 Energy band diagrams for the Ta₂NiSe₅/MoTe₂/PdSe₂ device under low and high bias, respectively.

[1] Advanced Optical Materials, 12(16), 2303088, 2024

[2] ACS Nano, 19(1), 1302–1315, 2025

[Actions]:

We have made the corresponding revisions on page 4, 5 of the revised manuscript, including Figs. 2a, 2b, 2f and Supplementary Figs. 6 to 8.

[Comments 3]: The manuscript lacks sufficient structural and interfacial characterization of the heterostructure. Raman spectroscopy alone is not sufficient to demonstrate interface quality, particularly for vertical heterostructures where interfacial properties critically affect device performance and transport behavior. Cross-sectional STEM characterization is required.

[Response]:

We thank the reviewer for this important comment regarding the structural and interfacial characterization of the heterostructure. We fully agree that Raman spectroscopy alone is not sufficient to comprehensively evaluate the interface quality, particularly for vertical heterostructures where interfacial properties critically affect charge transport.

In response to this suggestion, we have strengthened the structural characterization in the revised manuscript by incorporating cross-sectional transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDS) analyses. The cross-sectional TEM images (Figure R5.a) clearly reveal a well-defined and clean interface between the stacked layers, with no

observable interfacial amorphous regions or significant contamination. The sharp contrast between the layers further confirms the high crystallinity and integrity of the heterostructure. In addition, EDS elemental mapping (Figure R5.b-d) demonstrates a well-confined spatial distribution of the constituent elements, with clear boundaries across the interface.

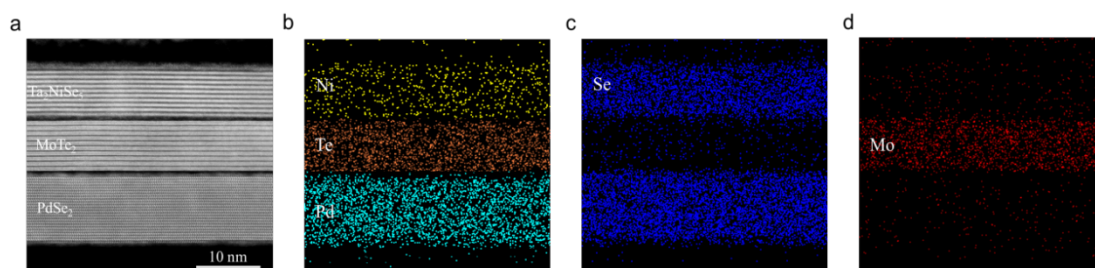


Figure R5 Structural and interfacial characterization of the heterostructure. (a) Cross-sectional TEM image of the vertical heterostructure, showing a well-defined and clean interface between the stacked layers. (b-d) EDS elemental mapping of the corresponding region, illustrating the spatial distribution of the constituent elements. The elemental maps confirm a clear separation of each layer, indicating the formation of a sharp and high-quality interface.

[Actions]:

We have made the corresponding revisions on page 5 of the revised manuscript, Supplementary Figs. 4.

[Comments 4]: *There is no discussion of how structural parameters such as layer thickness, lateral size, or contact area influence device performance. These factors are important for understanding both the transport mechanism and the scalability of the device. The authors should investigate and discuss how these parameters affect device behavior and the underlying mechanisms.*

[Response]:

We sincerely thank the reviewer for the valuable comments. In response, we have additionally fabricated and systematically investigated devices with different layer thicknesses and varying junction areas to clarify the role of structural geometry in carrier transport and device scalability.

Our key findings are as follows:

(i) The dark current increases with junction area but decreases with barrier thickness, which is consistent with previous reports on area-dependent dark current scaling in photodetectors and the exponential suppression of tunneling current in thicker barriers.

- (ii) Despite these variations in dark current, overall responsivity remains stable across all devices.
- (iii) Most importantly, the bias-dependent fast and slow response times remain unchanged across different geometries, confirming that the fundamental carrier transport mechanism is governed by interfacial barrier physics rather than geometric scaling.

The detailed experimental results are provided below:

1. Effect of junction area

We fabricated devices with junction areas of $\sim 20 \mu\text{m}^2$, $\sim 32 \mu\text{m}^2$, and $\sim 100 \mu\text{m}^2$ (Figure R6). The dark current at $V_{\text{ds}} = 0.1 \text{ V}$ increases with increasing area, from $\sim 1.32 \text{ nA}$ at $20 \mu\text{m}^2$ to $\sim 3.8 \text{ nA}$ at $32 \mu\text{m}^2$ and further to $\sim 8 \text{ nA}$ 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.

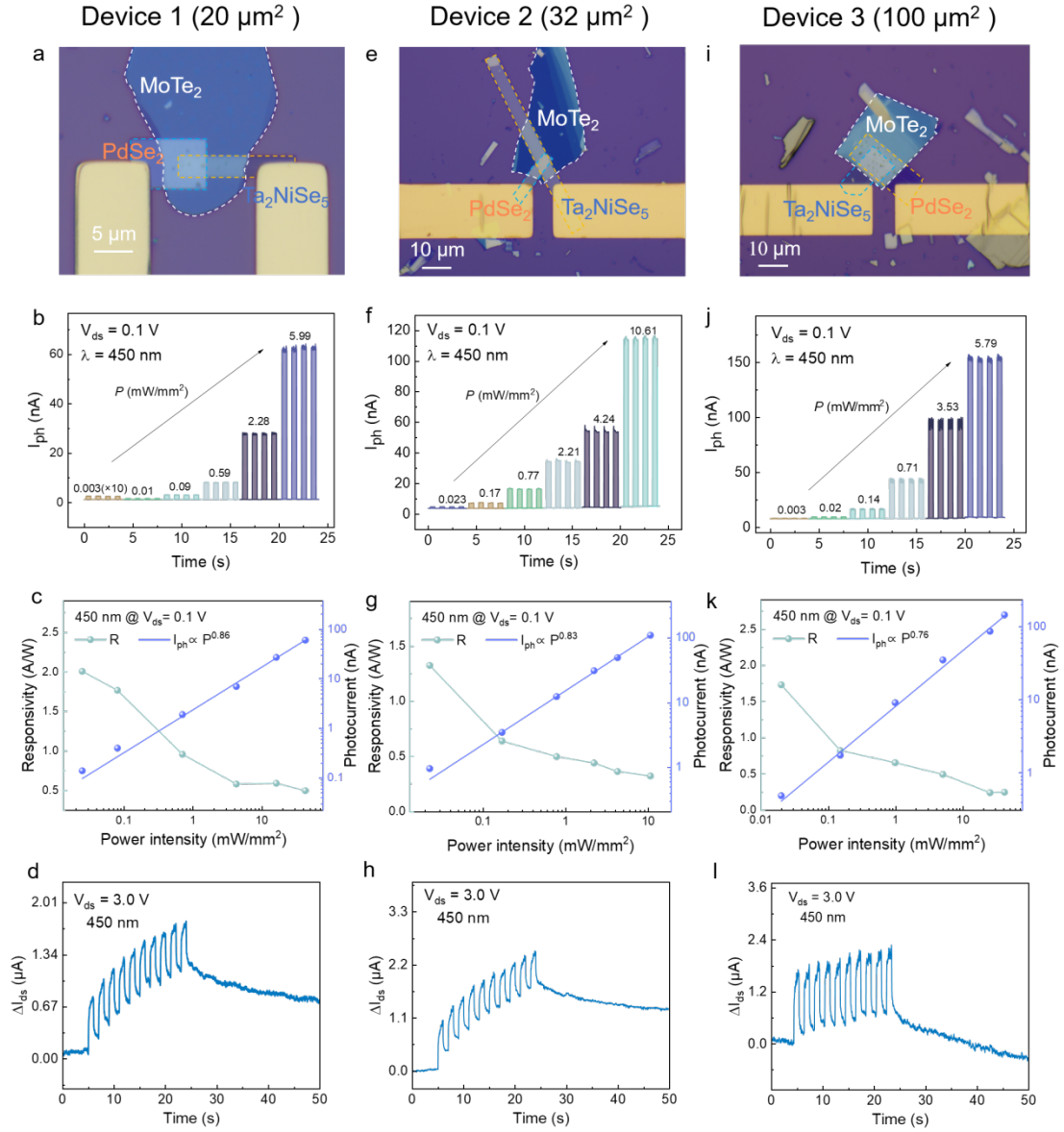


Figure R6 Photoresponse characteristics of the device in different areas. (a, e, i) Optical images of the 3 devices. (b, f, j) Low-bias time-resolved switching response under 450 nm illumination of the 3 devices, showing stable and repeatable photocurrent modulation. (c, g, k) Photocurrent and responsivity 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 PPC effect with slow decay behavior.

2. Effect of layer thickness

We have fabricated devices with different layer thicknesses (Figure R7). The thickness of the Ta_2NiSe_5 absorption layer and PdSe_2 contact layer were kept approximately constant across all

devices, while the thickness of the MoTe₂ barrier layer was systematically varied. Specifically, the MoTe₂ thicknesses are 23.3 nm (Device 4), 17.08 nm (Device 5), and 8.96 nm (Device 6) (Figure R8). 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:

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

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 close to each other. Importantly, all devices with different MoTe₂ thicknesses exhibit the same bias-dependent fast-to-slow switching behavior (Figure R6).

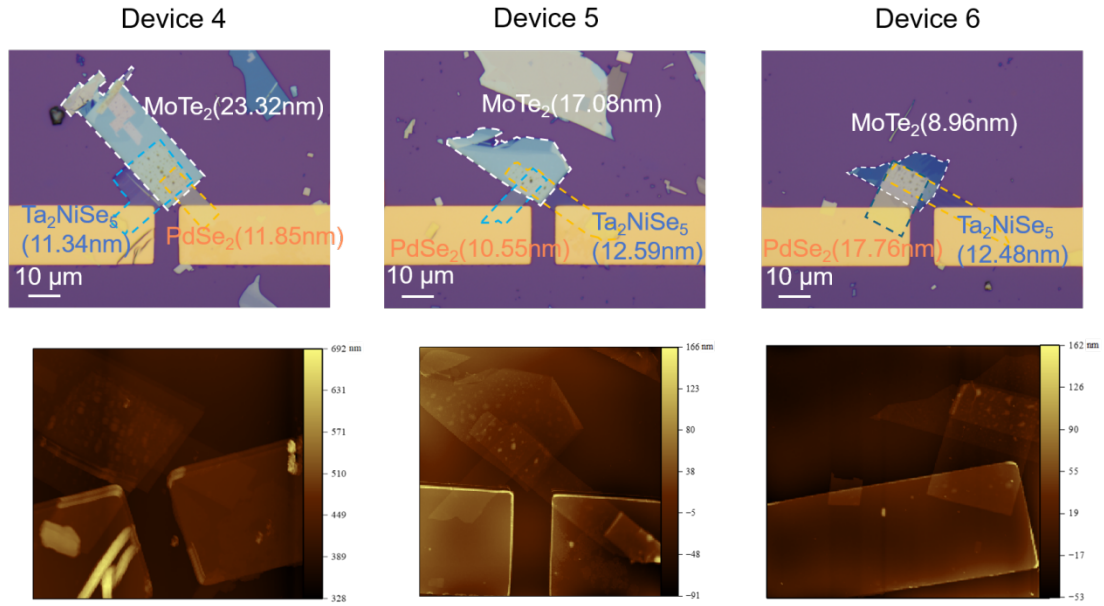


Figure R7. Optical images and corresponding atomic force microscopy (AFM) images of three Ta₂NiSe₅/MoTe₂/PdSe₂ heterostructure devices with different layer thicknesses.

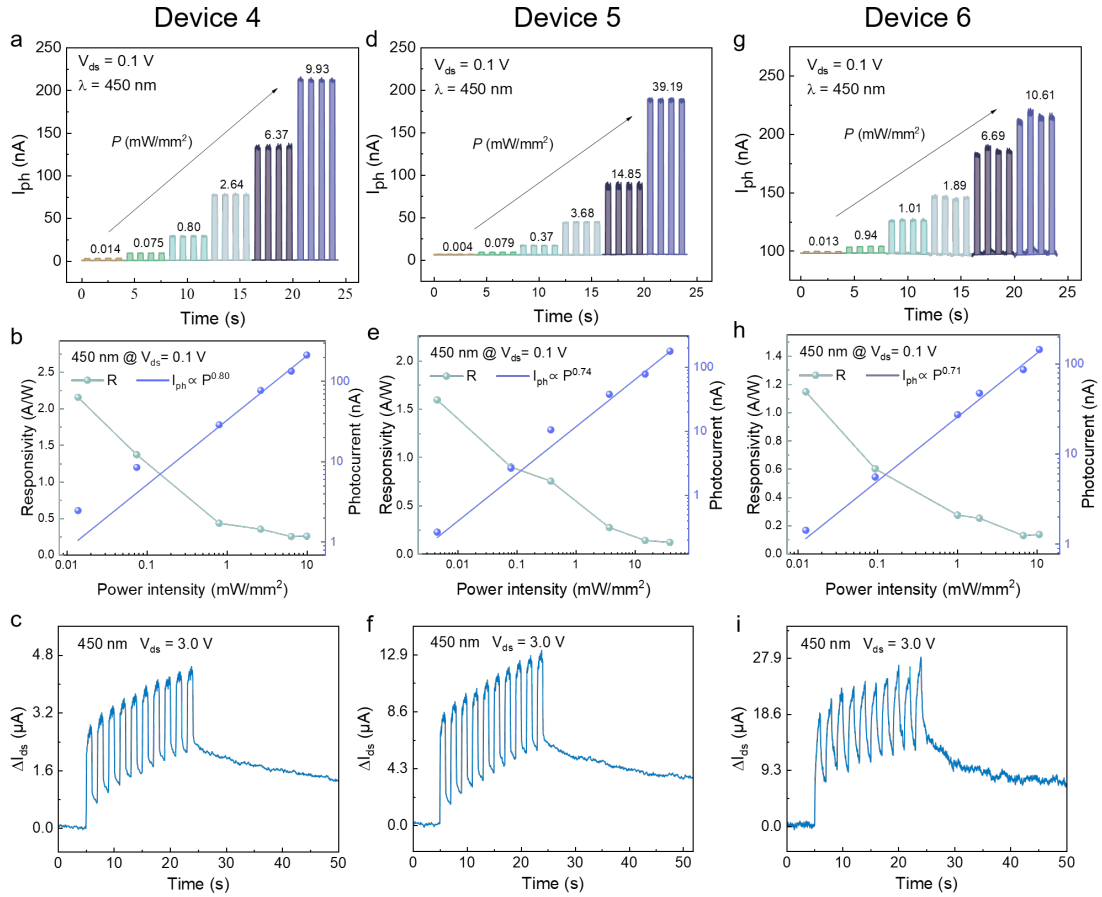


Figure R8 Photoresponse characteristics of devices with different thicknesses. (a, d, g) Low-bias time-resolved switching response under 450 nm illumination of the 3 devices, showing stable and repeatable photocurrent modulation. (b, e, h) Photocurrent and responsivity as a function of incident optical power density under 450 nm illumination of the 3 devices. (c, f, i) Photocurrent response under higher bias, revealing a PPC effect with slow decay behavior.

[Actions]:

We have made the corresponding revisions on page 6 of the revised manuscript, Supplementary Fig. 13- 15.

[Comments 5]: Although the device exhibits attractive photodetection performance, including sub-microsecond response time and mA/W-level responsivity, the manuscript does not provide sufficient physical insight into the origin of these properties. The underlying mechanisms responsible for the high performance are not adequately analyzed. A more comprehensive physical model, supported by experimental data, is required to explain the observed behavior.

[Response]:

We sincerely thank the reviewer for this constructive comment. We fully agree with the assessment and provide the following detailed analysis.

The high photodetection performance at low bias ($V_{ds} = 0.1$ V) can be attributed to the direct tunneling transport mode. Under DT, photogenerated holes traverse the interfacial barrier via quantum tunneling without the need to acquire thermal energy, which greatly shortens the carrier transit time $\tau_{transit}$. According to the photoconductive gain relation:

$$G = \frac{\tau_{lifetime}}{\tau_{transit}}$$

Where G is the gain, $\tau_{lifetime}$ is the carrier lifetime, and $\tau_{transit}$ is the carrier transit time. Reduction directly enhances the gain, thereby contributing to the observed high responsivity while maintaining a fast sub-microsecond response speed.

Beyond the high performance at low bias, the bias-dependent evolution of the transport mechanism is central to the device's reconfigurable functionality. To quantitatively describe this behavior, we developed a multi-channel transport model, as presented below.

1. Band structure foundation established by first-principles calculations

We first employed first-principles calculations to study the electronic structure of Ta_2NiSe_5 , $MoTe_2$, $PdSe_2$, and their heterojunctions (Figure R2).

After the formation of the $Ta_2NiSe_5/MoTe_2$ heterojunction, a conduction band offset of approximately 0.74 eV is created (Figure R9). This conduction band offset may play a dual role:

(1) First, it largely separates the transport paths of electrons and holes, such that the response time is primarily governed by the hole transport mechanism. Photogenerated electrons are confined to the Ta_2NiSe_5 side and cannot easily overcome the conduction band offset to enter $MoTe_2$. Therefore, recombination relies mainly on holes crossing the interface, making the photoresponse speed sensitive to the hole transport mechanism. When holes cross by the direct tunneling, the device exhibits a fast response; when they need to overcome the barrier by slow thermal excitation, the response time extends to the slow regime. This conduction band offset thus enables the hole transport mechanism to largely influence the overall response time with less interference from electron transport.

(2) Second, it provides a favorable condition for the appearance of PPC and long-term memory

effects. Under high bias, when the DT channel for holes is suppressed, photogenerated holes tend to remain on the Ta₂NiSe₅ side and become spatially separated from the blocked electrons, significantly reducing recombination and giving rise to PPC.

(3) Simultaneously, the VBM of Ta₂NiSe₅ (−0.18 eV) is higher than that of MoTe₂ (−0.53 eV), yielding a valence band offset of about 0.35 eV. This provides a bias-tunable interfacial barrier for holes. As the external electric field increases, the energy levels on both sides shift upward collectively, with the valence band edge dominated by the Ni–Se orbitals of Ta₂NiSe₅ and with the Te/Mo states of MoTe₂ participating in the interfacial coupling, leading to noticeable changes in the width and effective height of the valence band barrier.

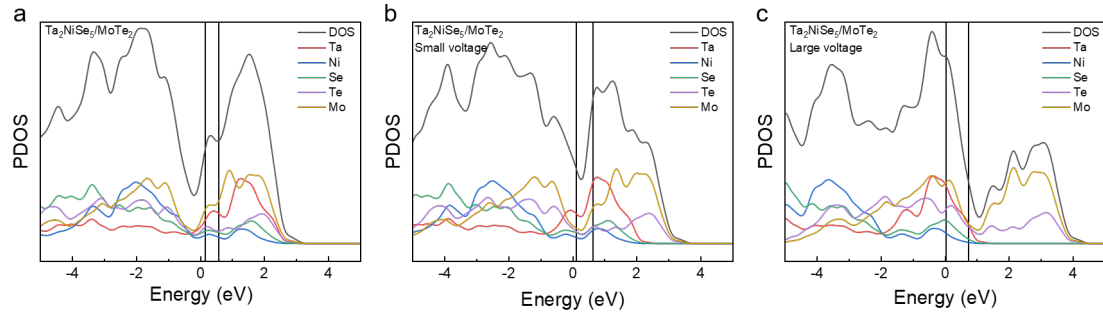


Figure R9 First-principles calculations of the electronic structure. (a–c) Density of states of the Ta₂NiSe₅/MoTe₂ heterostructure under zero bias, small bias, and large bias, showing the total DOS and element-projected contributions. Energy is referenced to the Fermi level in all panels.

2. Experimental identification of the two transport modes

At $V_{ds} = 0.1$ V, the photocurrent is nearly independent of temperature in the range of 80–300 K (Figure R10). This is consistent with tunneling-dominated transport.^[1]

At $V_{ds} = 3$ V, the photocurrent exhibits a strong temperature dependence, following the thermal excitation relation:

$$J = AT^2 \exp\left(-\frac{\phi}{k_B T}\right)$$

The extracted activation energy is consistent with the barrier height. This supports that carriers overcome the barrier mainly by thermal excitation.

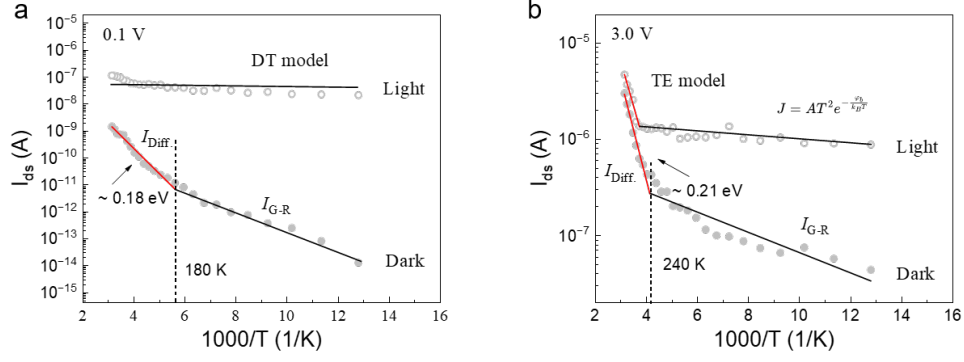


Figure R10 Temperature-dependent optoelectronic measurements of $V_{ds} = 0.1$ V and $V_{ds} = 3$ V.

Furthermore, we measured the bias-dependent photocurrent at both low temperature (80 K) and room temperature (300 K) (Figure R11). At low bias, the photoresponse is fast at both temperatures, consistent with direct tunneling that is insensitive to thermal activation. At high bias, the photoresponse at 80 K remains fast, while at 300 K the same bias produces obviously slow decay with PPC effect. This obvious temperature contrast at high bias indicates that the slow response is thermal excitation effect, largely suppressed at low temperature and becoming dominant at room temperature.

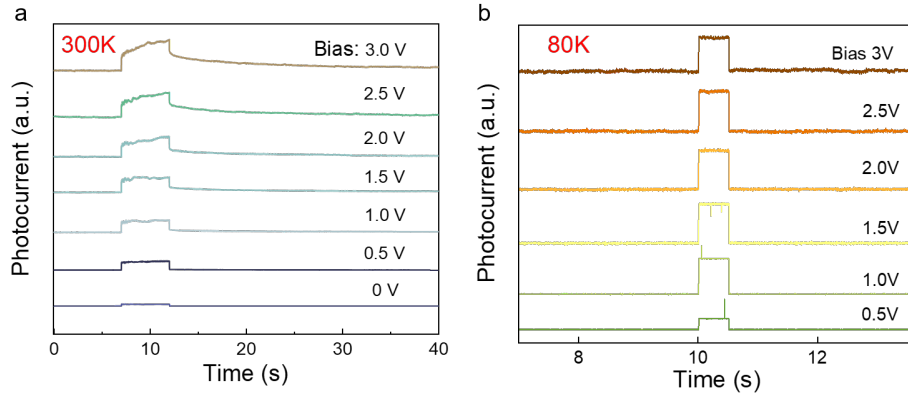


Figure R11 Bias-dependent photocurrent measured at 80 K and 300 K.

To further identify the transition between these two regimes, we analyzed the bias-dependent photocurrent characteristics. By plotting $\ln(I/V^2)$ versus $1/V$, two distinct regions emerge: at low bias, the curve exhibits behavior characteristic of direct tunneling; as the bias increases to approximately $V_{ds} \approx 0.5$ V, a clear change in the slope is observed, suggesting that the DT channel is gradually suppressed, and thermal excitation begins to take over (Figure R12). The experimental data can be described by the DT current expression:

$$I_{DT} = V \exp\left(-\frac{4\pi\sqrt{2m^*\phi}}{h}\right)$$

This transition point coincides roughly with the bias at which the response time jumps from the sub-microsecond to the millisecond-second regime in our experiments.

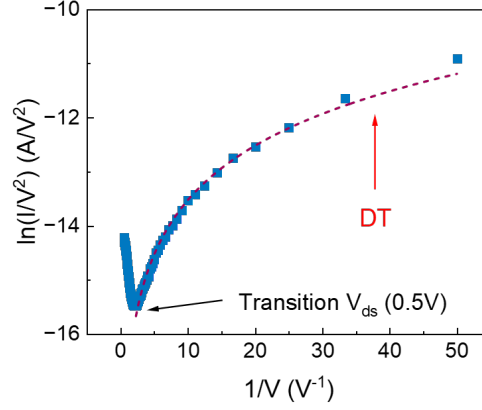


Figure R12 Fowler–Nordheim plots of the device at positive V_{ds} under illumination.

Additionally, we have performed systematic temperature-dependent I–V measurements in the dark at various bias voltages and extracted the effective barrier height $\phi(V)$ via Arrhenius fitting (Figure R13). ϕ exhibits a non-monotonic V-shaped variation: it decreases from 0.18 eV at 0.1 V to a minimum of ~ 0.10 eV around 0.5 V, and then monotonically rises to 0.29 eV at 3 V. The physical origin of this behavior can be understood as follows: at low bias, the external electric field partially counteracts the built-in field, narrowing the depletion region and making the barrier thinner and lower; once the bias exceeds a threshold of ~ 0.5 V, the depletion region may extend through the entire MoTe_2 layer and the barrier height rises again. This non-monotonic behavior provides rather direct experimental evidence for the abrupt suppression of the DT channel near 0.5 V.

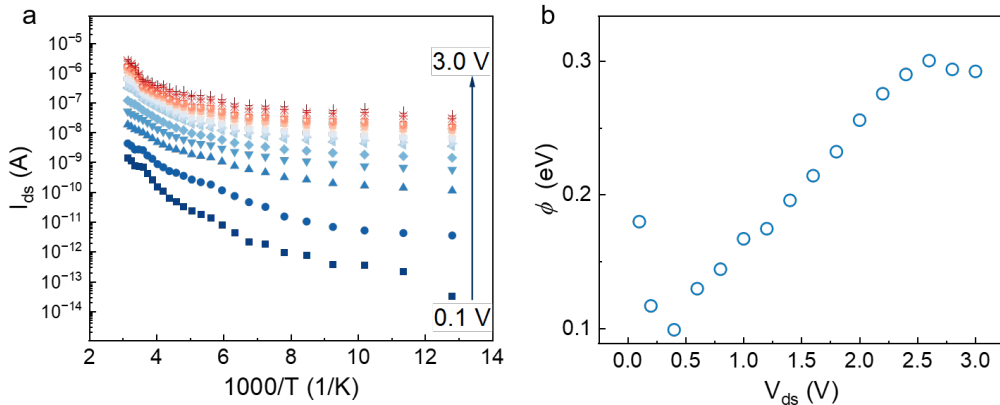


Figure R13. Temperature-dependent I–V measurements in the dark at various bias voltages and the extracted effective barrier height

The spatial distribution of photocurrent provides visual evidence for the effect of bias voltage on the depletion region (Figure R14). Compared with the 0V, 0.1 V condition, at 3 V the primary photocurrent generation region shifts significantly toward the edge of the Ta₂NiSe₅/MoTe₂ interface, indicating an expansion of the depletion region.

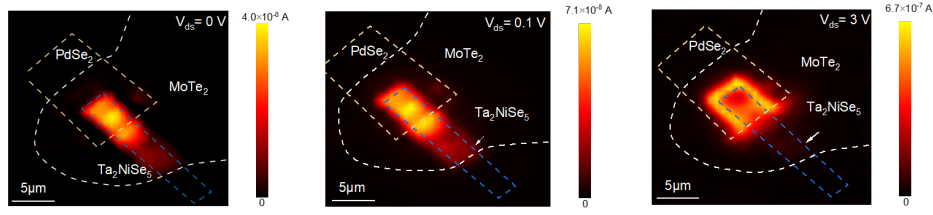


Figure R14 The spatially resolved photocurrent mapping images at $V_{ds} = 0$ V, $V_{ds} = 0.1$ V and $V_{ds} = 3$ V, respectively.

3. Transport model

Based on the above analysis, 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:

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

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:

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

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

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

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 is 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:

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

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.

To describe the competition between DT and TE mechanisms, a voltage-dependent weighting function $f(V)$ is introduced. This physical saturation behavior can be well described by a sigmoidal switching function, a form commonly used in semiconductor device modeling to describe smooth transitions between two limiting states:^[2-4]

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

Here, $f(V)$ represents the probability that charge transport occurs via the thermal excitation channel, while $1 - f(V)$ corresponds to the tunneling contribution. This formulation ensures a smooth and continuous transition between the two limiting transport regimes.

Physically, the sigmoidal form of $f(V)$ can be understood as a Fermi-like distribution arising from the competition between two transport pathways with different effective activation energies.

The transition voltage (V_{th}) corresponds to the point at which the contributions of DT and TE are comparable, while the parameter (γ) characterizes the sharpness of the transition and reflects the degree of energetic disorder or interface inhomogeneity in the device.

By substituting the experimentally determined parameters into the above equation, we calculated the theoretical response time over the full bias range. The resulting curve (Figure R15) reproduces the key features observed experimentally: a fast response at low bias, a sharp increase around 0.5 V, and a slow decay at high bias. The theoretical curve is in reasonably good agreement with our experimental data, which supports the validity of the proposed model and indicates that the seven-order-of-magnitude modulation of the response time originates primarily from the bias-driven DT-to-TE mechanism transition, with the trap states providing an additional contribution to the slow response at high bias.

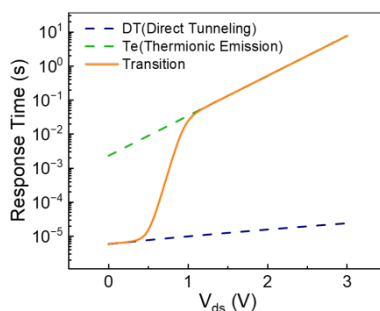


Figure R15. Calculated bias-dependent response time from the model, showing agreement with experimental data

- [1] Nature Communications 15, 141 (2024)
- [2] Nature 453, 80–83 (2008).
- [3] Nature Materials 12, 114–117 (2013).
- [4] Nature Communications 8, 15204 (2017).

[Actions]:

We have made the corresponding revisions on page 4, 5 of the revised manuscript, including Figs. 2a, 2b, 2f and Supplementary Figs. 9, 12.

[Comments 6]: *The synaptic characterization is relatively conventional and does not provide clear*

novelty. In addition, the physical origin of the strong bias dependence of the response speed is not discussed in sufficient depth. The explanation based solely on thermal emission appears oversimplified and does not fully account for the observed behavior.

[Response]:

We thank the reviewer for this comment. We address this concern from two aspects: the novelty in synaptic characterization and the physical origin of the strong bias dependence of the response speed.

1. We acknowledge that individual synaptic metrics such as long-term potentiation, paired-pulse facilitation, and frequency-dependent plasticity have been reported in previous optoelectronic synaptic devices. However, we would like to emphasize that the combination of broadband operation, frequency versatility, and wide dynamic range in a single, dynamically reconfigurable device represents an advance over conventional synaptic devices.

(i) Most previously reported optoelectronic synapses operate within a narrow spectral range, primarily in the visible or near-infrared region. In contrast, our device exhibits synaptic behavior across an broad wavelength range spanning from visible (450 nm) to mid infrared (3.8 μm) (Figure R16). This capability is significant because it allows the same synaptic device to process multispectral information, including wavelengths relevant to night vision, thermal sensing, and gas detection, without the need for spectral filtering or multiple detector arrays. To our knowledge, synaptic devices with response extending to 3.8 μm are still uncommon, and this broad bandwidth synaptic functionality opens new possibilities for neuromorphic vision systems that integrate visible and infrared information for improved perception in challenging environments such as fog, smoke, and low light.

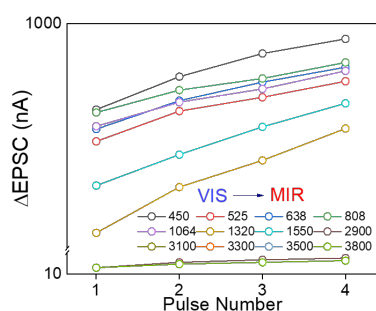


Figure R16. EPSC triggered by optical spikes across a broad spectrum from 450 nm to 3800 nm

(ii) Many reported synaptic devices are characterized at a single frequency or over a limited range, which does not fully capture their temporal processing capabilities. Our device operates across a wide frequency range from 0.1 Hz to 100 Hz, covering different timescales relevant to biological sensory processing (Figure R17). Low frequencies (0.1–1 Hz) are suitable for sustained memory and slow adaptation; moderate frequencies (5–10 Hz) are relevant to natural visual and auditory processing; and high frequencies (up to 100 Hz) enable temporal summation and rapid event detection. This frequency versatility allows the same device to adapt to different temporal processing demands, from slow environmental monitoring to fast dynamic scene analysis.

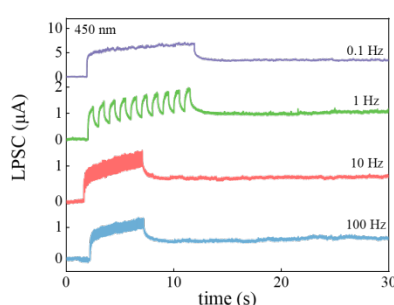


Figure R17. Frequency-dependent synaptic response of the device across a wide operating range from 0.1 Hz to 100 Hz.

(iii) While individual synaptic devices have demonstrated either fast or slow dynamics, only a few can continuously tune their response time over a wide range, and devices covering seven orders of magnitude in a single device are, to our knowledge, still uncommon. Our bias-reconfigurable design allows the response time to be continuously modulated from ~600 ns to ~8 s, effectively bridging the gap between ultrafast photodetection and long-term memory.

Furthermore, a comprehensive quantitative model that includes direct tunneling, thermal excitation, and trap-assisted processes has been presented in our response to Comments 5.

[Actions]:

We have made the corresponding revisions on page 10 of the revised manuscript and Supplementary Figs. 27.

[Comments 7]: *The energy-consumption analysis is unclear. In Fig. 4e, the authors present energy consumption values at different wavelengths, but do not explain how these values are calculated. It is also unclear whether the reported values include only optical energy or also account for electrical*

energy consumption during device operation.

[Response]:

We thank the reviewer for this insightful comment. In the original manuscript, the energy consumption values presented in Fig. 4e were calculated based on the electrical energy consumed during device operation. For a single synaptic event, the energy consumption is given by:

$$E = \int_{t_0}^{t_1} V \cdot I(t) dt$$

where V is the applied bias voltage, and I is the EPSC current under optical pulse excitation. This calculation reflects the intrinsic electrical energy consumed by the device under a single optical pulse. Taking the 450 nm illumination as an example, at a driving voltage of 0.8 V, yielding an electrical energy consumption of approximately 109 fJ per synaptic spike (Figure R18). The same calculation method and pulse conditions were applied to the other three wavelength bands (638 nm, 1064 nm, and 1550 nm) reported in Fig. 4e. This value facilitates comparison with other optoelectronic synaptic devices, as electrical driving energy is commonly reported as an important figure of merit for energy-efficient operation. Similar calculation approaches have been widely used for evaluating the energy consumption of various optoelectronic synaptic devices. [1–3]

[1] *Nature nanotechnology*, 14, 776-782, 2019.

[2] *Nano Letters*, 25, 16468-16476, 2025.

[3] *Nature Nanotechnology*, 18, 882-888, 2023.

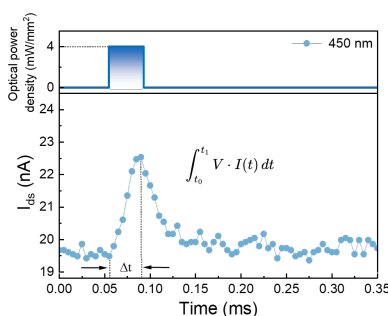


Figure R18. Diagram of energy consumption measurement methods for a single synaptic event.

[Actions]:

We have made the corresponding revisions on page 11 of the revised manuscript and Supplementary Figs. 30.

[Comments 8]: The presentation of Fig. 4h is not sufficiently clear. Although 16 different output

states are shown, the current format does not allow straightforward comparison. In addition, given the inherent variability in temporal decay processes, the absence of repeated measurements and statistical analysis raises concerns regarding the robustness of the results.

[Response]:

We thank the reviewer for this constructive comment.

In response to the concern about data presentation, we have revised Fig. 4h by reorganizing the output states into a clearer format that allows for more straightforward comparison between different conductance levels. The updated figure improves visual readability and highlights the distinction among the 16 states more effectively. Furthermore, we conducted ten repetitions of tests for each optical pulse combination, performed statistical calculations, and included the indication of the error range (standard deviation) to quantitatively reflect the differences among the multiple measurement results.

To address the concern regarding variability in temporal decay processes, we have performed repeated measurements and added new statistical data. Specifically, the device response corresponding to each state has been measured multiple times, and the results are now presented in the revised figures (Figure R19, Figure R20). The repeated measurements demonstrate good consistency in both amplitude and decay behavior across cycles.

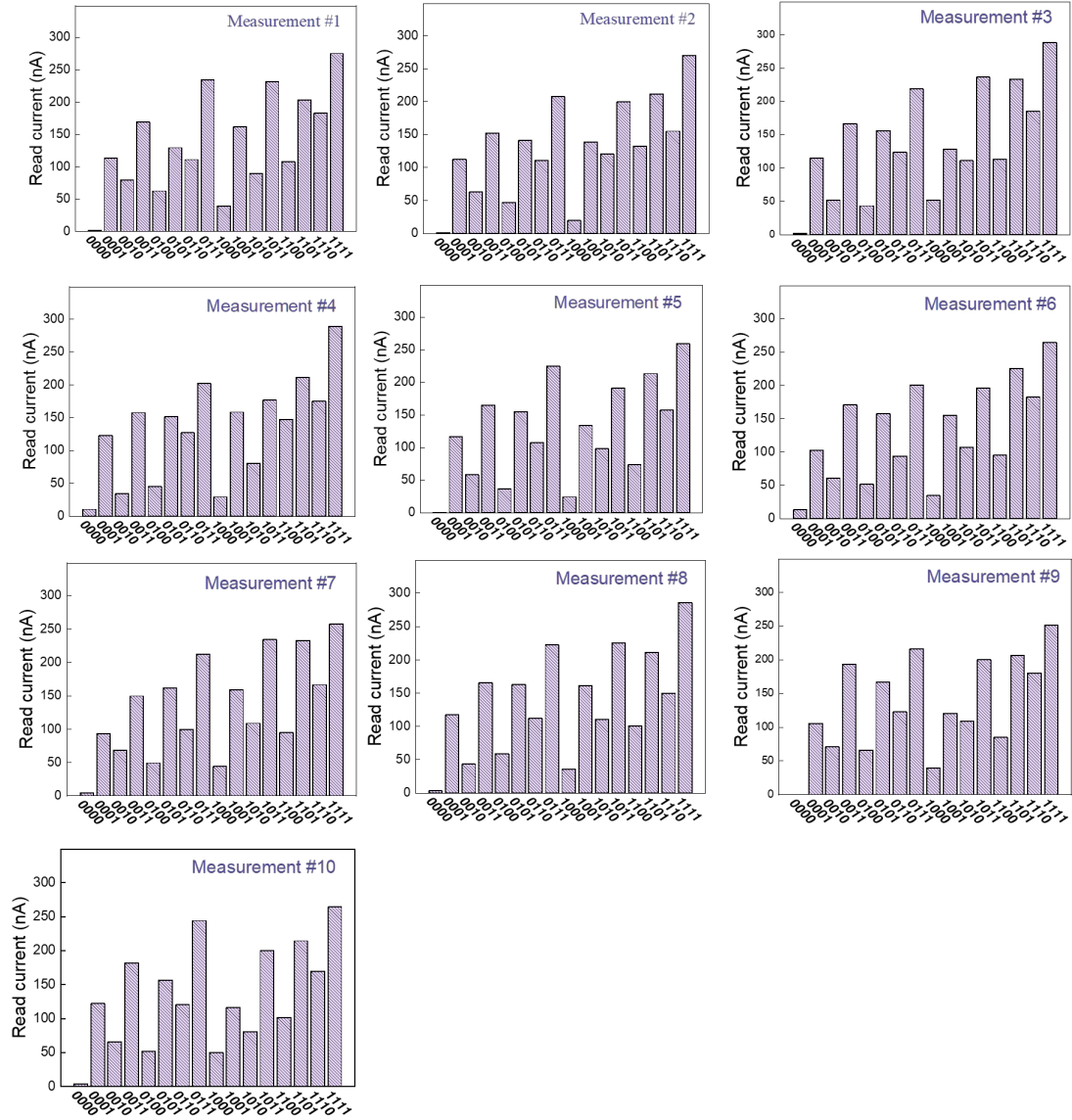


Figure R19. Statistical and temporal characteristics of multi-level photoresponse states.

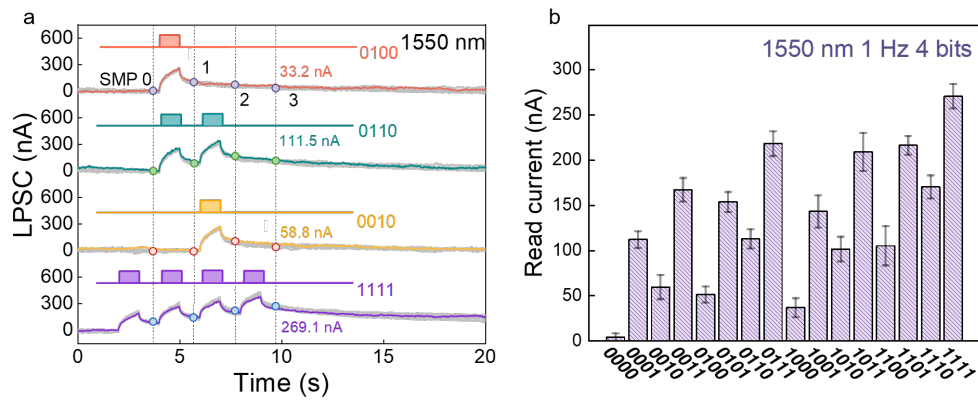


Figure R20. Statistical and temporal characteristics of multi-level photoresponse states. a. Sixteen distinct conductance states are achieved through controlled optical programming. The temporal

evolution of each state is plotted to show their decay dynamics over time. b. Each state was measured over multiple cycles ($n = 10$), and the averaged response is presented with error bars indicating the standard deviation. The results demonstrate good reproducibility, with minimal variation in both initial conductance levels and decay behavior.

[Actions]:

We have made the corresponding revisions on page 11 of the revised manuscript, including Fig. 4g, h and Supplementary Figs. 31.

[Comments 9]: *The connection between the tunable response dynamics and neuromorphic vision is not convincingly demonstrated. While the manuscript emphasizes the capability of integrating high-speed sensing and neuromorphic computation, the application demonstration does not utilize the fast response regime, nor does it experimentally validate reservoir computing functionality. Instead, the demonstration relies on the slow, volatile photoresponse, which has been widely reported in previous studies. As a result, this part lacks novelty and does not substantiate the claims made in the Introduction. Furthermore, no device array is fabricated or experimentally characterized, and the application is supported only by simulations, which significantly weakens the overall impact. The authors should design application scenarios that explicitly leverage the tunable response dynamics and experimentally validate these using device arrays. Discussions on scalability, device uniformity, and cycle-to-cycle stability are also necessary.*

[Response]:

We thank the reviewer for the constructive comments. We have made two major revisions to address these concerns. First, we designed an additional application scenario that demonstrates the combined use of fast and slow response dynamics. Second, we have fabricated device arrays and experimentally validated their scalability, uniformity, and cycle-to-cycle stability. The details are provided below.

1. First, we illustrate the collaborative framework in Figure R21. The input scene represents a dynamic traffic environment with moving vehicles under complex background conditions (Figure R21a). In Mode A (fast response, $V_{ds} = 0.1$ V), the device operates as a photodetector that captures instantaneous spatial information of the moving object but contains no temporal memory. In Mode

B (slow response, $V_{ds} = 3.0\text{ V}$), the device functions as a photomemristor that accumulates temporal signals and generates motion-dependent trailing features, although the output is severely degraded by environmental noise and device fluctuations. In Mode C (fusion mode), a spatial mask extracted from the fast-mode snapshot is applied to the temporally integrated signal from the slow mode, effectively suppressing background noise while preserving motion trajectories (Figure R21b). As shown in Figure R21c, Mode A and B exhibit low and unstable accuracy due to the lack of temporal information and noise interference, respectively, whereas Mode C achieves significantly improved and stable performance, demonstrating the effectiveness of the collaborative dynamics.

2. Second, we then quantitatively evaluate these three modes using a velocity classification task with video sequences of moving objects. Mode A produces only a single-frame intensity snapshot that retains no motion history. When a neural network performs motion inference using only single-frame spatial features, it cannot reliably extract velocity information. In a three-class velocity recognition task, Mode A achieved an accuracy of about 53%, and the corresponding confusion matrix (Figure R21d) shows near-random predictions across the three speed categories, confirming the absence of motion encoding. Mode B generates motion trailing through leaky integration of multiple frames. Although the trailing length theoretically encodes velocity, intrinsic device noise significantly contaminates the signals and prevents effective feature extraction. In the same task, Mode B achieved an accuracy of only about 50%, and its confusion matrix (Figure R21e) exhibits degraded and scattered classification patterns due to noise contamination in the accumulated signal. This shows that while the slow mode possesses the necessary temporal accumulation capability, its practical performance is limited by noise. Mode C combines the complementary strengths of the two modes (Figure R21f). The fast-mode snapshot, with its high spatial precision, is processed by an edge detection algorithm to extract the contour of the moving object. This contour is then used as a spatial mask applied to the temporally accumulated signal from the slow mode. The mask suppresses background noise and preserves only the motion trailing within the object contour. In the same velocity classification task, Mode C reached an accuracy of about 90%, and its confusion matrix shows clear diagonal dominance, indicating accurate and robust classification enabled by the noise-suppressed temporal features.

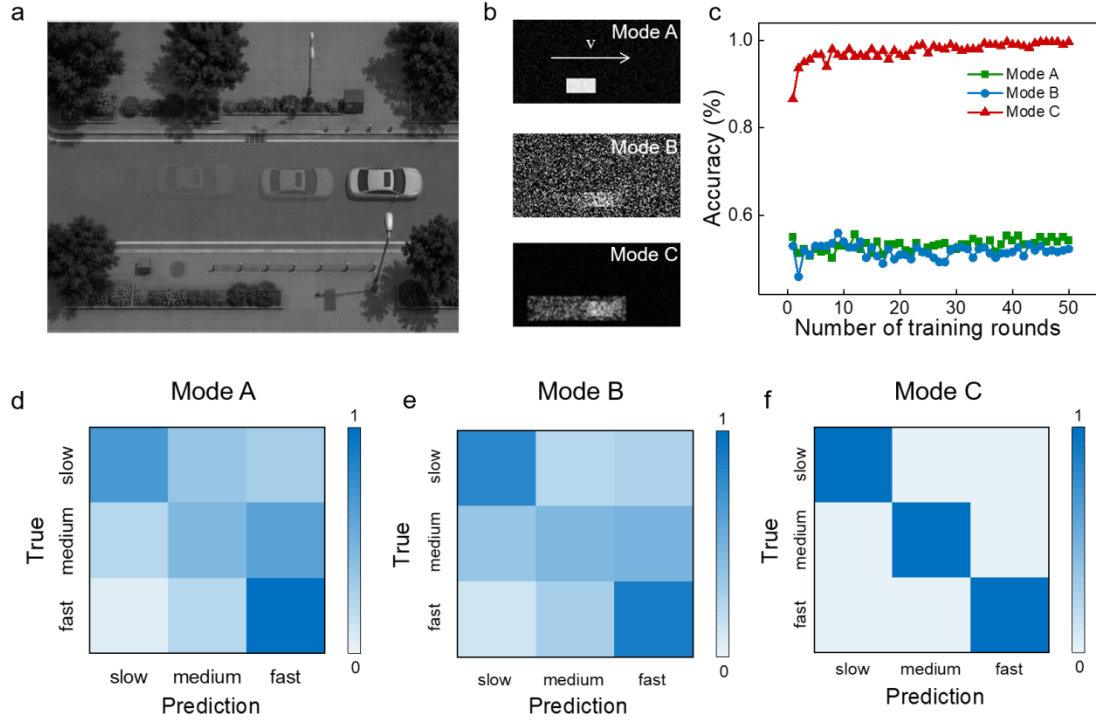


Figure R21 Tunable-response-enabled collaborative sensing and computation for motion perception. (a) Dynamic traffic scene with complex background. (b) Mode A (fast): instantaneous spatial snapshot, no temporal memory. Mode B (slow): temporal accumulation with motion trailing, degraded by noise. Mode C (fusion): a spatial mask from Mode A is applied to Mode B, suppressing background noise and preserving motion trajectories. (c) Classification accuracy comparison. (d–f) Confusion matrices for Mode A, Mode B, and Mode C, respectively. Mode C shows clear diagonal dominance, indicating robust velocity classification.

These results indicate that the fast mode can provide spatial references that help mitigate the noise sensitivity of the slow mode, while the slow mode contributes temporal accumulation that compensates for the fast mode's blindness to motion. The performance gain achieved through their synergy supports the practical value of the tunable response dynamics and aligns with our core claim of integrating high-speed sensing and neuromorphic processing within a single device.

2. Second, we have fabricated a 3×3 device array and characterized its performance (Figure R22). In this array, the PdSe₂ and MoTe₂ layers were grown by chemical vapor deposition, while the Ta₂NiSe₅ flakes were prepared by mechanical exfoliation, as the CVD growth of Ta₂NiSe₅ remains challenging at present. All nine devices were tested under both low bias and high bias. Under low bias, all devices exhibited fast photoresponse, while under high bias, all devices showed

slow response. This consistent bias-tunable behavior across the array demonstrates good device-to-device uniformity and reproducibility.

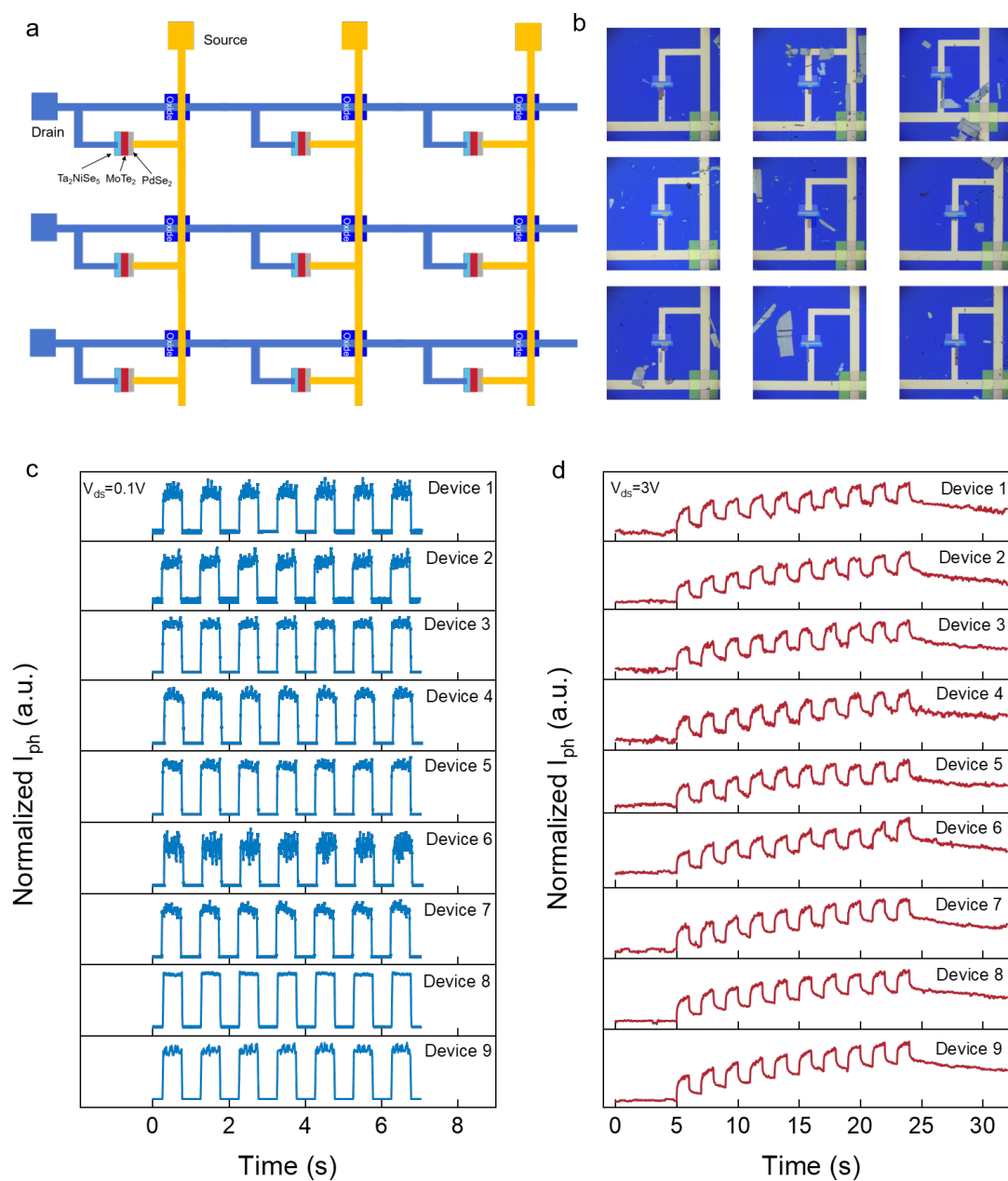


Figure R22 (a) Schematic illustration of the 3×3 device array based on the Ta₂NiSe₅/MoTe₂/PdSe₂ heterostructure. (b) Optical image of the 3×3 device array. (c) Photoresponse of the 3×3 device array under low and high bias, showing consistent bias-tunable behavior across all nine devices.

[Actions]:

We have made the corresponding revisions on page 7, 13 of the revised manuscript, including Fig. 5e and Supplementary Figs. 17.

[Comments 10]: Finally, there is a textual error at line 158, where “Under no illumination” should be corrected to “Under illumination.”

[Response]:

We thank the reviewer for carefully identifying this textual error. We have corrected the statement at line 158 from "Under no illumination" to "Under illumination" in the revised manuscript. We have also carefully proofread the entire manuscript to ensure that similar typographical errors are eliminated.

We appreciate the reviewer's attention to detail, which has helped improve the clarity and accuracy of the manuscript.

Reviewer #2

This paper presents a bias-reconfigurable photodetector based on a Ta₂NiSe₅, MoTe₂, PdSe₂ van der Waals heterostructure. The work moves toward the realization of compact neuromorphic systems by enabling a single photodetector to switch between sensing and memory functions. This integration reduces the need for multiple hardware components and simplifies system architecture. The device exhibits multifunctionality, combining photodetection, memory effects, and neuromorphic behavior, along with broadband and polarization-sensitive detection capabilities. Overall, the manuscript presents a promising step toward hardware-efficient intelligent vision systems. While this concept is widely discussed at present, the core aspects of physical interpretation and experimental verification remain weak. I cannot support publication now because much improvement remains in terms of analysis, performance comparison, and technical details.

[Response]:

We sincerely thank the reviewer for the overview and constructive feedback on our manuscript, which helped us to further improve its quality. We have carefully addressed the concerns point-by-point as below.

[Comments 1]: *The proposed mechanism relies critically on the modulation of interfacial barrier height under applied bias. However, the claimed evolution of band alignment lacks simulation support and direct experimental evidence. Key metrics such as memory capacity and nonlinearity*

are not quantitatively evaluated. In addition, the demonstrated results rely on a relatively small, custom dataset without validation on standard benchmark tasks.

[Response]:

We sincerely thank the reviewer for the constructive comments. In the revised manuscript, we have addressed these issues in three main aspects: (1) band alignment simulations and experimental validation, (2) quantitative evaluation of synaptic and reservoir computing metrics, and (3) benchmarking on standard datasets. Our point-to-point responses are presented below.

1. First, we employed first-principles calculations to study the electronic structure of Ta_2NiSe_5 , MoTe_2 , PdSe_2 , and their heterojunctions (Figure R1). The band edge positions of the individual materials are approximately as follows: Ta_2NiSe_5 has its VBM at about -0.18 eV and CBM at about 0.17 eV, with a bandgap of roughly 0.35 eV; MoTe_2 has its VBM at about -0.53 eV and CBM at about 0.56 eV, with a bandgap of about 1.09 eV; PdSe_2 has its VBM at about -0.01 eV and CBM at about 0.41 eV, with a bandgap of about 0.42 eV. Element-projected density of states reveals that the valence band edge of Ta_2NiSe_5 is dominated by the hybridization of Se $4p$ and Ni $3d$ states, while that of MoTe_2 is primarily composed of Te $5p$ states, with Mo $4d$ states contributing significantly near the conduction band edge.

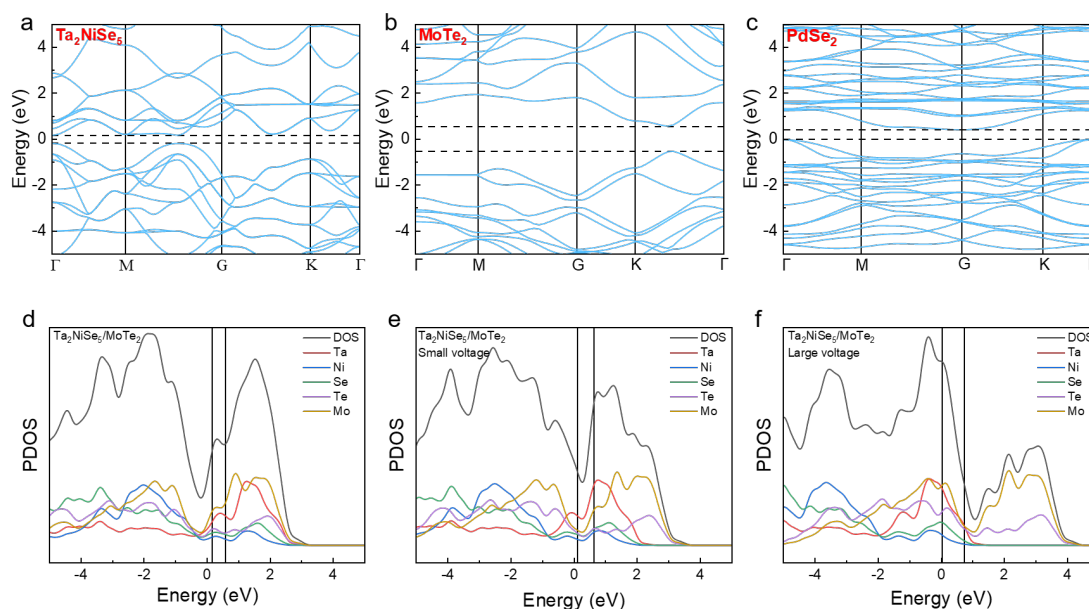


Figure R1 First-principles calculations of the electronic structure. (a–c) Band structures of the individual Ta_2NiSe_5 , MoTe_2 , and PdSe_2 , with the valence band maximum and conduction band minimum indicated. (d–f) Projected density of states of the $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2$ heterostructure 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.

After the formation of the Ta₂NiSe₅/MoTe₂ heterojunction, a conduction band offset of about 0.74 eV is created. To examine the influence of bias on the band alignment, we calculated the density of states under three conditions: zero bias, small bias, and large bias. The results show that the applied voltage mainly induces an overall rigid shift of the energy levels on both sides. Meanwhile, the VBM of Ta₂NiSe₅ (−0.18 eV) is higher than that of MoTe₂ (−0.53 eV), yielding a valence band offset of about 0.35 eV. This conduction band offset largely separates the transport paths of electrons and holes. Photogenerated electrons are confined to the Ta₂NiSe₅ side and cannot easily overcome the conduction band offset to enter MoTe₂. The recombination of photogenerated electron–hole pairs therefore proceed mainly via holes crossing the interface, so the photoresponse speed depends sensitively on the way holes traverse the interface. When holes cross by tunneling, the device exhibits a fast response; when they need to overcome the barrier by slow thermal excitation, the response time extends to a slow response.

Furthermore, to experimentally verify the dynamic modulation of the interfacial carrier transport barrier by the bias, we carried out temperature-dependent I–V measurements and extracted the effective barrier height $\phi(V)$ at various biases through Arrhenius fitting. The $\phi(V)$ decreases from 0.18 eV at $V_{ds} = 0.1$ V to a minimum of about 0.10 eV around 0.5 V, and then rises to 0.29 eV at 3 V. Besides the contribution from the interfacial band offset, it is also modulated by factors such as the depletion-region electric field, interfacial dipole layers, and trap states. Therefore, the significant variation of $\phi(V)$ with bias directly confirms that the applied electric field can effectively tune the carrier transport barrier near the interface, complementing the band alignment evolution revealed by DFT.

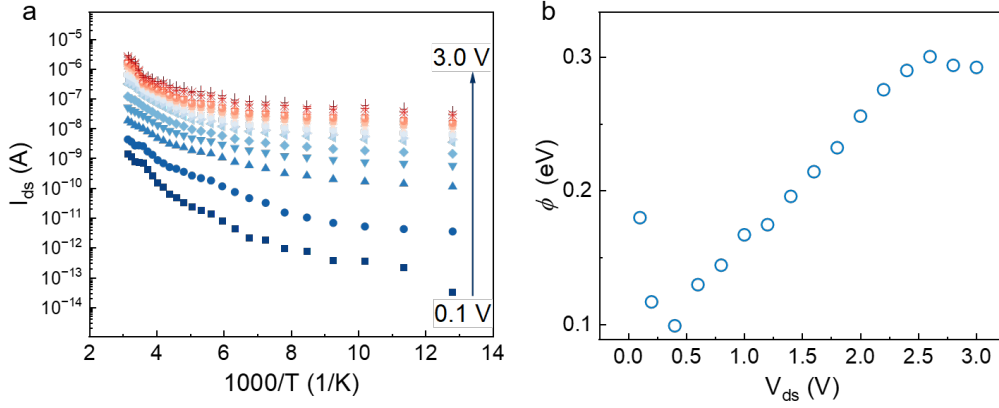


Figure R2. Temperature-dependent I–V measurements in the dark at various bias voltages and the extracted effective barrier height

2. Second, we have quantitatively evaluated the memory capacity and nonlinearity.

Figure R3a shows the cumulative evolution of the device current under consecutive pulse stimulation. The corresponding state distribution heatmap (Figure R3b) demonstrates that at least 128 distinguishable conductance states can be reliably accessed. This further supports the high state-space dimensionality and information capacity of the device.

Second, the nonlinearity is quantitatively characterized from two perspectives. On one hand, the normalized conductance evolution as a function of pulse number under different numbers of pulses (16 to 128) is shown in Figure R3c, where a clear nonlinear growth behavior is observed. Figure R3d shows how the dynamic range changes with the number of applied pulses. It can be observed that as the number of pulses increases, the dynamic range gradually expands and eventually saturates. The degree of nonlinearity is tunable with the applied pulse amplitude. On the other hand, we extract a nonlinearity factor ($|NL|$), defined based on the deviation of the device response from linear superposition of input pulse sequences. $|NL|$ is obtained from the following:

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

where G_{LTP} represents the conductance of the LTP curve; G_c is the fitting constant used to normalize the conductance range; L is the value of $|NL|$; P is the number of pulses applied; and $G_{\min}$ denotes the minimum conductance value.

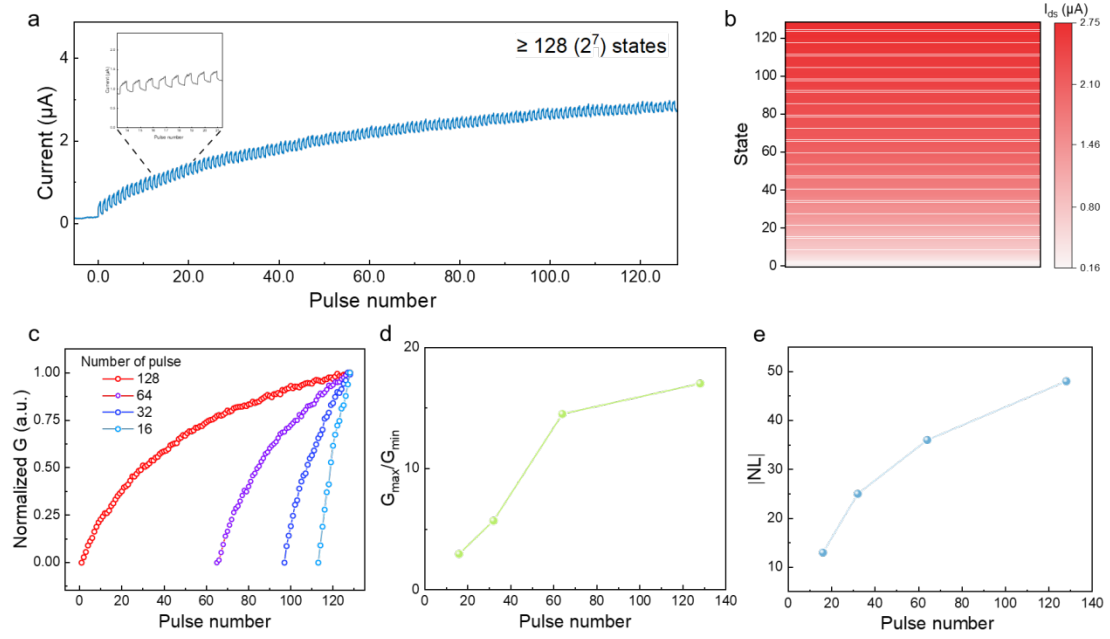


Figure R3 Quantitative evaluation of key reservoir computing metrics under pulsed operation. (a) Evolution of device current as a function of pulse number, along with (b) the corresponding state distribution heatmap, showing at least 128 distinguishable conductance states. (c) Normalized conductance versus pulse number under different pulse amplitudes, demonstrating pronounced and tunable nonlinear response characteristics. (d) The extraction dynamic range ($G_{\max}/G_{\min}$) as a function of pulse quantity. (e) Extracted nonlinearity factor (NL) as a function of pulse number, indicating progressively enhanced and saturating nonlinear mapping capability.

As presented in Figure R3e, the $|NL|$ value increases with pulse number, indicating that the device provides a progressively stronger nonlinear mapping capability during dynamic evolution. Such nonlinearity is essential for high-dimensional projection and complex temporal task processing.

The memory capacity (MC) is evaluated using a standard short-term memory (STM) task.³ Specifically, we calculate the squared correlation coefficient (Cor^2) between the device output and time-delayed input signals, and integrate over different delay steps to obtain the total memory capacity. As shown in Figure R4, 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.

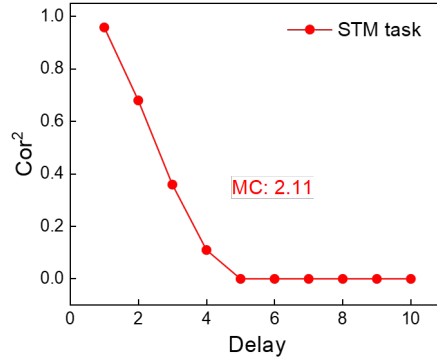


Figure R4 Memory capacity evaluation based on a STM task, showing the squared correlation coefficient (Cor^2) as a function of delay.

3. We have substantially extended our experimental validation by incorporating two widely used benchmark datasets, namely MNIST and Fashion-MNIST (f-MNIST), as shown in Figure R5. 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 “0” and a “dress” image from the f-MNIST dataset are presented in Figure R5a and c, respectively, demonstrating the capability of the device to encode and process distinct visual patterns. Quantitatively, as shown in Figure R5b, the system achieves a training accuracy of 99 % after 100 training epochs, and the corresponding confusion matrix confirms high recognition reliability. Furthermore, to evaluate performance on a more challenging task, the f-MNIST dataset was employed, where the categories exhibit higher inter-class similarity. As shown in Figure R5d, the system achieves an overall training accuracy of 90% across 10 categories, demonstrating robust pattern recognition capability under increased task complexity. These results clearly indicate that, beyond the originally used small custom dataset, the proposed system performs effectively on large-scale standard benchmarks with tens of thousands of samples.

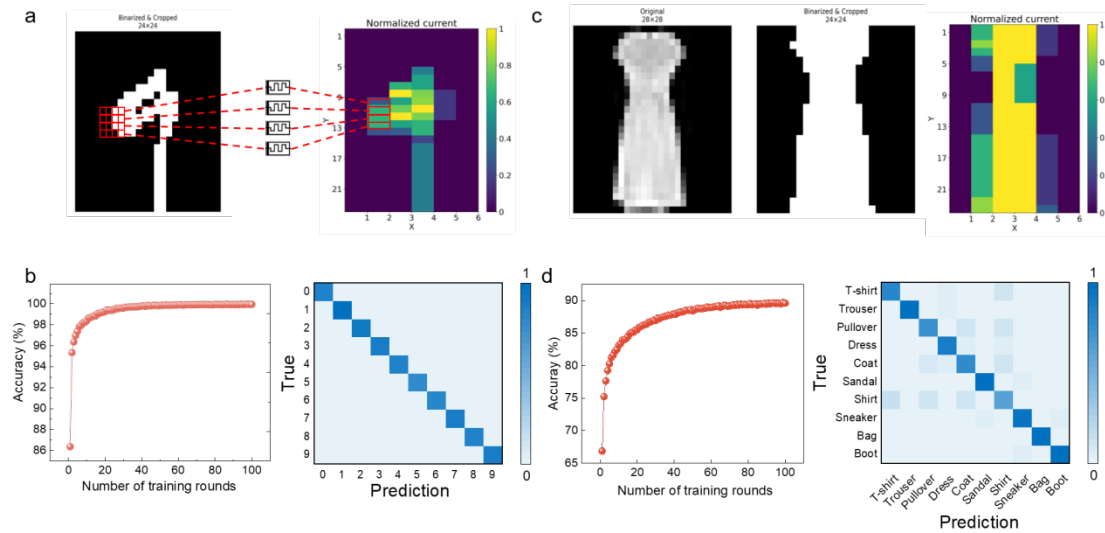


Figure R5. (a) Recognition of the handwritten digit "0" from the MNIST dataset, showing the reservoir output for digit "0". b) Recognition of a "dress" image from the Fashion-MNIST dataset, showing the reservoir output for "dress". c) Training accuracy after 100 epochs for digit pattern recognition and 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.

[1] *Nature Sensors*, 1-12, 2026

[2] *Advanced Functional Materials*, 36, e26966, 2026

[3] *IEEE Electron Device Letters*, 45, 108-111, 2023.

[Actions]:

We have made the corresponding revisions in page 5, 10, 13 and Supplementary Materials page 10, 34, 38.

[Comments 2]: The manuscript employs a Ta₂NiSe₅, MoTe₂, PdSe₂ heterostructure; however, its advantages over existing systems such as 2D neural network sensors, optoelectronic RRAM, and silicon-based platforms are not clearly articulated.

[Response]:

We sincerely thank you for pointing out the insufficient comparison with existing systems. Following your suggestion, we have substantially expanded the comparative discussion in both the Introduction and the revised manuscript. Below, we detail the advantages of our device over the

three representative categories: 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 (*Nature* 2020, 579, 62–66) in which individual devices exhibit a response time on the microsecond scale, but the photocurrent decays rapidly after light removal and lacks tunable persistent memory. Their subsequent 2D material-based classifier (*Nature* 2021, 597, 498–503) shows a response time constant of tens of microseconds, again without demonstrating long-term plasticity. Li et al. demonstrated a MoS₂-based optical synapse (*Nano Lett.* 2025, 25, 16468–16476) that emulates synaptic functions, but its response time is designed above the millisecond range and is incompatible with nanosecond-speed photodetection. Chen et al. reported a graded neuron sensor (*Nat. Nanotechnol.* 2023, 18, 882–888) that realizes motion perception through adjustable photoresponse, yet its speed modulation range is confined to the millisecond-to-second scale. 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. (*Nat. Nanotechnol.* 2019, 14, 776–782), whose conductance update time is in the microsecond-to-millisecond range and cannot achieve nanosecond response. Guo et al. reported a VO₂/WO₃ heterojunction device (*Small Methods* 2025, 9, e2400779) 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 tunable. Chen et al. demonstrated a 2D perovskite-based optoelectronic memristor (*Adv. Mater.* 2021, 33, 2004204) with multistate storage, yet the write/erase speeds are constrained by ionic kinetics, with

typical operation times in the sub-millisecond to millisecond range. Sun et al. employed an Ag/a-Si/TiO₂ structure (Nano Lett. 2020, 20, 2841–2848) that exhibits optical and electrical tunability but operates with discrete bi- or multi-level state switching at millisecond speeds. 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 (Nat. Electron. 2022, 5, 519–525); although high speed can be achieved, external gate bias circuitry is required, and the spectral response of silicon cannot reach the mid-infrared. Choi et al. reported a silicon-based floating-gate optoelectronic device (Science 2018, 360, 998–1003) capable of emulating synaptic plasticity, but weight updates also rely on external circuitry, and simultaneous broadband detection and polarization sensitivity were not demonstrated. More broadly, CMOS-based neuromorphic vision chips (Liao et al., IEEE J. Solid-State Circuits 2021, 56, 1972–1984) are powerful yet inherently multi-device circuit systems rather than a single device with intrinsic multimode reconfiguration. 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.

[Actions]:

We have made the corresponding revisions in Supplementary Materials page 2, 3.

[Comments 3]: *The reported modulation of response time from nanosecond level (0.1 V) to second level (3 V) spans over seven orders of magnitude. Attributing this dramatic change solely to barrier-height-induced transition from direct tunneling to thermal emission is not fully convincing, particularly given the observed long retention behavior. The authors should clarify how they exclude the possibility that high bias induced defects (e.g., due to the damaging in material under strong electric field) contribute to the super slow dynamics.*

[Response]:

We thank the reviewer for this important question. To assess whether the slow response at high bias could originate from bias-induced material damage, we carried out three sets of control experiments. First, we performed cyclic bias sweeps from 0.1 V to 3 V and back to 0.1 V. The device can be reversibly and repeatedly switched between the fast and slow response regimes, which suggests that the slow dynamics are not caused by permanent material damage, as such damage would be irreversible and would not allow recovery of the fast response upon returning to low bias (Figure R6). Second, we measured Raman spectra at both 0.1 V and 3 V, and no additional peaks indicative of structural damage were observed. Third, we measured the bias-dependent photocurrent at both low temperature (80 K) and room temperature (300 K). At low bias, the photoresponse is fast at both temperatures, consistent with direct tunneling that is insensitive to thermal activation. At high bias ($V_{ds} = 3$ V), the photoresponse at 80 K remains fast, while at 300 K the same bias produces a markedly slow decay with PPC effect (Figure R7). If the slow dynamics were dominated by field-induced damage, they would be expected to persist at low temperature.

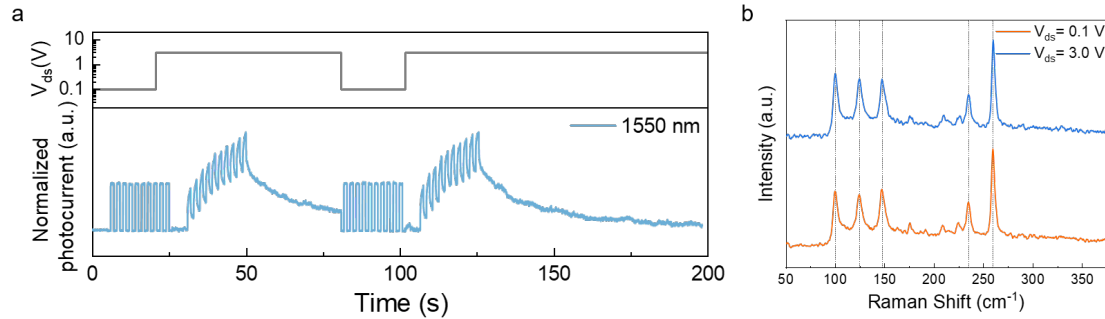


Figure R6. (a) Raman spectra of the device measured after prolonged operation under low bias and high bias. (b) Time-resolved photoresponse of the device under pulsed illumination (1550 nm) at different bias conditions.

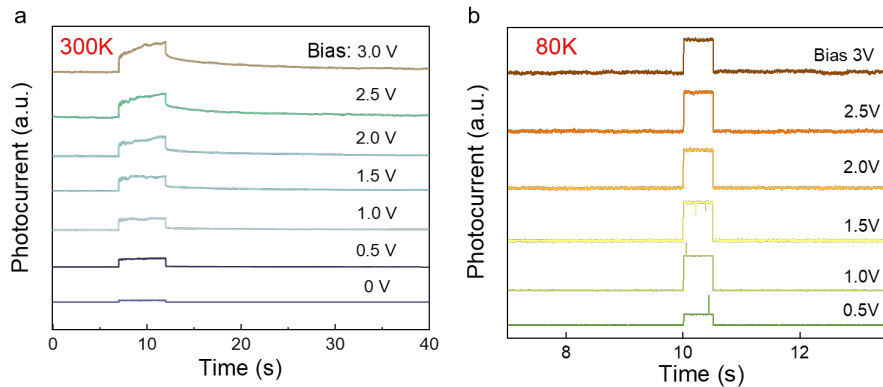


Figure R7 Bias-dependent photocurrent measured at 80 K and 300 K.

These results indicate that the slow response at high bias is not dominated by bias-induced material damage but is more likely attributed to the intrinsic bias-tunable transport mechanisms. To further clarify the underlying physics, we developed the quantitative multi-channel transport model as described below.

1. Band structure foundation established by first-principles calculations

A detailed description of the first-principles calculations of the band structure, including the band alignment of the individual materials and the heterojunction under different bias conditions, has been provided in our response to Comments 1.

2. Experimental identification of the two transport modes

At $V_{ds} = 0.1$ V, the photocurrent is nearly independent of temperature in the range of 80–300 K (Figure R3). This is consistent with tunneling-dominated transport. ^[1]

At $V_{ds} = 3$ V, the photocurrent exhibits a strong temperature dependence (Figure 1b), following the thermal excitation relation:

$$J = AT^2 \exp\left(-\frac{\phi}{k_B T}\right)$$

The extracted activation energy is consistent with the barrier height. This supports that carriers overcome the barrier mainly by thermal excitation.

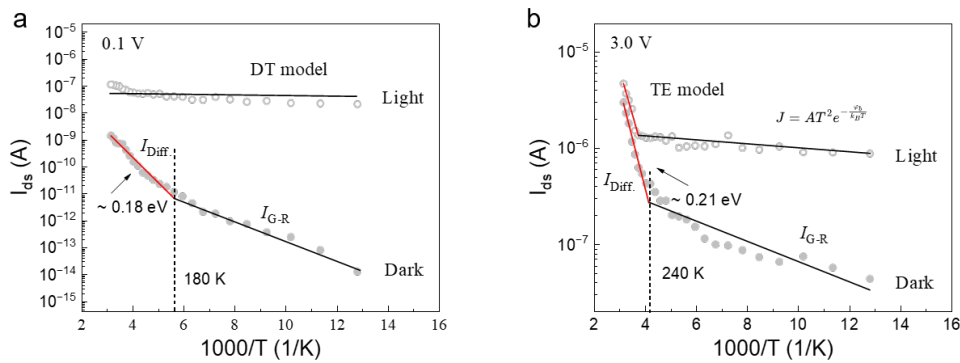


Figure R8 Temperature-dependent optoelectronic measurements of $V_{ds} = 0.1$ V and $V_{ds} = 3$ V.

Furthermore, we measured the bias-dependent photocurrent at both low temperature (80 K) and room temperature (300 K) (Figure R7). At low bias, the photoresponse is fast at both temperatures, consistent with direct tunneling that is insensitive to thermal activation. At high bias,

the photoresponse at 80 K remains fast, while at 300 K the same bias produces obviously slow decay with PPC effect. This obvious temperature contrast at high bias indicates that the slow response is thermal excitation effect, largely suppressed at low temperature and becoming dominant at room temperature.

To further identify the transition between these two regimes, we analyzed the bias-dependent photocurrent characteristics. By plotting $\ln(I/V^2)$ versus $1/V$, two distinct regions emerge: at low bias, the curve exhibits behavior characteristic of direct tunneling; as the bias increases to approximately $V_{ds} \approx 0.5$ V, a clear change in the slope is observed, suggesting that the DT channel is gradually suppressed, and thermal excitation begins to take over (Figure R9). The experimental data can be described by the DT current expression:

$$I_{DT} = V \exp\left(-\frac{4\pi\sqrt{2m^*}\phi}{h}\right)$$

This transition point coincides roughly with the bias at which the response time jumps from the sub-microsecond to the millisecond-second regime in our experiments.

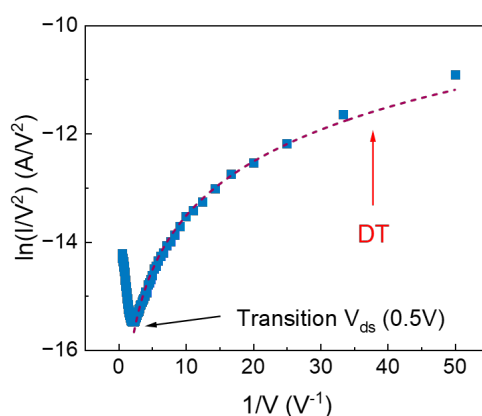


Figure R9 Fowler–Nordheim plots of the device at positive V_{ds} under illumination.

Additionally, we have performed systematic temperature-dependent I–V measurements in the dark at various bias voltages and extracted the effective barrier height $\phi(V)$ via Arrhenius fitting (Figure R10). ϕ exhibits a non-monotonic V-shaped variation: it decreases from 0.18 eV at 0.1 V to a minimum of ~ 0.10 eV around 0.5 V, and then monotonically rises to 0.29 eV at 3 V. The physical origin of this behavior can be understood as follows: at low bias, the external electric field partially counteracts the built-in field, narrowing the depletion region and making the barrier thinner and

lower; once the bias exceeds a threshold of ~ 0.5 V, the depletion region may extend through the entire MoTe_2 layer and the barrier height rises again. This non-monotonic behavior provides rather direct experimental evidence for the abrupt suppression of the DT channel near 0.5 V.

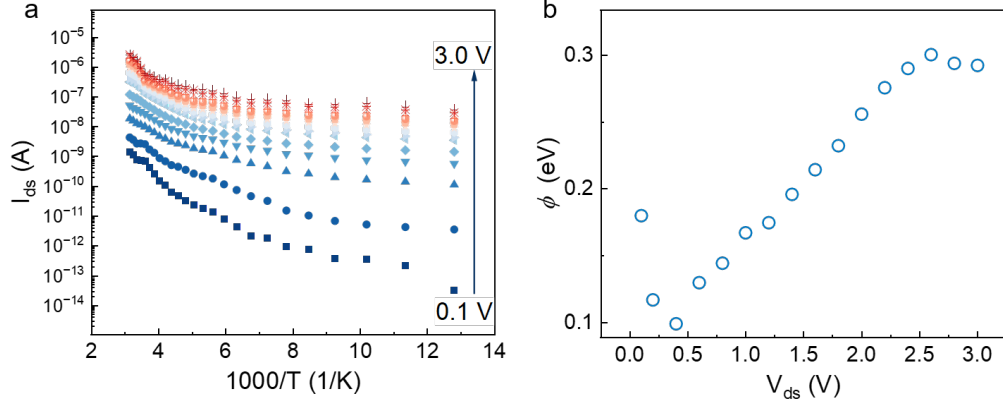


Figure R10. Temperature-dependent I–V measurements in the dark at various bias voltages and the extracted effective barrier height

The spatial distribution of photocurrent provides visual evidence for the effect of bias voltage on the depletion region (Figure R11). Compared with the 0V, 0.1 V condition, at 3 V the primary photocurrent generation region shifts significantly toward the edge of the $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2$ interface, indicating an expansion of the depletion region.

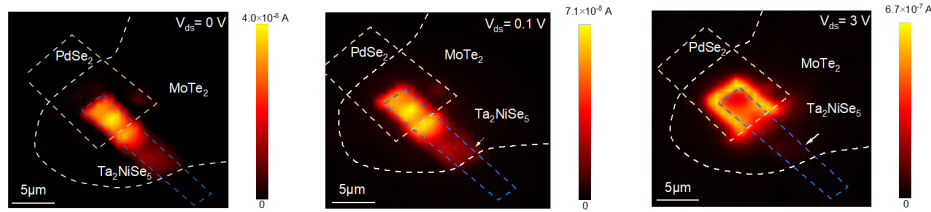


Figure R11 The spatially resolved photocurrent mapping images at $V_{ds} = 0$ V, $V_{ds} = 0.1$ V and $V_{ds} = 3$ V, respectively.

3. Transport model

Based on the above analysis, 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:

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

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:

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

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

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

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:

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

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.

To describe the competition between DT and TE mechanisms, a voltage-dependent weighting function $f(V)$ is introduced. This physical saturation behavior can be well described by a sigmoidal

switching function, a form commonly used in semiconductor device modeling to describe smooth transitions between two limiting states:^[2-4]

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

Here, $f(V)$ represents the probability that charge transport occurs via the thermal excitation channel, while $1 - f(V)$ corresponds to the tunneling contribution. This formulation ensures a smooth and continuous transition between the two limiting transport regimes.

Physically, the sigmoidal form of $f(V)$ can be understood as a Fermi-like distribution arising from the competition between two transport pathways with different effective activation energies. The transition voltage (V_{th}) corresponds to the point at which the contributions of DT and TE are comparable, while the parameter (γ) characterizes the sharpness of the transition and reflects the degree of energetic disorder or interface inhomogeneity in the device.

By substituting the experimentally determined parameters into the above equation, we calculated the theoretical response time over the full bias range. The resulting curve (Figure R12) reproduces the key features observed experimentally: a fast response at low bias, a sharp increase around 0.5 V, and a slow decay at high bias. The theoretical curve is in reasonably good agreement with our experimental data, which supports the validity of the proposed model and indicates that the seven-order-of-magnitude modulation of the response time originates primarily from the bias-driven DT-to-TE mechanism transition, with the trap states providing an additional contribution to the slow response at high bias.

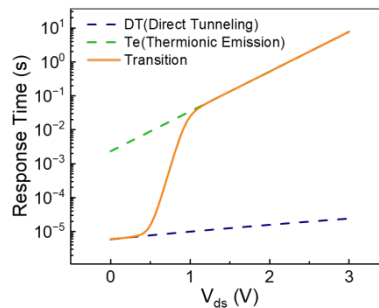


Figure R12. Calculated bias-dependent response time from the model, showing agreement with experimental data

- [1] Nature Communications 15, 141 (2024)
- [2] Nature 453, 80–83 (2008).
- [3] Nature Materials 12, 114–117 (2013).
- [4] Nature Communications 8, 15204 (2017).

[Actions]:

We have made the corresponding revisions in the page 4-7 and Supplementary Figs. 9, 12.

[Comments 4]: *The time-domain photoresponse at 0.1 V shows a rise time of ~ 257 ns, which corresponds to an estimated bandwidth of ~ 1.36 MHz ($f=0.35/t$). However, the frequency-domain measurement presented in Supplementary Fig. 11 shows a bandwidth of only ~ 0.11 MHz, which is over one order of magnitude lower. Please estimate this discrepancy.*

[Response]:

We sincerely thank the reviewer for raising this critical point. Upon revisiting the experimental setup, we identified that the primary cause of this discrepancy lies in the measurement configuration used for the frequency-domain characterization. In our initial measurement, shown in Supplementary Fig. 11, the signal readout employed a transimpedance amplifier with a relatively high gain setting. This high-gain configuration, while beneficial for sensitivity, inherently limited the overall system bandwidth due to the gain-bandwidth product constraint, resulting in a suppressed -3 dB cutoff frequency of only about 0.11 MHz.

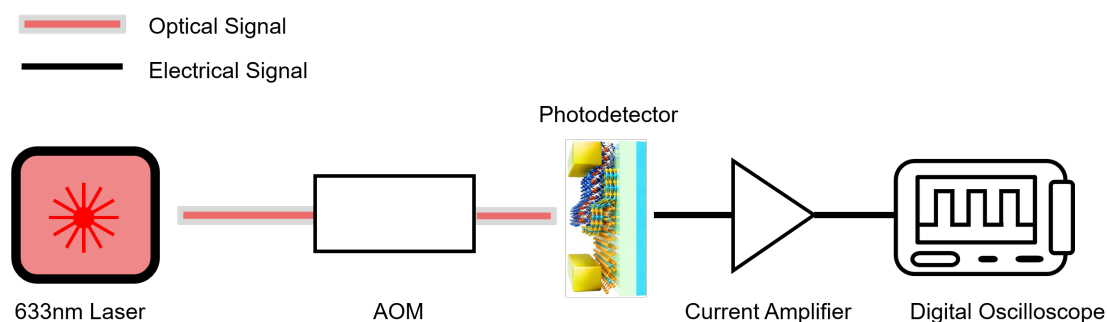


Figure R13 Experimental set-up for the high-frequency measurement.

To address this issue, we repeated the frequency-domain measurements with an optimized setup (Figure R13). Specifically, we reduced the gain of the transimpedance amplifier to extend its

effective bandwidth, while carefully maintaining an adequate signal-to-noise ratio. As shown in the newly added Figure R14, the measured -3 dB bandwidth has been significantly improved to approximately 0.5 MHz. This optimized measurement largely bridges the gap observed previously.

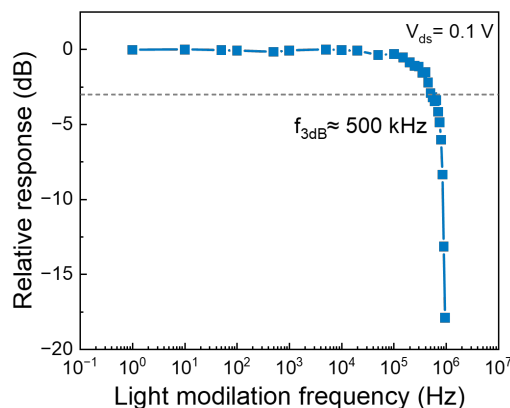


Figure R14 Normalized photoresponse as a function of modulation frequency extracted at $V_{ds} = 0.1$ V under 633 nm laser illumination for $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2/\text{PdSe}_2$ devices.

Regarding the time-domain analysis, we would also like to provide a further clarification. The previously quoted rise time of about 257 ns was derived from exponential fitting of the transient edge, which corresponds to the time constant of the exponential function. However, in the relation $f = 0.35/t$, the parameter t conventionally refers to the 10%–90% rise time. In fact, our fitting gave rise and fall time constants of ~ 257 ns and ~ 274 ns, respectively, while the corresponding 10%–90% values are 528 ns (rise) and 415 ns (fall). Substituting this value into $f = 0.35/t$ yields an estimated bandwidth of approximately 0.66 MHz, which is close to our newly measured frequency-domain cutoff of about 0.5 MHz. The remaining modest difference can be reasonably attributed to residual parasitic RC effects and nonideal device parasitics, which are not fully captured by the simplified single-pole rise-time approximation. We have updated the main text and the supplementary information accordingly.

[Actions]:

We have made the corresponding revisions in the page 9 and Supplementary Fig. 22.

[Comments 5]: *The barrier layer of the proposed unipolar photodetector is MoTe₂. However, MoTe₂ has a relative low bandgap (~ 1 eV), which means MoTe₂ is sensitive to most of the wavelengths that the authors used (e.g. 450 nm – 1320 nm). So, MoTe₂ cannot really prevent electron transport, because it can generate photocarriers itself. This further leads to the incomplete*

of the bias-related response speed mechanism, as the response speed (e.g. Fig. 3e) is measured under 633nm, which can cause the photoresponse in MoTe₂ “barrier” layer. Please evaluate contribution of photogenerated carriers in MoTe₂ and clarify how this affects the interpretation of the barrier-controlled response speed mechanism.

[Response]:

We thank the reviewer for the insightful comment. We have carefully addressed this point through additional experiments and revised discussion, as detailed below.

1. We fabricated a single-MoTe₂ device and confirmed that it shows no photoresponse at 1550 nm (Figure R15). 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 (Figure R16). This demonstrates that the tunable response speed does not primarily rely on photogeneration within the MoTe₂ layer.

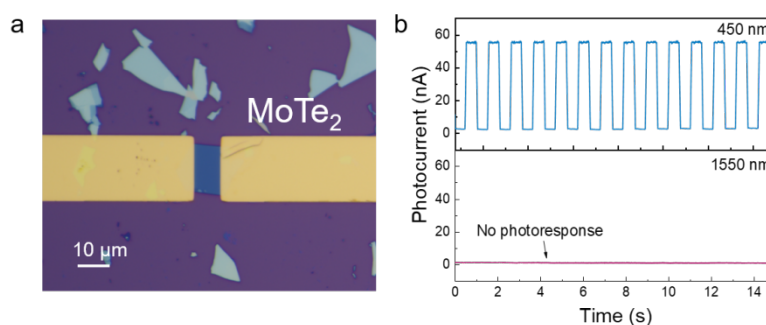


Figure R15 Photoresponse of the MoTe₂ device. (a) Optical image of the MoTe₂ device. (b) Time-resolved photocurrent measured under 1550 nm and 450 nm illumination.

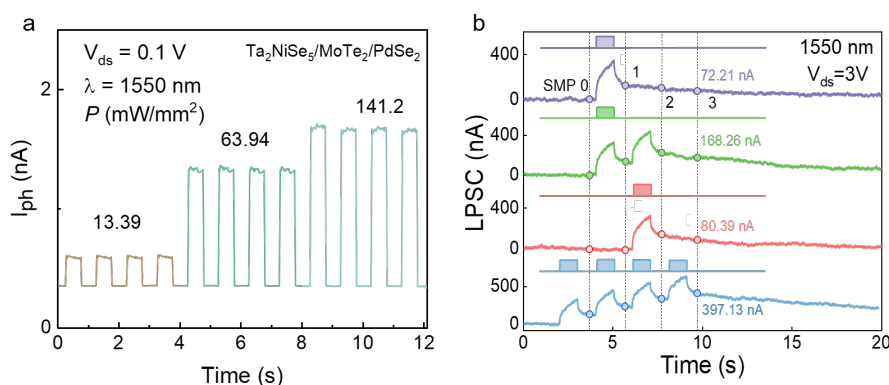


Figure R16 Photoresponse of the Ta₂NiSe₅/MoTe₂/PdSe₂ device under 1550 nm illumination. (a) Fast photoresponse at $V_{ds} = 0.1$ V. (b) Slow photoresponse at $V_{ds} = 3$ V.

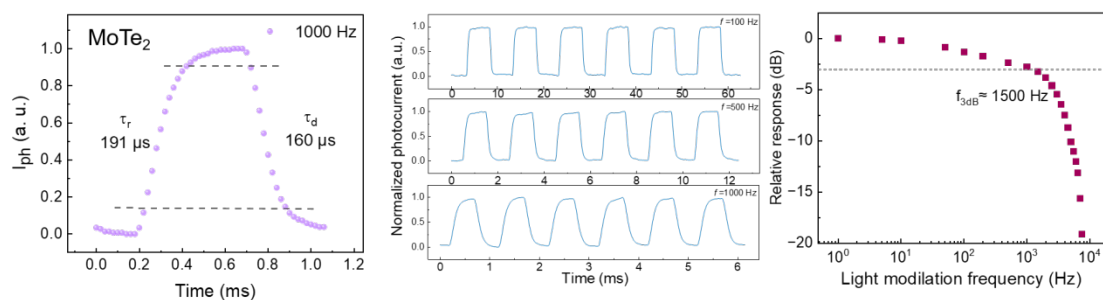


Figure R17 The Photoresponse of MoTe₂.

2. We measured the temporal response of a single 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 (Figure R17). These values are approximately three orders of magnitude slower than the 257 ns / 274 ns response of the heterostructure at low bias (0.1 V). 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.

3. We acknowledge that at photon energies above ~1 eV, MoTe₂ does absorb light and generates electron-hole pairs. However, in our barrier design, MoTe₂ serves primarily as a conduction-band blocker with a small valence-band offset. Photogenerated holes in MoTe₂ can still be rapidly extracted via the direct-tunneling pathway into PdSe₂, while electrons are blocked by the large conduction-band offset at the MoTe₂/PdSe₂ interface. This additional absorption contributes modestly to the total photocurrent but does not alter the dominant carrier-transport mechanism. The measured response time remains largely determined by the interfacial barriers that carriers need to traverse, which is consistent with our observation.

[Actions]:

We have made the corresponding revisions in the page 7 and Supplementary Materials page 10.

[Comments 6]: *The central claim is based on barrier modulation within a specific Ta₂NiSe₅/MoTe₂/PdSe₂ heterostructure. However, no experiments are provided to demonstrate the generality or reproducibility of this mechanism. For instance, it would be valuable to examine whether similar behavior persists under variations, such as different material thicknesses and alternative barrier materials.*

[Response]:

We thank the reviewer for this valuable suggestion. We fully agree that demonstrating the robustness of the observed behavior across different device configurations is essential for establishing the broad applicability of our findings.

In response, we have performed systematic experiments in two aspects.

1. First, we fabricated devices with three different thicknesses of the MoTe_2 barrier layer: 8.9 nm, 17.1 nm, and 23.3 nm. In all cases, we consistently observed the same bias-dependent transition in photoresponse characteristics: fast and reversible photodetection at low bias (0.1 V) and pronounced synaptic-like persistent photoconductivity at high bias (3 V) (Figure R18, R19). This thickness-independent behavior confirms that the mechanism is not sensitive to the exact barrier thickness.

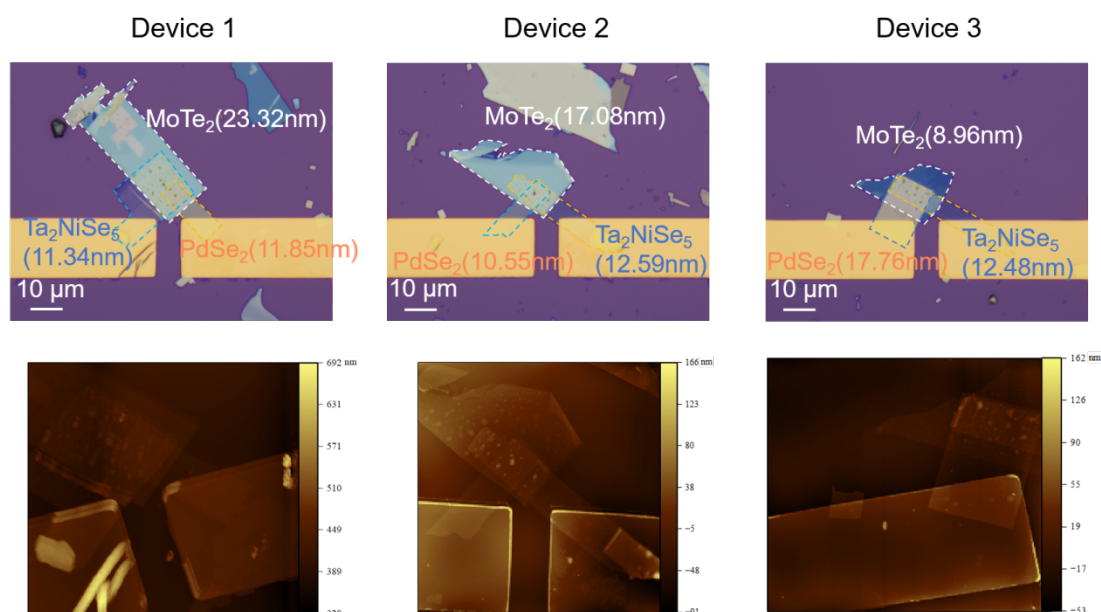


Figure R18. Optical microscopy images and corresponding AFM images of three $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2/\text{PdSe}_2$ heterostructure devices with different layer thicknesses.

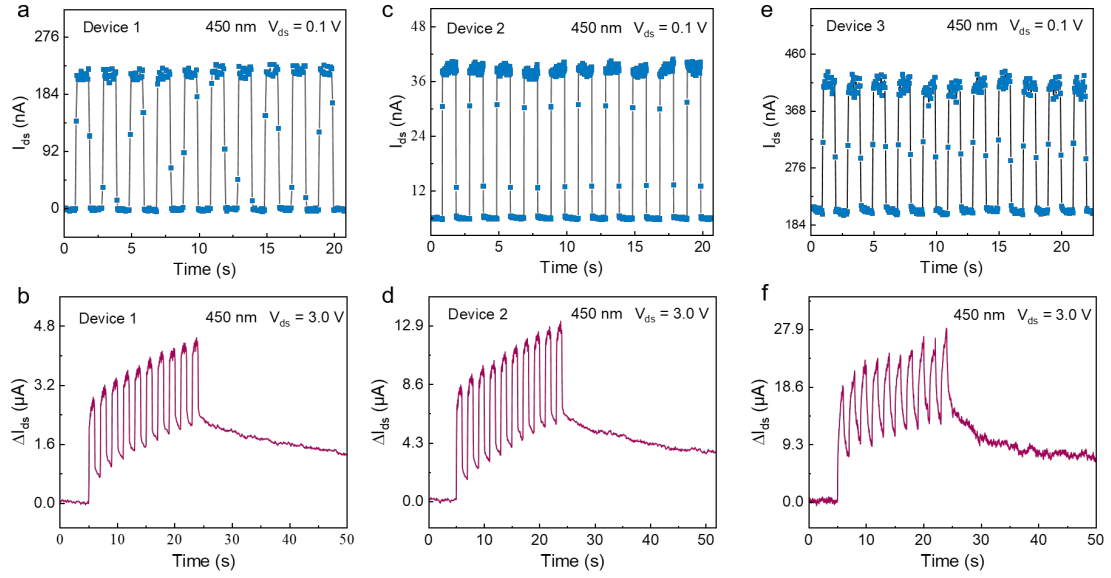


Figure R19 Photoresponse characteristics of devices with different thicknesses. (a, c, e) Low-bias time-resolved switching response under 450 nm illumination of the 3 devices, showing stable and repeatable photocurrent modulation. (b, d, f) Photocurrent response under higher bias, revealing a pronounced PPC effect with slow decay behavior.

Second, and more importantly, 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 (Figure R20). Remarkably, these heterostructures also exhibit the same bias-dependent transition: fast photoresponse under low bias and slow, persistent response under high bias (Figure R21). The observation of similar phenomena in devices with chemically distinct barrier materials, including MoTe₂, WS₂, and WSe₂, indicates that the underlying mechanism is not unique to a specific material combination but rather arises from a more general band engineering principle applicable to a broad class of van der Waals heterostructures with suitable band alignment.

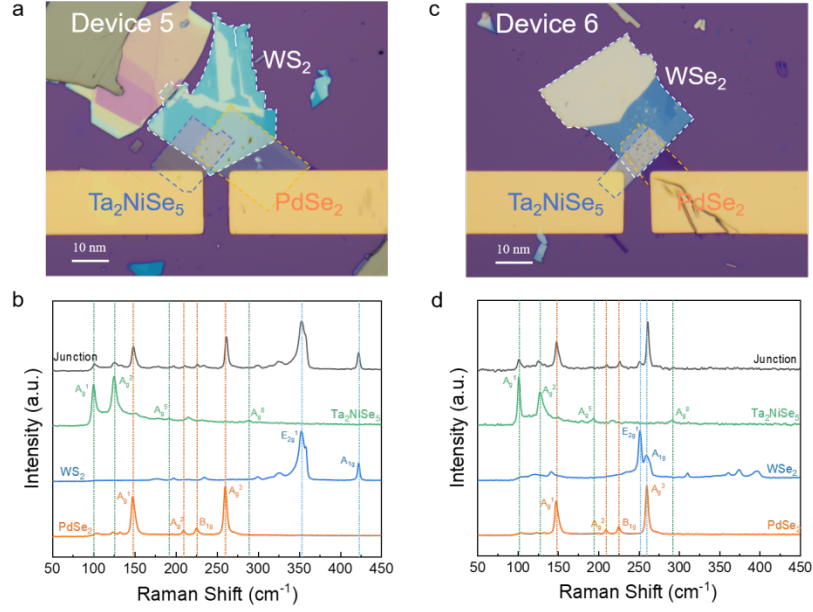


Figure R20 Optical image and Raman spectrum of the $\text{Ta}_2\text{NiSe}_5/\text{WS}_2/\text{PdSe}_2$ and $\text{Ta}_2\text{NiSe}_5/\text{WSe}_2/\text{PdSe}_2$ device.

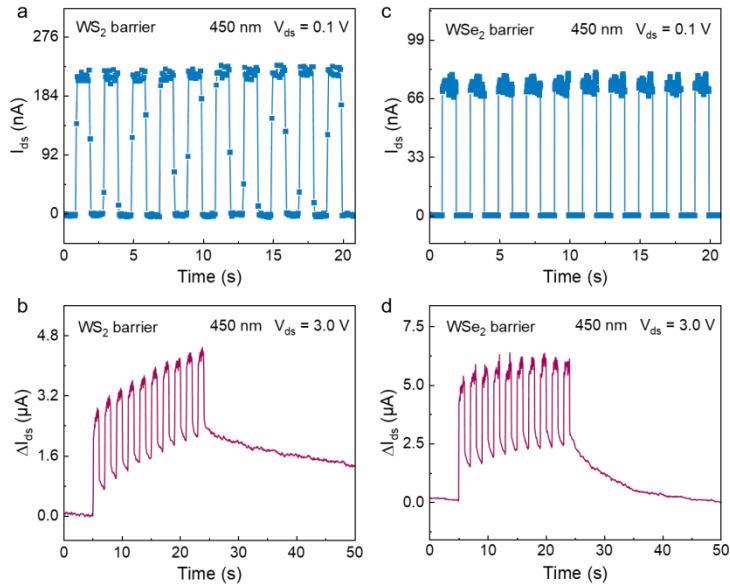


Figure R21 Photoresponse characteristics of the $\text{Ta}_2\text{NiSe}_5/\text{WS}_2/\text{PdSe}_2$ and $\text{Ta}_2\text{NiSe}_5/\text{WSe}_2/\text{PdSe}_2$ devices. (a, c) Low-bias time-resolved switching response under 450 nm illumination of the 2 devices, showing stable and repeatable photocurrent modulation. (b, d) Photocurrent response under higher bias, showing a pronounced PPC effect with slow decay behavior.

[Actions]:

We have made the corresponding revisions in the page 9 and Supplementary Fig. 13-16.

[Comments 7]: *The manuscript presents polarization-sensitive photodetection characteristics (Fig. 3d). However, this functionality seems disconnected from the core device concept and is not utilized in the demonstrated applications (Fig. 5). The authors should clarify the role and significance of polarization sensitivity in the context of the proposed system.*

[Response]:

We thank the reviewer for this constructive comment. We agree that the connection between polarization-sensitive photodetection and the core device concept could be more clearly articulated. The polarization sensitive photodetection is related to the anisotropic crystal structures of Ta₂NiSe₅ and PdSe₂, which are part of the material selection and device architecture, and it does not require external polarizers or complex metamaterial structures.

The role of polarization sensitivity in our proposed system is in three aspects.

First, it provides an additional information dimension for perception. Polarization encodes optical properties that are not accessible through conventional intensity-based imaging, such as the orientation and polarization state of light. In scenarios like autonomous driving and machine vision, where objects may be obscured by glare, scattering, or low-contrast environments, polarization information can enhance target detection and scene interpretation. By integrating this capability directly into the sensor, our device enables the extraction of richer visual information at the hardware level, which is desirable for edge intelligence where computational resources are limited.

Second, it complements the broadband and neuromorphic functionalities. The polarization sensitivity is compatible with the broadband operation and the bias-reconfigurable fast/slow response dynamics of the device, meaning that all these functionalities coexist within the same hardware without mutual interference. This compatibility ensures that polarization information can be simultaneously acquired with spectral and temporal information in a single detector.

Third, it offers future integration potential. While the motion recognition demonstration in Fig. 5 focuses on temporal dynamics and trajectory prediction and does not explicitly utilize polarization information, the intrinsic polarization capability remains available for future extensions of the system to more complex perception tasks, such as distinguishing objects under low visibility conditions or improving contrast in scenes with strong background interference. Recent studies have demonstrated the potential of polarization-sensitive neuromorphic devices for polarization-resolved

imaging and feature extraction, further supporting the value of integrating polarization sensitivity into neuromorphic vision hardware.^[1, 2] The unique combination of broadband operation, ultrawide dynamic range, and intrinsic polarization sensitivity in our device makes it promising for future multidimensional visual perception systems.

[1] Lin, Y. et al. Antiferroelectric polarization enabling physical activation in CuBiP2Se6 for medical image processing. *Nature Communications* 2026.

[2] Li, C. et al. Coupled polarization dynamics and charge tunneling enable reconfigurable heterojunctions. *Nature Communications* 2026.

[Actions]:

We have made the corresponding revisions in the page 9.

[Comments 8]: *The author claims broadband photoresponse from the visible (450nm) to mid-infrared (3.8um). However, the presented photodetector performance data (e.g., I-V characteristics, responsivity, and response time) are only experimentally demonstrated up to approximately 3.1um. The evidence at 3.8um appears to be limited to synaptic (EPSC) measurements, without corresponding photodetector performance. Therefore, the claim of operation up to 3.8um is not sufficiently supported by the current data.*

[Response]:

We thank the Reviewer for this careful observation. The limited signal at 3.8 μm in the earlier measurements arose because this wavelength lies near the absorption edge of Ta₂NiSe₅, resulting in a relatively weak photoresponse under the low-bias conditions used for photodetector characterization. In our revised experiments, we have optimized the measurement conditions and now obtained clear photodetector performance data at 3.8 μm . These new data confirm that the device does function as a photodetector at this wavelength.

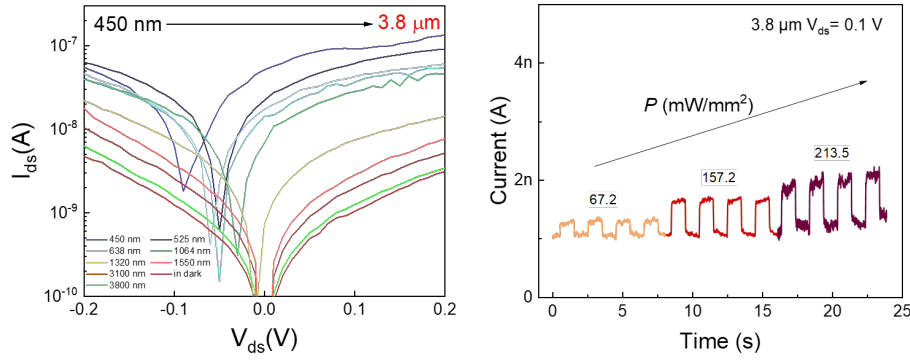


Figure R22 (a) Logarithmic I_{ds} - V_{ds} curves measured in the dark and under illumination at wavelengths ranging from 450 nm to 3800 nm. (b) Photoresponse characteristics of devices under $3.8 \mu m$ illumination.

[Actions]:

We have made the corresponding revisions in Fig. 3a, c.

[Comments 9]: *The simulation task remains unclear how such functionality can be practically realized at the hardware level. In particular, the manuscript appears to rely on a single device. Could the authors clarify how this approach enables scalable on-chip integration? How would multiple devices be interconnected to support real-time processing, and what are the limitations for large-scale system implementation?*

[Response]:

We thank the reviewer for this constructive comment. We have fabricated a 3×3 device array and experimentally validated its performance (Figure R23). In this array, the $PdSe_2$ and $MoTe_2$ layers were grown by chemical vapor deposition, while the Ta_2NiSe_5 flakes were prepared by mechanical exfoliation, as the CVD growth of Ta_2NiSe_5 remains challenging at present. All devices exhibit consistent bias-tunable behavior, showing fast photoresponse under low bias and slow response with persistent photoconductivity under high bias, which demonstrates the feasibility of extending our single-device results to a multi-pixel architecture. Regarding how multiple devices could be interconnected for real-time processing, we envision an architecture in which each pixel in the array is individually addressable and its bias voltage can be dynamically switched according to the task requirements. In the collaborative fusion framework, the same pixel can first operate in the fast photodetector mode to capture instantaneous spatial information, and then switch to the slow photomemristor mode to accumulate temporal signals and generate motion dependent features. This

time division multiplexing strategy leverages the core advantage of our device, its bias tunable response time, allowing each pixel to flexibly alternate between fast sensing and slow memory functions, rather than assigning different functions to different devices. The outputs from the two modes can be combined through a simple readout circuit to implement the fusion algorithm. For more advanced neuromorphic processing, the array can also be configured as a crossbar architecture, where each device serves as a tunable synaptic weight. The bias reconfigurable response time naturally provides a mechanism for encoding temporal information at the hardware level, which can be exploited for reservoir computing or spiking neural networks.

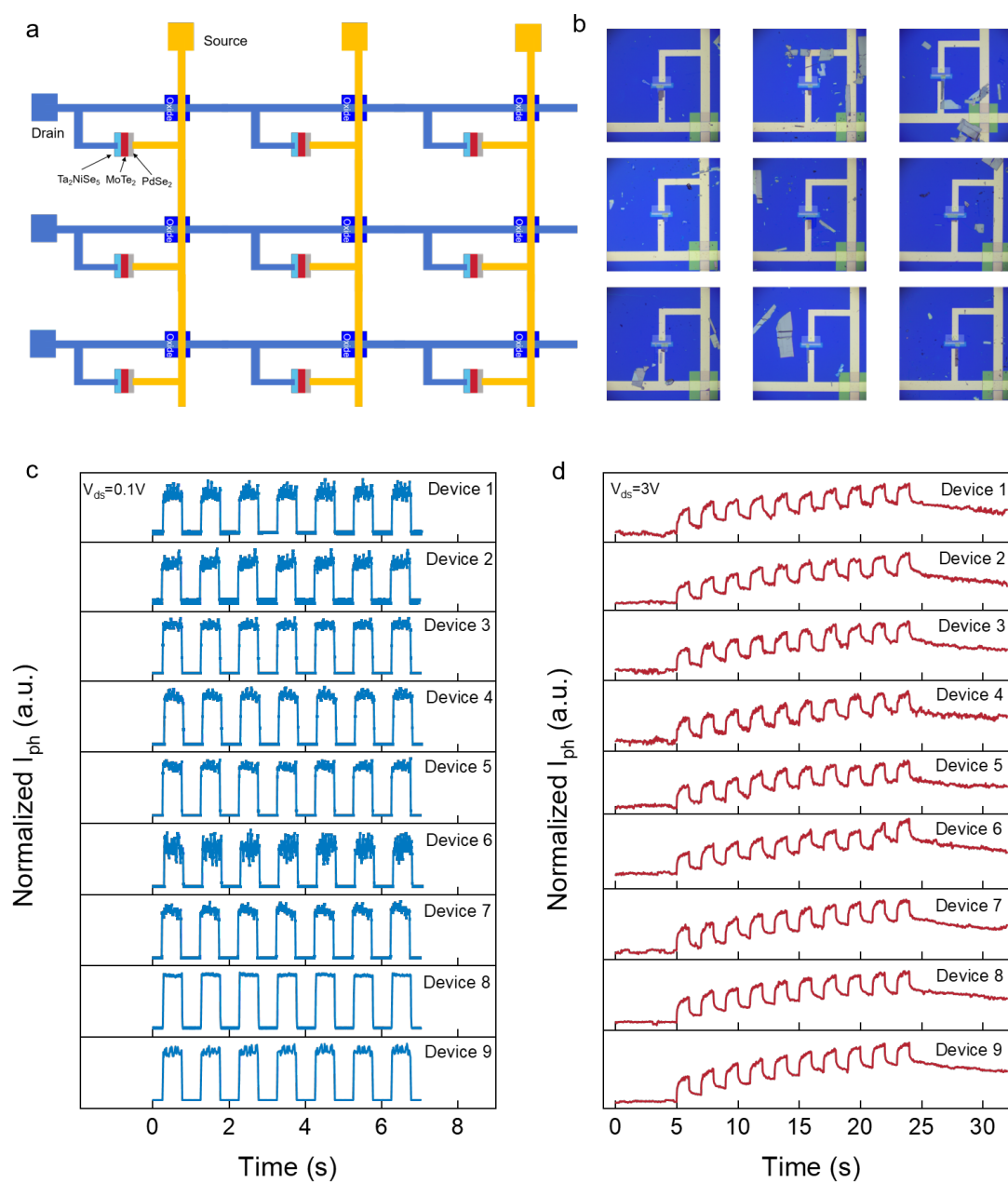


Figure R23 (a) Schematic illustration of the 3×3 device array based on the $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2/\text{PdSe}_2$ heterostructure. (b) Optical image of the 3×3 device array. (c) Photoresponse of the 3×3 device array under low and high bias, showing consistent bias-tunable behavior across all nine devices.

[Actions]:

We have made the corresponding revisions in the page 7, 13 and Supplementary Fig. 17.

[Comments 10]: *In Supplementary Fig. 12, the transition from short-term memory (STM) to long-term memory (LTM) is demonstrated. However, the analysis remains largely qualitative. Quantitative metrics such as retention time constants and decay rates are not provided. Could the authors include a more rigorous and quantitative characterization of the synaptic dynamics to better substantiate the claimed memory behavior?*

[Response]:

We thank the reviewer for this constructive comment. To provide a more quantitative characterization of the synaptic dynamics and the transition from STM to LTM, we performed control experiments by measuring the EPSC decay under various light power densities, illumination durations, and pulse numbers (Figure R24, R25, R26).

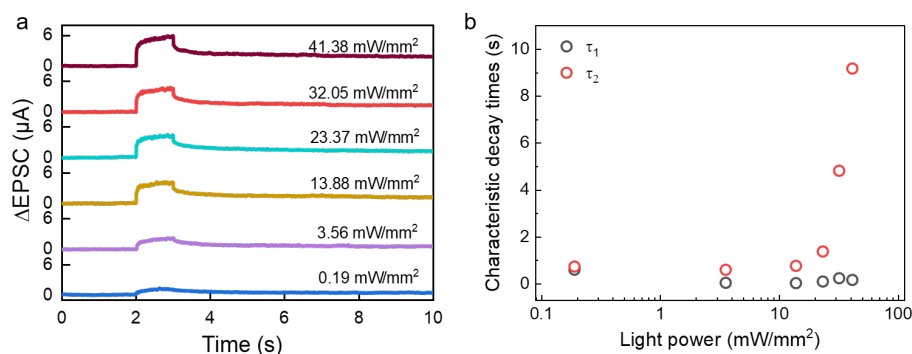


Figure R24 Light-intensity-dependent optoelectronic synaptic response and decay dynamics analysis. (a) Time-resolved EPSC responses under different light power densities. (b) Extracted characteristic decay time constants at different light intensities obtained from bi-exponential fitting.

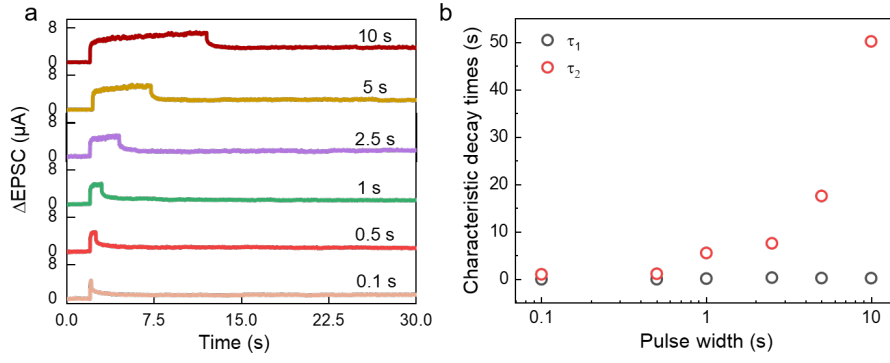


Figure R25 Illumination-duration-dependent decay dynamics. (a) Time-resolved EPSC responses under different illumination durations. (b) Extracted characteristic decay time constants at illumination durations.

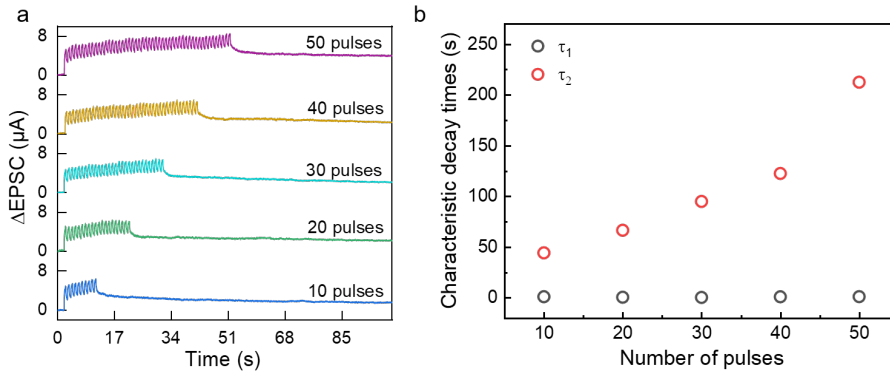


Figure R26 Illumination-duration-dependent decay dynamics. (a) Time-resolved EPSC responses under different illumination durations. (b) Extracted characteristic decay time constants at illumination durations.

The decay curves were fitted using a bi-exponential function:

$$I(t) = C_1 \exp\left(-\frac{t}{\tau_1}\right) + C_2 \exp\left(-\frac{t}{\tau_2}\right)$$

where τ_1 and τ_2 are the fast and slow decay time constants, respectively. The slow component τ_2 is directly related to the retention behavior of the synaptic device. We extracted these parameters systematically for all measurement conditions. As reported by Kah-Wee Ang et. al (Adv. Mater. 2024, 36, 2307393), STM is defined as temporary storage with a retention time of less than 60 s, whereas LTM corresponds to retention longer than 60 s. Our quantitative results show that when the pulse number is below 20s, τ_2 remains smaller than 60 s, indicating STM behavior. In contrast, when

the pulse number reaches 20 or more, τ_2 exceeds 60 s and continues to increase with more pulses which clearly transit into the LTM regime.

[Actions]:

We have made the corresponding revisions in the page 10 and Supplementary Materials page 27-29.

Reviewer #3:

The manuscript reports a van der Waals heterostructure photodetector based on Ta₂NiSe₅/MoTe₂/PdSe₂, designed to enable bias-controlled modulation of carrier transport across an interfacial barrier. By tuning the applied bias, the authors claim a continuous transition from direct tunneling to thermionic emission, resulting in a modulation of the photoresponse time over seven orders of magnitude (from ~600 ns to >8 s). This allows the device to operate either as a high-speed photodetector or as an optoelectronic synapse. The device further exhibits broadband photodetection (450 nm to ~3–3.8 μ m), polarization sensitivity, and is demonstrated in neuromorphic tasks such as reservoir computing and motion recognition, achieving reported accuracies above 96%. The work aims to provide a unified hardware platform for integrated sensing and neuromorphic vision. Overall, this work contains a wealth of experiments, treats a timely topic, and exhibits some performance parameters that seem to go beyond the state-of-the-art. However, the central claims — particularly the continuous seven-orders-of-magnitude modulation mechanism and its interpretation in terms of transport physics — are not sufficiently substantiated. As such, it may be of interest to the broad readership of Nature Communications, but this would require substantial additional analysis and clarification. This manuscript could become acceptable for publication in Nature Communications after major revisions.

[Response]:

We sincerely thank the reviewer for the positive comments on our manuscript. We have carefully addressed the concerns raised by the reviewer, as shown below.

[Comments 1]: *The central claim of continuous response-time modulation over seven orders of magnitude is attributed to a transition between direct tunneling and thermionic emission. However, the evidence provided is indirect and largely qualitative. The tunneling vs. thermal emission regimes*

are inferred from I - V characteristics and Arrhenius fits, but no direct spectroscopic or time-resolved carrier dynamics measurements are presented to validate this transition.

[Response]:

We sincerely thank the reviewer for raising this important point. To directly verify this mechanism transition at the microscopic carrier dynamics level, we performed pump-probe ultrafast spectroscopy on our devices. Figure R1 shows the transient reflectivity change ($\Delta R/R$) measured by pump-probe spectroscopy under two bias conditions.

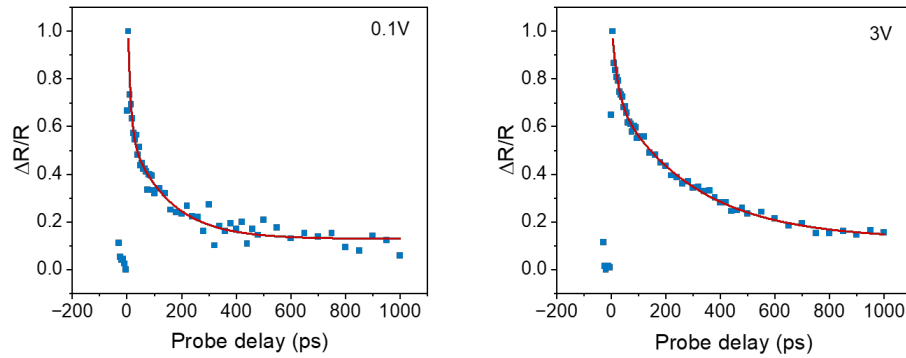


Figure R1. Pump-probe measurements of the device under low bias and high bias.

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

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

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.

[Actions]:

We have made the corresponding revisions in the page 7 and Supplementary Fig. 11.

[Comments 2]: *To this point, it is puzzling that the extracted barrier heights (~ 0.18 – 0.21 eV) show only modest variation, which does not obviously explain such a dramatic change in response time. A quantitative model linking barrier modulation to time constants is missing.*

[Response]:

We sincerely thank the reviewer for this insightful and critical comment. Through systematic first-principles calculations, temperature-dependent transport measurements, and multi-mechanism quantitative modeling, we provide the following analysis:

1. Band structure foundation established by first-principles calculations

We first employed first-principles calculations to study the electronic structure of Ta₂NiSe₅, MoTe₂, PdSe₂, and their heterojunctions (Figure R2).

The band edge positions of the individual materials are approximately as follows:

Ta₂NiSe₅: VBM ≈ -0.18 eV, CBM ≈ 0.17 eV, bandgap ≈ 0.35 eV;

MoTe₂: VBM ≈ -0.53 eV, CBM ≈ 0.56 eV, bandgap ≈ 1.09 eV;

PdSe₂: VBM ≈ -0.01 eV, CBM ≈ 0.41 eV, bandgap ≈ 0.42 eV.

After the formation of the Ta₂NiSe₅/MoTe₂ heterojunction, a conduction band offset of approximately 0.74 eV is created. This conduction band offset may play a dual role:

(1) First, it largely separates the transport paths of electrons and holes, such that the response time is primarily governed by the hole transport mechanism. Photogenerated electrons are confined to the Ta₂NiSe₅ side and cannot easily overcome the conduction band offset to enter MoTe₂. Therefore, recombination relies mainly on holes crossing the interface, making the photoresponse speed sensitive to the hole transport mechanism. When holes cross by the direct tunneling, the device exhibits a fast response; when they need to overcome the barrier by slow thermal excitation, the response time extends to the slow regime. This conduction band offset thus enables the hole transport mechanism to largely influence the overall response time with less interference from

electron transport.

(2) Second, it provides a favorable condition for the appearance of PPC and long-term memory effects. Under high bias, when the DT channel for holes is suppressed, photogenerated holes tend to remain on the Ta_2NiSe_5 side and become spatially separated from the blocked electrons, significantly reducing recombination and giving rise to PPC.

(3) Simultaneously, the VBM of Ta_2NiSe_5 (-0.18 eV) is higher than that of MoTe_2 (-0.53 eV), yielding a valence band offset of about 0.35 eV. This provides a bias-tunable interfacial barrier for holes. As the external electric field increases, the energy levels on both sides shift upward collectively, with the valence band edge dominated by the Ni–Se orbitals of Ta_2NiSe_5 and with the Te/Mo states of MoTe_2 participating in the interfacial coupling, leading to noticeable changes in the width and effective height of the valence band barrier.

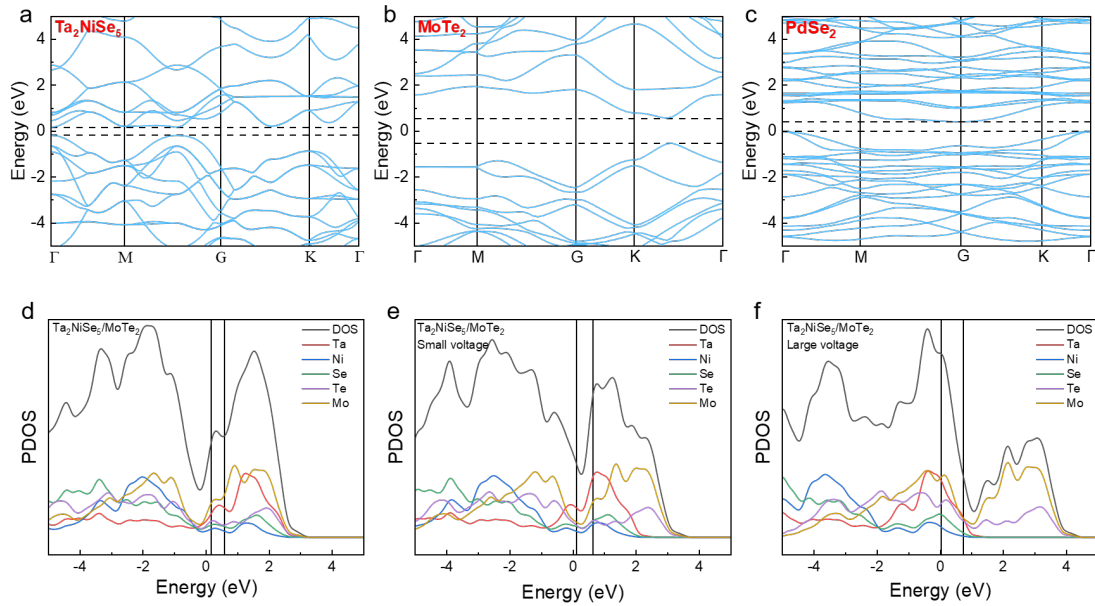


Figure R2 First-principles calculations of the electronic structure. (a–c) Band structures of the individual Ta_2NiSe_5 , MoTe_2 , and PdSe_2 , with the valence band maximum and conduction band minimum indicated. (d–f) Density of states of the $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2$ heterostructure 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.

2. Experimental identification of the two transport modes

At $V_{\text{ds}} = 0.1$ V, the photocurrent is nearly independent of temperature in the range of 80–300

K (Figure R3). This is consistent with tunneling-dominated transport. ^[1]

At $V_{ds} = 3$ V, the photocurrent exhibits a strong temperature dependence (Figure 1b), following the thermal excitation relation:

$$J = AT^2 \exp\left(-\frac{\phi}{k_B T}\right)$$

The extracted activation energy is consistent with the barrier height. This supports that carriers overcome the barrier mainly by thermal excitation.

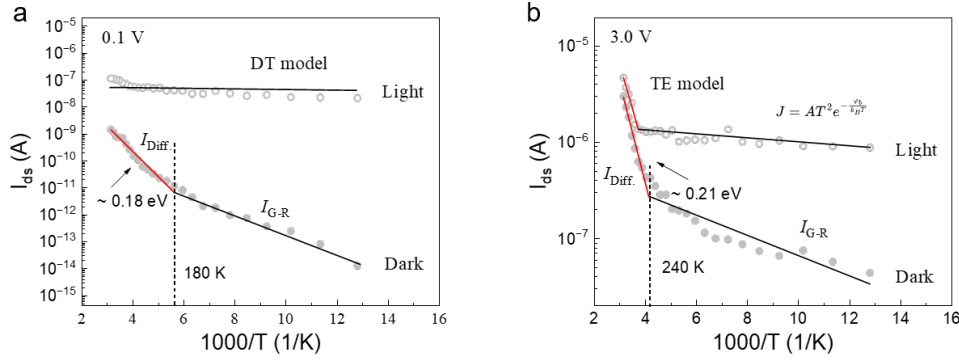


Figure R3 Temperature-dependent optoelectronic measurements of $V_{ds} = 0.1$ V and $V_{ds} = 3$ V.

Furthermore, we measured the bias-dependent photocurrent at both low temperature (80 K) and room temperature (300 K) (Figure R4). At low bias, the photoresponse is fast at both temperatures, consistent with direct tunneling that is insensitive to thermal activation. At high bias, the photoresponse at 80 K remains fast, while at 300 K the same bias produces obviously slow decay with PPC effect. This obvious temperature contrast at high bias indicates that the slow response is thermal excitation effect, largely suppressed at low temperature and becoming dominant at room temperature.

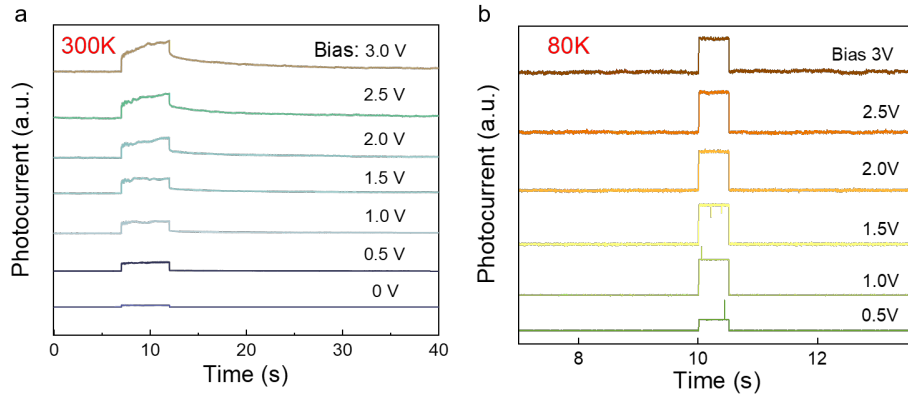


Figure R4 Bias-dependent photocurrent measured at 80 K and 300 K.

To further identify the transition between these two regimes, we analyzed the bias-dependent photocurrent characteristics. By plotting $\ln(I/V^2)$ versus $1/V$, two distinct regions emerge: at low bias, the curve exhibits behavior characteristic of direct tunneling; as the bias increases to approximately $V_{ds} \approx 0.5$ V, a clear change in the slope is observed, suggesting that the DT channel is gradually suppressed, and thermal excitation begins to take over (Figure R5). The experimental data can be described by the DT current expression:

$$I_{DT} = V \exp\left(-\frac{4\pi\sqrt{2m^*}\phi}{h}\right)$$

This transition point coincides roughly with the bias at which the response time jumps from the sub-microsecond to the millisecond-second regime in our experiments.

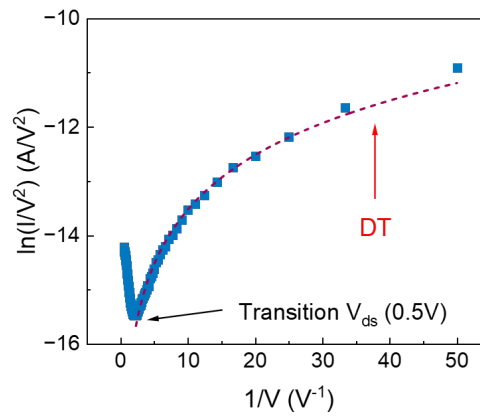


Figure R5 Fowler–Nordheim plots of the device at positive V_{ds} under illumination.

Additionally, we have performed systematic temperature-dependent I–V measurements in the dark at various bias voltages and extracted the effective barrier height $\phi(V)$ via Arrhenius fitting (Figure R6). ϕ exhibits a non-monotonic V-shaped variation: it decreases from 0.18 eV at 0.1 V to a minimum of ~ 0.10 eV around 0.5 V, and then monotonically rises to 0.29 eV at 3 V. The physical origin of this behavior can be understood as follows: at low bias, the external electric field partially counteracts the built-in field, narrowing the depletion region and making the barrier thinner and lower; once the bias exceeds a threshold of ~ 0.5 V, the depletion region may extend through the entire MoTe_2 layer and the barrier height rises again. This non-monotonic behavior provides rather direct experimental evidence for the abrupt suppression of the DT channel near 0.5 V.

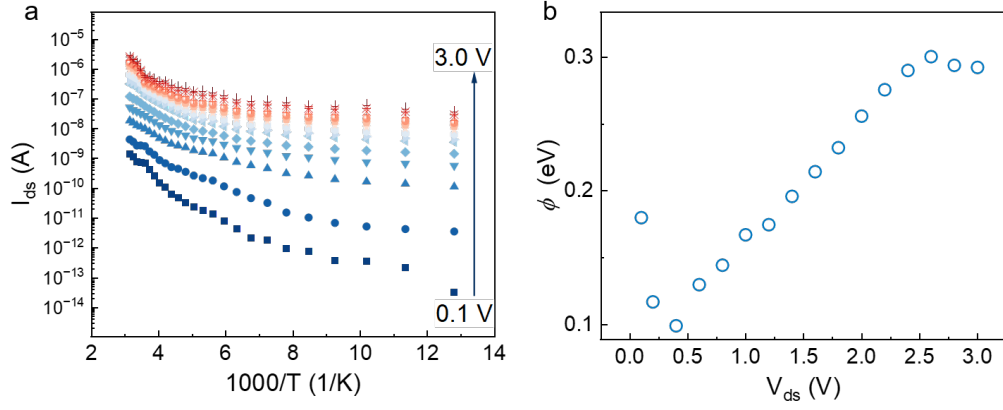


Figure R6. Temperature-dependent I–V measurements in the dark at various bias voltages and the extracted effective barrier height

The spatial distribution of photocurrent provides visual evidence for the effect of bias voltage on the depletion region (Figure R7). Compared with the 0V, 0.1 V condition, at 3 V the primary photocurrent generation region shifts significantly toward the edge of the $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2$ interface, indicating an expansion of the depletion region.

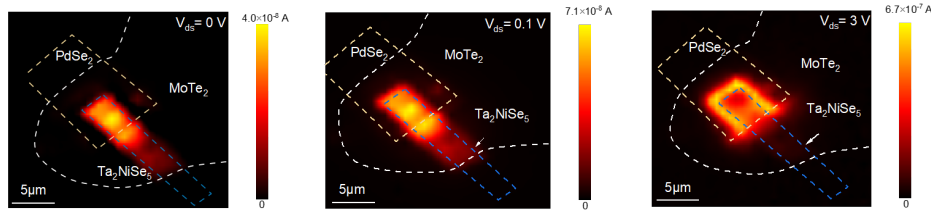


Figure R7 The spatially resolved photocurrent mapping images at $V_{ds} = 0$ V, $V_{ds} = 0.1$ V and $V_{ds} = 3$ V, respectively.

3. Transport model

Based on the above analysis, 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:

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

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:

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

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

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

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:

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

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.

To describe the competition between DT and TE mechanisms, a voltage-dependent weighting function $f(V)$ is introduced. This physical saturation behavior can be well described by a sigmoidal switching function, a form commonly used in semiconductor device modeling to describe smooth transitions between two limiting states:^[2-4]

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

Here, $f(V)$ represents the probability that charge transport occurs via the thermal excitation channel, while $1 - f(V)$ corresponds to the tunneling contribution. This formulation ensures a smooth and continuous transition between the two limiting transport regimes.

Physically, the sigmoidal form of $f(V)$ can be understood as a Fermi-like distribution arising from the competition between two transport pathways with different effective activation energies. The transition voltage (V_{th}) corresponds to the point at which the contributions of DT and TE are comparable, while the parameter (γ) characterizes the sharpness of the transition and reflects the degree of energetic disorder or interface inhomogeneity in the device.

By substituting the experimentally determined parameters into the above equation, we calculated the theoretical response time over the full bias range. The resulting curve (Figure R8) reproduces the key features observed experimentally: a fast response at low bias, a sharp increase around 0.5 V, and a slow decay at high bias. The theoretical curve is in reasonably good agreement with our experimental data, which supports the validity of the proposed model and indicates that the seven-order-of-magnitude modulation of the response time originates primarily from the bias-driven DT-to-TE mechanism transition, with the trap states providing an additional contribution to the slow response at high bias.

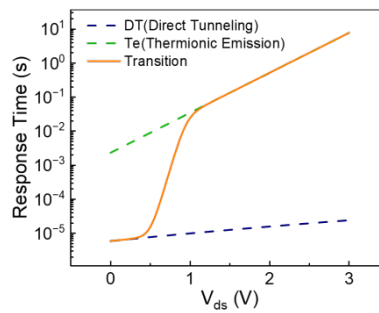


Figure R8. Calculated bias-dependent response time from the model, showing agreement with experimental data

[1] Nature Communications 15, 141 (2024)

[2] Nature 453, 80–83 (2008).

[3] Nature Materials 12, 114–117 (2013).

[4] Nature Communications 8, 15204 (2017).

[Actions]:

We have made the corresponding revisions in the page 4-7 and Supplementary Fig. 7-9, 12.

[Comments 3]: *Furthermore, I would like to see a more rigorous demonstration of the highly bias-dependent response times shown in Fig. 1b. In principle, this has been done well in Fig. 3e, however, only for the isolated case of $V = 0.1$ V and a repetition rate of 20 kHz. The choice of the repetition rate for this bias is sufficiently justified by Fig. S11b. I would like to see the corresponding analyses for three more voltages up to $V = 3.0$ V, including the power spectra to justify the applied repetition rates (which, I assume, would have to be smaller for larger voltages).*

[Response]:

We thank the reviewer for this valuable observation. We have performed additional frequency response measurements at $V_{ds} = 0.6$ V, 0.7 V, and 0.8 V, and the corresponding -3 dB cutoff frequencies are provided (Figure R9). As the bias increases from 0.1 V to 0.8 V, the -3 dB cutoff frequency decreases from approximately 500 kHz to approximately 3.5 kHz, consistent with the trend of increasing response time shown in Fig. 1b. The repetition rates used at each bias were chosen based on the respective frequency response spectra. For bias voltages beyond approximately 0.8 V, the device response moves into the millisecond-to-second regime, where the device transitions into a regime dominated by PPC. Under these conditions, the photocurrent no longer follows the modulated optical input in a simple linear manner, and a considerable portion of the response is governed by barrier-limited transport and slow trap-assisted carrier release. As a result, the -3 dB bandwidth, which is most meaningful for linear time-invariant systems, becomes less representative of the underlying device dynamics. For the high-bias synaptic regime, the behavior is more appropriately characterized in the time domain through metrics such as the EPSC decay time constant, PPF index, and long-term retention time, as presented in the main manuscript.

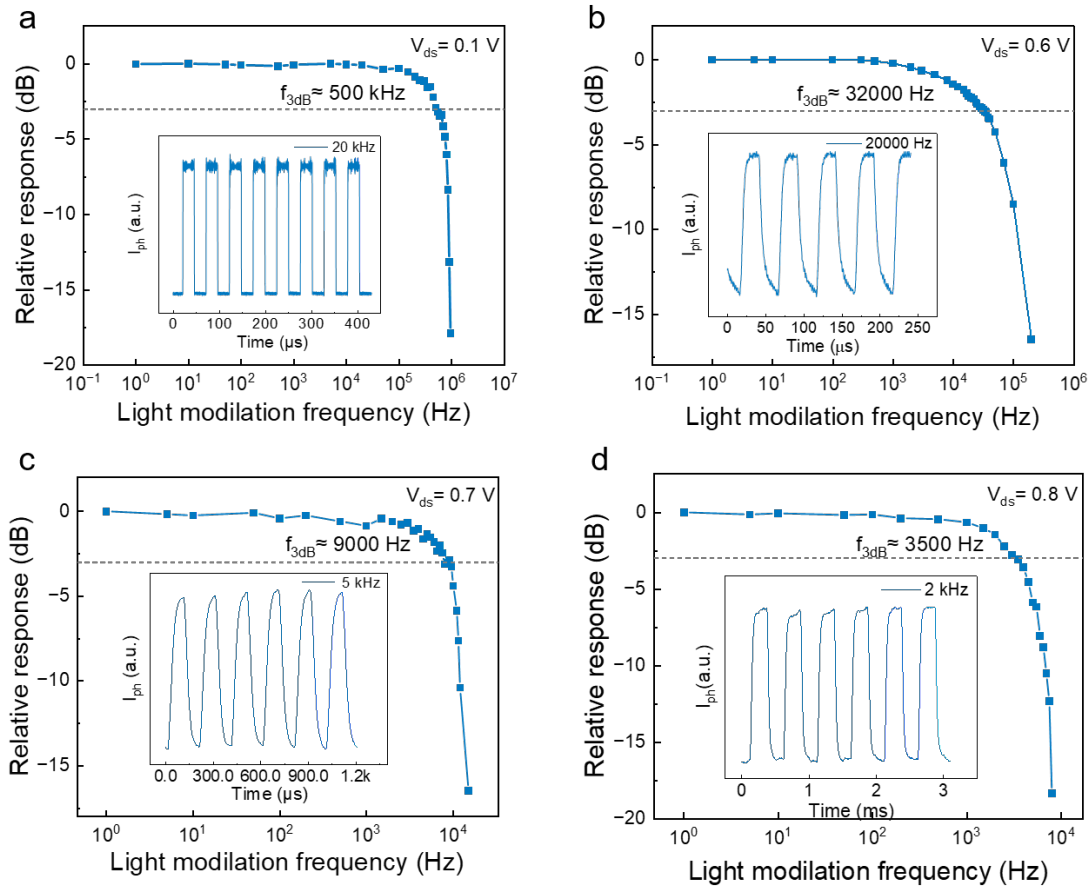


Figure R9. Relative response with the modulation frequency measured for the heterostructure under different bias.

[Actions]:

We have made the corresponding revisions in page 9 and Supplementary Fig. 22.

[Comments 4]: In Figure 3f, the authors claim a response time of ~ 300 ns (justified by Fig. 3e) in combination with a responsivity of 2 A/W. However, this responsivity was only shown for the lowest power density in Fig. 7a. At which power density was the response time obtained? Was this really the same (very low) power density? This would surprise me. I would like to see the unnormalized plot of Fig. 3e to see the absolute current values.

[Response]:

We thank the reviewer for this valuable observation. We apologize for not clearly stating the measurement conditions in the original manuscript. We would like to clarify this point and provide a detailed explanation, together with the requested unnormalized voltage trace and a schematic of

the speed-measurement setup.

First, the responsivity measurement was performed under continuous-wave (CW) illumination at a relatively low power density of 0.0014 mW/mm^2 . At such a low power density, the signal is weak. Even with a transimpedance amplifier, a very high gain (e.g., 10^8 V/A) would be required to obtain a measurable signal, but this would severely limit the amplifier bandwidth and thus prevent the oscilloscope from capturing the transient response. In order to preserve the temporal dynamics, we chose a higher power density of $\sim 50 \text{ mW/mm}^2$, the generated photocurrent allowing us to record the pulse shape with reasonable fidelity. The experimental setup is shown in Figure R10: a continuous-wave laser is modulated by an acousto-optic modulator (AOM) at a repetition frequency of 20 kHz, and the device output is fed into a current amplifier (DHPCA-100); the amplified voltage is directly recorded by a digital oscilloscope. At the higher power density used for the speed measurement, the responsivity was $\sim 0.1 \text{ A/W}$, which may be due to slight saturation.

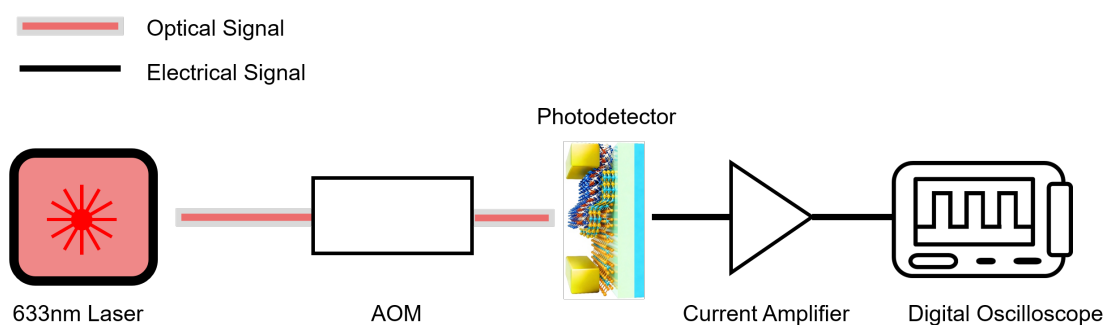


Figure R10. Experimental set-up for the high-frequency measurement.

Second, we provide the raw voltage-time trace directly captured by the oscilloscope, without any normalization as Figure R11.

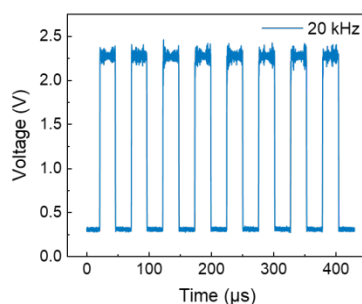


Figure R11. The original voltage-time trajectory of the photodetector captured directly by the oscilloscope.

[Actions]:

We have made the corresponding revisions in page 9 and Supplementary Fig. 22.

[Comments 5]: *The long response times at high bias are attributed to trapped carriers and barrierlimited transport. However, PPC is a common phenomenon in 2D materials and heterostructures and is often dominated by defect states or interface traps. The manuscript does not convincingly distinguish between intrinsic barrier effects and extrinsic trap-assisted processes. Control experiments (e.g., temperature-dependent decay dynamics, illumination history dependence) would be necessary.*

[Response]:

We thank the reviewer for this insightful comment. To better distinguish between trap-assisted processes and barrier-limited transport, we performed a series of control experiments examining the decay dynamics under varying light power density, illumination duration, and pulse number (Figure R12, R13, R14). The results show distinct trends under different excitation conditions, which may help to differentiate the two mechanisms.

First, Figure R10a shows the excitatory postsynaptic current (EPSC) as a function of time under various light power densities. The transient decay curves were fitted using a bi-exponential function:

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

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

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

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.

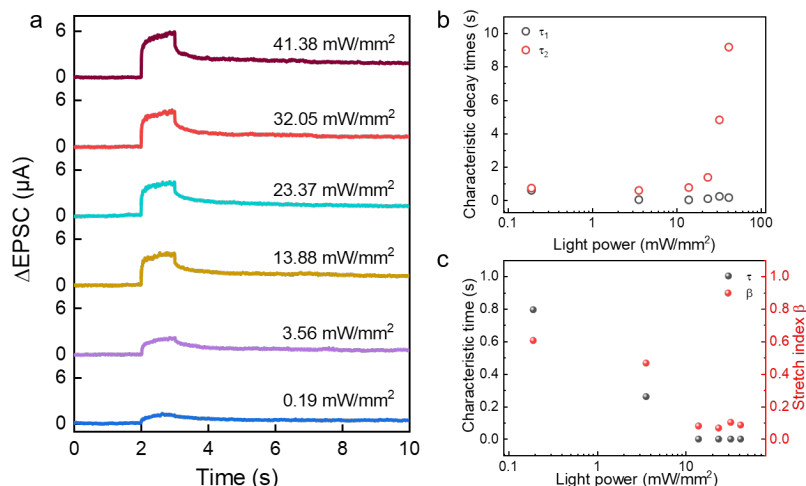


Figure R12 Light-intensity-dependent optoelectronic synaptic response and decay dynamics analysis. (a) Time-resolved EPSC responses under different light power densities. (b) Extracted characteristic decay time constants at different light intensities obtained from bi-exponential fitting. (c) Light-intensity dependence of the characteristic relaxation time (τ) and stretching exponent (β) extracted from the stretched exponential fitting.

In contrast, when we increase the illumination duration (Figure R11) or pulse number (Figure R12), 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 .

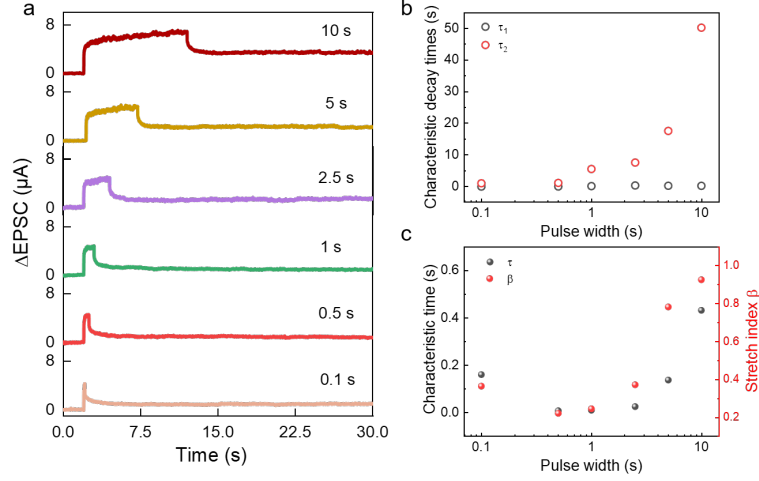


Figure R13 Illumination-duration-dependent decay dynamics. (a) Time-resolved EPSC responses under different illumination durations. (b) Extracted characteristic decay time constants at illumination durations. (c) Illumination duration dependence of the τ and β .

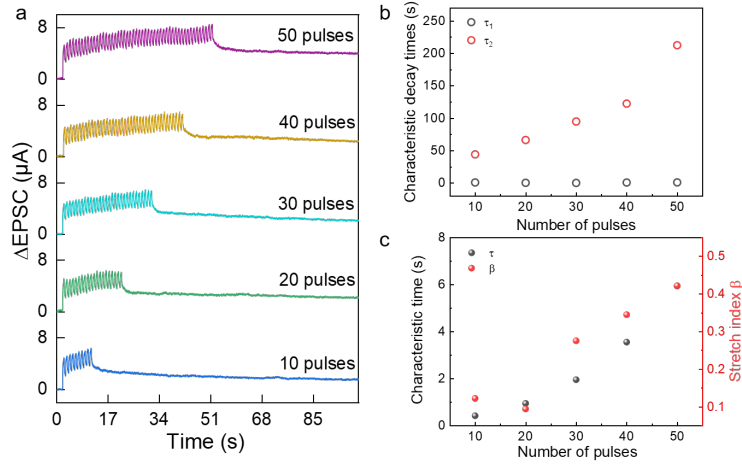


Figure R14 Illumination-duration-dependent decay dynamics. (a) Time-resolved EPSC responses under different illumination durations. (b) Extracted characteristic decay time constants at illumination durations. (c) Illumination durations dependence of the τ and β .

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.

[1] *Adv. Mater.* **37**, e2500049, 2025

[Actions]:

We have made the corresponding revisions in the page 10 and Supplementary Materials page 27-29.

[Comments 6]: *It is unclear what the color map in Figure 1a refers to.*

[Response]:

We thank the reviewer for the helpful comment. In the original figure, the color scheme was intended to distinguish different materials in the schematic; however, we agree that the correspondence between the colors and the device structure was not sufficiently clear.

In the revised manuscript, we have removed the ambiguous color mapping and instead directly labeled each material in the schematic using arrows and annotations. This modification improves the clarity and readability of the figure.

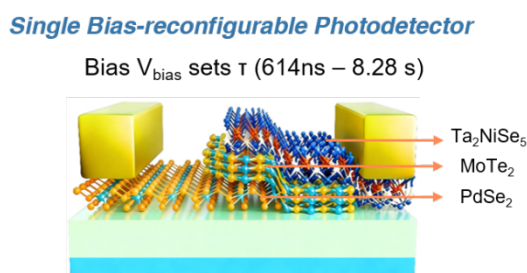


Figure R15 Schematic diagram of the broadband bias tunable reconfigurable optical response

[Comments 7]: *The y-axis label in Figure 5d should read “true”*

[Response]:

We thank the reviewer for carefully identifying this typographical error. The y-axis label in Fig. 5d has been corrected from “ture” to “true” in the revised manuscript.

[Comments 8]: *What is the rational behind the wavelength-dependent polarization dependence shown in Figure S9?*

[Response]:

We thank the reviewer for raising this important question. The wavelength-dependent

polarization ratio shown in Figure S9 is a result of two intertwined physical factors: the wavelength-dependent spatial distribution of absorbed light among the constituent layers and the intrinsic photon-energy dependence of the transition dipole matrix elements in each anisotropic material.

We note that similar wavelength-dependent polarization sensitivity has been observed in many other 2D materials and heterostructures. For instance, $(\text{TaSe}_4)_2\text{I}/\text{MoTe}_2$ exhibits a dichroism reversal from visible to infrared; $\text{MoS}_2/\text{Ta}_2\text{NiSe}_5$ shows a rotation of the preferred polarization direction; $\text{Nb}_2\text{GeTe}_4/\text{MoS}_2$ displays wavelength-tunable polarization polarity; and $\text{PdSe}_2/\text{NbSe}_2$ achieves a very high polarization ratio at 1550 nm.^[1-4] These reports collectively indicate that such behaviour is a general feature of anisotropic layered systems, originating from the energy-dependent optical anisotropy of the individual layers. In our device, the variation of the polarization ratio with wavelength can be understood as follows:

In the visible region, all three layers absorb efficiently, with photocarriers generated mainly near the top interface. In the near-infrared, the MoTe_2 layer becomes transparent, allowing deeper light penetration and shifting the absorption weight toward the top and bottom layers. Each layer has its own distinct anisotropic absorption spectrum; notably, the top layer exhibits strong excitonic anisotropy near its band edge owing to its quasi-1D chain structure. Therefore, the overall photocurrent anisotropy, being a weighted superposition of the layer contributions, naturally varies with wavelength.

At the same time, even within a single material, the transition matrix elements along different crystallographic axes are inherently energy-dependent. Thus, the intrinsic linear dichroism of each layer is not constant across the spectrum but changes with photon energy. The measured polarization ratio therefore reflects the combined effect of both the layer-weighted absorption and the energy-dependent anisotropy of each component.

[Actions]:

We have made the corresponding revisions in the page 9 and Supplementary Materials page 24 to clarify the origin of the observed wavelength dependence.

[1] *Advanced Materials*, 38, e13698. 2026.

[2] *Applied Physics Letters*, 122, 2023.

[3] *Advanced Materials*, 35, 2305594, 2023.

[4] *ACS Applied Materials & Interfaces*, 17, 5213-5222, 2025.

[Comments 9]: The x-axis in Figure 7, especially the most relevant 7a, should be logarithmic to resolve the first 4 datapoints.

[Response]:

We thank the reviewer for this valuable suggestion. As suggested, we have replotted Supplementary Fig. 7a using a logarithmic scale on the x-axis. The revised figure has been updated in the manuscript.

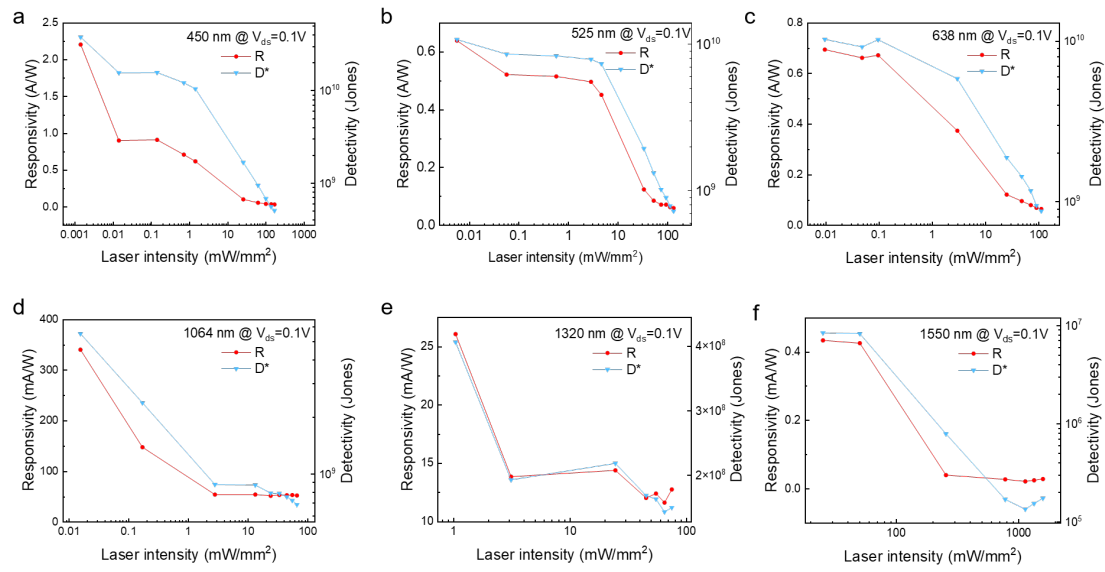


Figure R16. Device characterization under different light illumination at $V_{ds} = 0.1$ V.

REVIEWER COMMENTS

Dear editor,

Thank you very much for providing us with the constructive comments from the reviewers. We would like to thank all the referees for their critical comments that allowed us to further improve our manuscript. The manuscript has been revised according to the reviewer's comments, and a point-by-point response is given below.

Reviewer #1:

I am very pleased to see the extensive experiments and analysis added by the authors, which makes the manuscript more substantial and reliable. I believe the authors has provided sufficient responses to the reviewer's comments, so I suggest publishing it in Nature Communications.

[Response]:

We sincerely thank the reviewer for the encouraging assessment and recommendation for publication. The insightful comments have greatly improved the quality of the manuscript.

Reviewer #2:

The revisions to the manuscript, especially the additional information supporting the presented energy band characteristics, evolution, and charge carrier transport mechanisms are welcome, but further questions remain particularly with regards to the claimed neuromorphic applications of the presented devices.

[Response]: We thank the Reviewer for the encouraging feedback. These comments are very important for improving the quality of our manuscript. We provide point-to-point responses as follows.

[Comments 1]: *Authors explained that polarization sensitivity could provide an additional information dimension for machine vision, particularly under glare, scattering, or low-contrast conditions. However, the neuromorphic demonstrations (Fig. 5) still did not directly utilize the polarization information. Although authors' explanation makes sense, the direct demonstration of the role of polarization is still absence. Polarization-related demonstrations may be difficult because the device's core operating mechanism stems from heterojunction band alignment rather than material anisotropy. Authors should reconsider where to present the results of the polarization*

sensitivity analysis—whether in the main body of the paper or in supplementary materials—to ensure the paper focuses more consistently on the device's core mechanism and demonstrated applications.

[Response]:

We thank the Reviewer for this valuable suggestion. To ensure the manuscript focuses more consistently on the core device mechanism and demonstrated applications, we have moved all the polarization-related data, analysis, and discussions to the supplementary materials.

[Actions]: We have made the corresponding revisions on Fig. 3 and page 8 of the revised manuscript, Supplementary Material page 25.

[Comments 2]: *Regarding Reviewer 1: Comment 5 and Reviewer 3: Comment 2, the DT to TE transition is described by the introduction of a sigmoidal switching function connecting the DT and TE models laid out in the manuscript – though intuitive, there is little physical explanation given as to the choice of switching function and the fit of the proposed model to the experimental data is simply described as “reasonably good agreement”, and the purported experimental transition data does not appear to be included in Figure R15 for comparison. I would like to know the parameters utilized to conclude adequate fit of the model to the data for the sake of robustness.*

[Response]:

We sincerely thank the Reviewer for this valuable comment. We address this issue from the following three aspects.

1. Physical rationale for the sigmoidal function

First, 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. 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 response (*Nature* 2008, 453, 80–83). 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 framework (*Nat. Mater.* 2013, 12, 114–117). In resistive switching devices, the field-driven growth and rupture of conductive filaments have also been modeled using sigmoidal functions (*Nat. Commun.* 2017, 8, 15204). 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.

2. Model fitting parameters

The DT response time was fitted using the expression

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

the TE response time was fitted using

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

where the barrier height varies linearly with bias

$$\phi(V) = \phi_0 + \alpha V$$

The total response time across the full bias range is obtained by connecting the two mechanisms via the sigmoidal function:

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

$\tau_0 = 257$ 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.

3. Comparison between model and experiment

The theoretical curves and experimental data are shown together in Figure R1. Overall, the model captures the general trend of the bias-dependent response time. We note two deviations, which we discuss below.

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 MoTe_2 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 MoTe_2 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 MoTe_2 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.

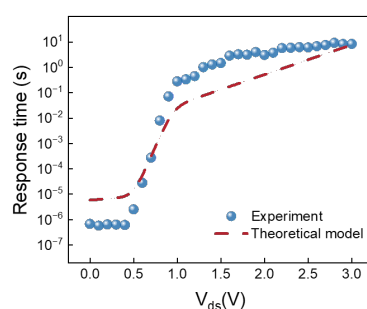


Figure R1. Comparison between the theoretical model and experimental data for the bias-dependent response time.

[Actions]: We have made the corresponding revisions on Supplementary Material page 16-18.

[Comments 3]: *The presented response made in evaluation of the platform's memory capacity and*

nonlinearity presents several highly puzzling issues that require further explanation from the authors. There are multiple data points in Figure R3a that can be clearly identified where the ‘sawtooth’ after one pulse is lower than that of the preceding pulse, suggesting that the claimed 128+ distinguishable states are an exaggeration. Though visually appealing, the use of normalized G with a varying number of pulses means that Figure R3c is useless for statistically and proportionately representing the nonlinearity in the conductance change between pulses due to the different scales between data points for each trace – the four curves could all belong to the exact same reading and it would be similarly difficult to differentiate. The usage of normalized G as the Y axis is more applicable for cases such as differentiating at a glance the difference in nonlinearity between traces of varying pulse intensity/time but with the same number of pulses. Figure R3d is effectively meaningless as $G_{\min}$ barring environmental factors should be a constant value, which suggests the graph is no different from any of the other G /pulse traces just divided by the common factor $G_{\min}$. Finally and perhaps most significantly, Figure R3e and its associated equation appear to have misinterpreted a term obtained from a general fitted curve equation – L should be a measure of linearity, not nonlinearity, which directly contradicts their explanation and understanding of the nonlinear characteristic of the presented platform (which notably is still present just in the reverse form – nonlinearity is strongest across the initial pulses and weakest the closer it approaches saturation). This may be observed via rearranging the equation and obtaining the second derivative (itself a measure of nonlinearity) w.r.t the number of input pulses:

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

$$\frac{G_{LTP} - G_{\min}}{G_c} = G_{\text{normalised}} = 1 - e^{-\frac{P}{L}} \quad \frac{dG_{\text{normalised}}}{dP^2} = -\frac{1}{L^2} e^{-\frac{P}{L}}$$

hence |curvature| $1/L^2$ - L inversely scales with nonlinearity. Alternatively, this may intuitively be shown by the observation that the change in conductance decreases the closer the platform approaches saturation – relative difference between pulses decreases – the graph of the peak conductance of each pulse approaches a flatline (fully linear).

[Response]:

We thank the Reviewer for the valuable comments. We respond to the specific points below.

1. In original Figure R3a, some of the conductance values recorded after a pulse are slightly lower

than those recorded after the preceding pulse, which may be attributed to two factors. The first is the readout noise of the measurement system. Because the conductance change between adjacent pulses is relatively small, random fluctuations in the instantaneous readout can occur when the noise amplitude is comparable to the state increment, leading to occasional decreases in the values. The second is the fast short-term relaxation of the device between pulses. In synaptic devices, after a stimulation pulse, a fast decay typically coexists with the long-term changes being characterized. These factors suggest that the evaluation of distinguishable states should be treated with caution, and the statement in the original manuscript was indeed not sufficiently rigorous. To better assess the multi-level conductance modulation, repeated measurements were performed under the same conditions: the LTP test was repeated four times, each with 32 consecutive pulses (Figure R2). The monotonic increasing trend of conductance with pulse number was reproducible across repeats, and the average increment between pulses was larger than the readout noise. Based on these results, the corresponding description has been adjusted in the revised manuscript, placing less emphasis on a specific number of distinguishable states and instead presenting the multi-level conductance tunability as qualitative evidence of synaptic plasticity.

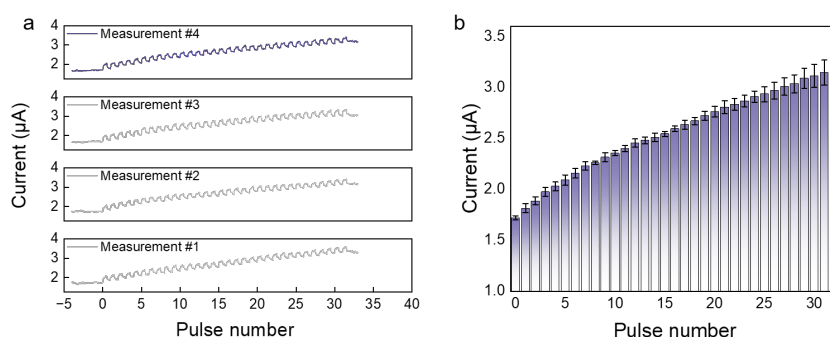


Figure R2. Repeated LTP measurements under identical stimulation conditions.

2. The original Figure R3c used normalized conductance, which made it difficult to fairly compare the nonlinearity under different conditions. The normalized plot has been removed and replaced with a plot showing the raw conductance change (Figure R3).
3. The original Figure R3d has been removed from the revised manuscript.
4. Regarding parameter L , we thank the Reviewer for the correction. The parameter L in the fitting equation was interpreted as a nonlinearity factor, which was not accurate. As the Reviewer pointed out, the second derivative of the normalized conductance with respect to the pulse number is

$$\frac{dG_{normalised}}{dP^2} = -\frac{1}{L^2} e^{-\frac{P}{L}}$$

whose magnitude reflects the local curvature of the conductance trace. Therefore, a larger L corresponds to a smaller curvature and a response closer to linear, while a smaller L corresponds to a larger curvature. Accordingly, L is more appropriately regarded as a linearity index or saturation constant, rather than a direct measure of nonlinearity. To address this issue rigorously, we re-analyzed the conductance data. We first redefined G_c as the theoretical maximum achievable conductance swing, i.e., the extrapolated saturation limit, and fixed it as a global constant. This eliminates the coupling between G_c and L that arose when G_c was treated as a floating fitting parameter. We then re-fitted all non-normalized conductance traces under 16, 32, 64, and 128 pulse conditions using the same equation, with only L as the single parameter. These results show that the fitted L values exhibit small variations across different pulse-number conditions, which supports the interpretation of L as an intrinsic saturation parameter of the device.

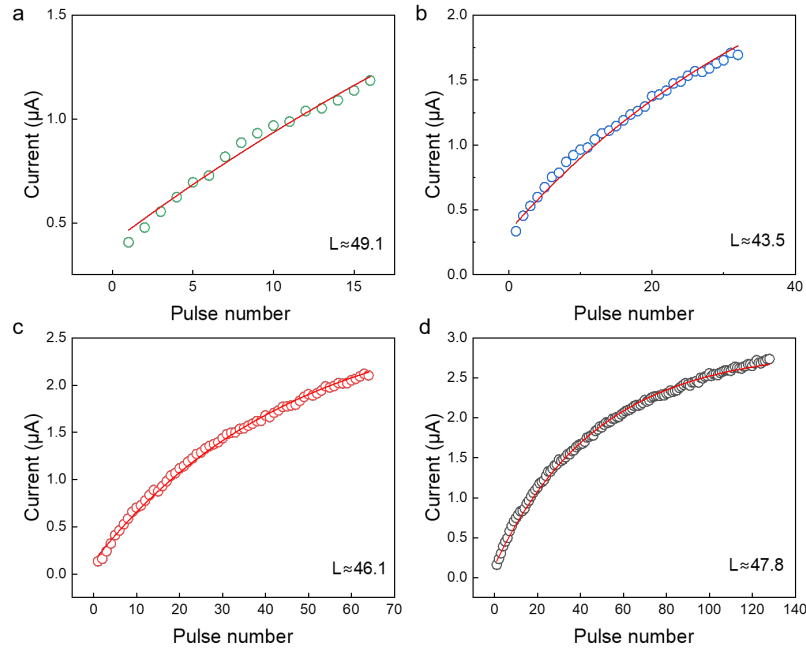


Figure R3 Conductance evolution as a function of pulse number under 16-pulse, 32-pulse, 64-pulse, and 128-pulse conditions, respectively, with the corresponding fitted curves.

[Actions]: We have made the corresponding revisions on page 10 of the revised manuscript, Supplementary Material page35.

[Comments 4]: *There are further issues when examining the newly presented benchmark testing. Figures only for training accuracy instead of test accuracy are given and what is clearly a*

handwritten “9” is presented as a “0” in Figure R5a – unless mislabeled in the dataset (in which case why use this data point as a figure), it is odd how such a mistake could occur. In addition, Fashion-MNIST was not intended to be used in 1-bit depth applications (as is presented with binarization), and 90% accuracy even in training warrants further scrutiny. Finally, the actual model architecture is entirely opaque to the reader of the manuscript – a basic overview of the model and clarity on its architecture should be a fundamental requirement, as well as description as to how the simulation/actual device readings were performed.

[Response]:

We sincerely thank the Reviewer for the thorough and careful review. We address these concerns from the following three aspects.

1. First, regarding the mislabeling of the handwritten digit in Figure R5a, this was a writing error during data compilation, where the displayed digit is clearly a “9”. We have corrected the label and the corresponding caption in the revised manuscript, and we have carefully double-checked all other dataset-related figures to avoid similar mistakes (Figure R4). We apologize for any confusion this may have caused.

2. Second, concerning the binarized Fashion-MNIST input and the absence of test accuracy, we fully agree that Fashion-MNIST, as a grayscale dataset, contains far richer information than its binarized version, and a training accuracy of 90% indeed reflects the information loss caused by binarization. We adopted binarized inputs for two reasons. First, such simplified input schemes have been used as reference benchmarks in several neuromorphic hardware evaluations. Second, we intended the binarized experiment to serve only as a baseline reference, to show that the device can still perform basic classification under simplified conditions. It merely provided supplementary functional evidence. Moreover, we have now added the test accuracy for the binarized Fashion-MNIST task. The training and test accuracy are 90% and 83%, respectively.

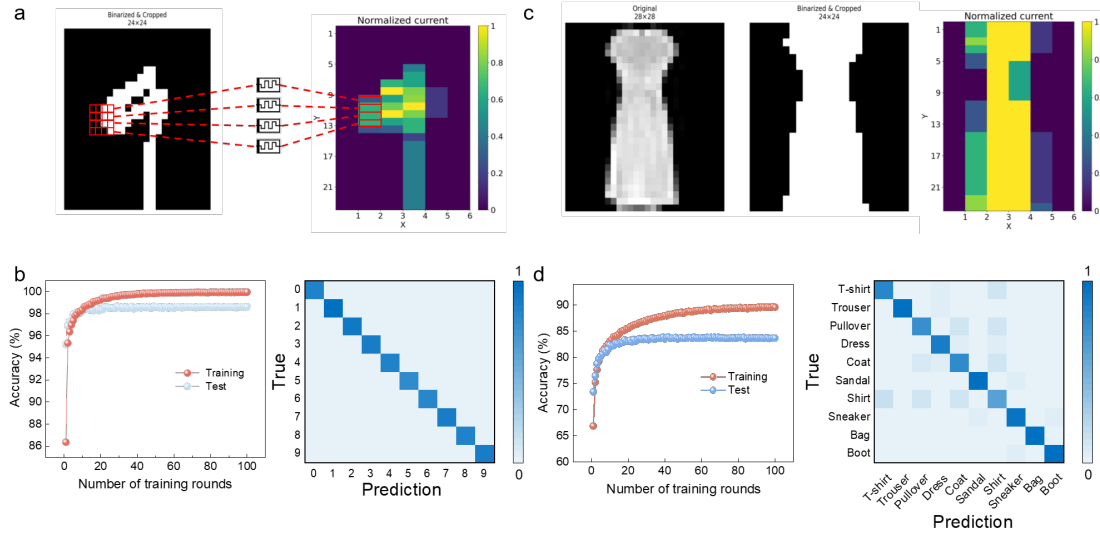


Figure R4. (a) Recognition of the handwritten digit "9" from the MNIST dataset, showing the reservoir output for digit "9". b) Recognition of a "dress" image from the Fashion-MNIST dataset, showing the reservoir output for "dress". c) Training and test accuracy after 100 epochs for digit pattern recognition 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.

3. Third, regarding the lack of transparency in the model architecture and simulation details, we agree that the original manuscript lacked a clear description. In the revised manuscript, we have now included the full details. For the motion trajectory recognition task, we built a custom dataset containing four types of car motion trajectories from real-world scenes (straight driving, left turn, right turn, and U-turn), resulting in 24 class labels in total. The dataset consists of 21,600 training images and 2,400 test images, and the neural network architecture is an artificial neural network (ANN). The input layer takes grayscale spatiotemporally fused images of size 300×300 pixels. The network body consists of three fully connected hidden layers with 1024, 512, and 256 neurons, respectively, all using ReLU activation and Dropout regularization. The output layer has 24 nodes and uses the Softmax function for multi-class prediction. Training uses the Adam optimizer with a learning rate of 1×10^{-4} , a batch size of 64, and a maximum of 100 epochs, with mixed-precision training enabled to accelerate GPU computation. A learning rate decay strategy is also employed to prevent overfitting. For the speed classification task, we generated a large-scale speed classification dataset using a memristor array simulator. The dataset includes six speed levels, covering a full

range from very slow to fast. The six speed levels are defined as: very slow (10–50 pixels/s), slow (50–100 pixels/s), moderately slow (100–150 pixels/s), medium (150–200 pixels/s), moderately fast (200–250 pixels/s), and fast (250–300 pixels/s). The data generation process adopts a memristor decay function to simulate spatiotemporal fusion. For each sample, the system captures four uniformly distributed motion frames on a 100×500 pixel array, and the temporal information is encoded into a single image through the double-exponential decay characteristics of the memristor. The final dataset contains 10,800 training images and 1,200 test images, with 900 training samples and 100 test samples per class. We used an ANN speed classification. The network architecture includes an input layer (flattened 50,000-dimensional vector), four fully connected layers (1024, 512, 256, 128 neurons, all using ReLU activation and Dropout regularization), and a 12-class output layer with Softmax activation. Regarding the simulation of device readings, we modeled the measured synaptic decay characteristics of the device as the weight update rules of a programmable resistor array, thereby connecting the device physics with the algorithm model at the simulation level. All simulation programs were written in Python 3.9 and built on the PyTorch deep learning framework.

[Actions]: We have made the corresponding revisions on page 12 of the revised manuscript, Supplementary Material page 4, 39.

[Comments 5]: *The x-axis label of the third graph in Figure R17 should spell “Light modulation frequency”.*

[Response]:

Thank you for pointing out this typographical error. We have corrected the x-axis label to “Light modulation frequency” in the revised Figures. In addition, we have carefully reviewed and corrected the formatting and spelling throughout the manuscript.

[Actions]: We have made the corresponding revisions on Fig. 3 of the revised manuscript, Supplementary Fig. 22.

[Comments 6]: *Previously, photodetector performance was experimentally demonstrated only up to approximately 3.1 μm , while the 3.8 μm result was limited to synaptic measurements. The authors*

explained that 3.8 μm lies near the absorption edge of Ta_2NiSe_5 , resulting in a weak low-bias photoresponse. After optimizing the measurement conditions, they added dark/light I–V curves and power-dependent photocurrent measurements at 3.8 μm . These results demonstrate the existence of photodetection at 3.8 μm . However, wavelength-specific responsivity, detectivity, and response time at 3.8 μm are not clearly provided. Therefore, operation at 3.8 μm is supported, but its quantitative photodetector performance remains insufficiently characterized.

[Response]:

We thank the Reviewer for this careful assessment. Following the suggestion, we have extracted the key figures of merit at 3.8 μm from the measurements. The responsivity is approximately 0.3 mA/W, the specific detectivity is approximately 3.9×10^6 Jones, and the response time is approximately 39.7 ms (Figure R5). These values reflect the weaker photoresponse expected near the absorption edge of Ta_2NiSe_5 .

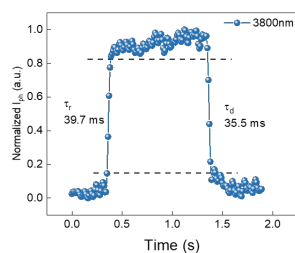


Figure R5. Response time of the device at 3.8 μm .

[Actions]: We have made the corresponding revisions on Supplementary Material page 24.

Reviewer #3:

The authors have expanded their already impressive wealth of experimental data to support their claims. While not always easy to read and follow, after full inspection of the various new datasets provided, I find the additional work mostly adequate and suitable to sufficiently support the main conclusions of this manuscript. This was my main concern with the previous version, and this has been removed. Therefore, I can now recommend the acceptance of this work for publication in Nature Communications.

[Response]:

We sincerely thank the reviewer for the positive assessment and recommendation for acceptance. The constructive comments have been very helpful in improving the manuscript.

Summary

The manuscript reports a van der Waals heterostructure photodetector based on $\text{Ta}_2\text{NiSe}_5/\text{MoTe}_2/\text{PdSe}_2$, designed to enable bias-controlled modulation of carrier transport across an interfacial barrier. By tuning the applied bias, the authors claim a continuous transition from direct tunneling to thermionic emission, resulting in a modulation of the photoresponse time over seven orders of magnitude (from ~ 600 ns to >8 s). This allows the device to operate either as a high-speed photodetector or as an optoelectronic synapse. The device further exhibits broadband photodetection (450 nm to $\sim 3\text{--}3.8$ μm), polarization sensitivity, and is demonstrated in neuromorphic tasks such as reservoir computing and motion recognition, achieving reported accuracies above 96%. The work aims to provide a unified hardware platform for integrated sensing and neuromorphic vision.

Overall, this work contains a wealth of experiments, treats a timely topic, and exhibits some performance parameters that seem to go beyond the state-of-the-art. However, the central claims — particularly the continuous seven-orders-of-magnitude modulation mechanism and its interpretation in terms of transport physics — are not sufficiently substantiated. As such, it may be of interest to the broad readership of Nature Communications, but this would require substantial additional analysis and clarification.

Major points

- The central claim of continuous response-time modulation over seven orders of magnitude is attributed to a transition between direct tunneling and thermionic emission. However, the evidence provided is indirect and largely qualitative. The tunneling vs. thermal emission regimes are inferred from I–V characteristics and Arrhenius fits, but no direct spectroscopic or time-resolved carrier dynamics measurements are presented to validate this transition.
- To this point, it is puzzling that the extracted barrier heights ($\sim 0.18\text{--}0.21$ eV) show only modest variation, which does not obviously explain such a dramatic change in response time. A quantitative model linking barrier modulation to time constants is missing.
- Furthermore, I would like to see a more rigorous demonstration of the highly bias-dependent response times shown in Fig. 1b. In principle, this has been done well in Fig. 3e, however only for the isolated case of $V = 0.1$ V and a repetition rate of 20 kHz. The choice of the repetition rate for this bias is sufficiently justified by Fig. S11b. I would like to see the corresponding analyses for three more voltages up to $V = 3.0$ V, including the power spectra to justify the applied repetition rates (which, I assume, would have to be smaller for larger voltages)

- In Figure 3f, the authors claim a response time of ~ 300 ns (justified by Fig. 3e) in combination with a responsivity of 2 A/W . However, this responsivity was only shown for the lowest power density in Fig. 7a. At which power density was the response time obtained? Was this really the same (very low) power density? This would surprise me. I would like to see the unnormalized plot of Fig. 3e to see the absolute current values.
- The long response times at high bias are attributed to trapped carriers and barrier-limited transport. However, PPC is a common phenomenon in 2D materials and heterostructures and is often dominated by defect states or interface traps. The manuscript does not convincingly distinguish between intrinsic barrier effects and extrinsic trap-assisted processes. Control experiments (e.g., temperature-dependent decay dynamics, illumination history dependence) would be necessary.

Minor points

- It is unclear what the color map in Figure 1a refers to.
- The y-axis label in Figure 5d should read “true”
- What is the rationale behind the wavelength-dependent polarization dependence shown in Figure S9?
- The x-axis in Figure 7, especially the most relevant 7a, should be logarithmic to resolve the first 4 datapoints.

The revisions to the manuscript, especially the additional information supporting the presented energy band characteristics, evolution, and charge carrier transport mechanisms are welcome, but further questions remain particularly with regards to the claimed neuromorphic applications of the presented devices.

1. Authors explained that polarization sensitivity could provide an additional information dimension for machine vision, particularly under glare, scattering, or low-contrast conditions. However, the neuromorphic demonstrations (Fig. 5) still did not directly utilize the polarization information. Although authors' explanation makes sense, the direct demonstration of the role of polarization is still absence. Polarization-related demonstrations may be difficult because the device's core operating mechanism stems from heterojunction band alignment rather than material anisotropy. Authors should reconsider where to present the results of the polarization sensitivity analysis—whether in the main body of the paper or in supplementary materials—to ensure the paper focuses more consistently on the device's core mechanism and demonstrated applications.
2. Regarding Reviewer 1: Comment 5 and Reviewer 3: Comment 2, the DT to TE transition is described by the introduction of a sigmoidal switching function connecting the DT and TE models laid out in the manuscript – though intuitive, there is little physical explanation given as to the choice of switching function and the fit of the proposed model to the experimental data is simply described as “reasonably good agreement”, and the purported experimental transition data does not appear to be included in Figure R15 for comparison. I would like to know the parameters utilised to conclude adequate fit of the model to the data for the sake of robustness.
3. The presented response made in evaluation of the platform's memory capacity and nonlinearity presents several highly puzzling issues that require further explanation from the authors. There are multiple data points in Figure R3a that can be clearly identified where the 'sawtooth' after one pulse is lower than that of the preceding pulse, suggesting that the claimed 128+ distinguishable states are an exaggeration. Though visually appealing, the use of normalised G with a varying number of pulses means that Figure R3c is useless for statistically and proportionately representing the nonlinearity in the conductance change between pulses due to the different scales between data points for each trace – the four curves could all belong to the exact same reading and it would be similarly difficult to differentiate. The usage of normalised G as the Y axis is more applicable for cases such as differentiating at a glance the difference in nonlinearity between traces of varying pulse intensity/time but with the same number of pulses. Figure R3d is effectively meaningless as $G_{\min}$ barring environmental factors should be a constant value, which suggests the graph is no different from any of the other G/pulse traces just divided by the common factor $G_{\min}$. Finally and perhaps most significantly, Figure R3e and its associated equation appear to have misinterpreted a term obtained from a general fitted curve

equation – L should be a measure of linearity, not nonlinearity, which directly contradicts their explanation and understanding of the nonlinear characteristic of the presented platform (which notably is still present just in the reverse form – nonlinearity is strongest across the initial pulses and weakest the closer it approaches saturation). This may be observed via rearranging the equation and obtaining the second derivative (itself a measure of nonlinearity) w.r.t the number of input pulses:

$$G_{LTP} = G_C \left(1 - e^{-\frac{P}{L}}\right) + G_{min}$$

$$\frac{G_{LTP} - G_{min}}{G_C} = G_{normalised} = 1 - e^{-\frac{P}{L}}$$

$$\frac{d^2 G_{normalised}}{dP^2} = -\frac{1}{L^2} e^{-\frac{P}{L}}$$

hence $|\text{curvature}| \propto 1/L^2$ - L inversely scales with nonlinearity. Alternatively, this may intuitively be shown by the observation that the change in conductance decreases the closer the platform approaches saturation – relative difference between pulses decreases – the graph of the peak conductance of each pulse approaches a flatline (fully linear).

4. There are further issues when examining the newly presented benchmark testing. Figures only for training accuracy instead of test accuracy are given and what is clearly a handwritten “9” is presented as a “0” in Figure R5a – unless mislabeled in the dataset (in which case why use this data point as a figure), it is odd how such a mistake could occur. In addition, Fashion-MNIST was not intended to be used in 1-bit depth applications (as is presented with binarisation), and 90% accuracy even in training warrants further scrutiny. Finally, the actual model architecture is entirely opaque to the reader of the manuscript – a basic overview of the model and clarity on its architecture should be a fundamental requirement, as well as description as to how the simulation/actual device readings were performed.
5. The x-axis label of the third graph in Figure R17 should spell “Light modulation frequency”.
6. Previously, photodetector performance was experimentally demonstrated only up to approximately 3.1 μm , while the 3.8 μm result was limited to synaptic measurements. The authors explained that 3.8 μm lies near the absorption edge of Ta_2NiSe_5 , resulting in a weak low-bias photoresponse. After optimizing the measurement conditions, they added dark/light I–V curves and power-dependent photocurrent measurements at 3.8 μm . These results demonstrate the existence of photodetection at 3.8 μm . However, wavelength-specific responsivity, detectivity, and response time at 3.8 μm are not clearly provided. Therefore, operation at 3.8 μm is supported, but its quantitative photodetector performance remains insufficiently characterized.