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Dual-mode 0D/2D spatial asymmetry optoelectronic device enabled by in situ microzone femtosecond laser deposition

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Abstract

To develop artificial intelligence and humanoid robotics, it is crucial to fabricate advanced vision systems with high efficiency and versatility. A key challenge is the functional integration of high-speed photodetectors (PD) and neuromorphic vision sensors (NVS) into a single device, as current studies suffer from complex architectures or fabrication processes. Hence, we propose a universal Microzone Femtosecond Laser Deposition (M-FLD) technique that enables the localized, in situ deposition of zero-dimensional (0D) black phosphorus (BP) nanoparticles onto a two-dimensional (2D) MoS₂ channel by ablating a micro-scale solid-state target. By M-FLD and h-BN nanomask, we fabricated a spatial asymmetric 0D/2D heterostructure for highly integrated dual-mode optoelectronic device. By changing the direction of V_{ds} , the device can be converted from PD to NVS. Under the PD mode, the device can sense high-frequency optical signals up to 3030 Hz. Under the NVS mode, the device's optical energy consumption per activity is only 191.2 pJ. Based on the sensing and memory capabilities, the device is simulated for MNIST handwritten digit recognition, achieving an accuracy of up to 96.20%. This work provides a flexible and powerful platform for fabricating complex heterostructures, paving the way for highly integrated and reconfigurable neuromorphic vision systems.

Introduction

The rapid development of artificial intelligence (AI), humanoid robotics, and extended reality has placed increasingly demanding requirements on the efficiency and versatility of vision systems^{1–3}. A key direction in this field is the functional integration of high-speed photodetector (PD) and neuromorphic vision sensor (NVS) into a single, dual-mode device, which can enhance adaptability and system integration^{4–7}. To achieve the conversion of PD and NVS, current studies focus on three-terminal gate modulation^{4–7}, or adjustments to the bias

voltage magnitude^{4,6} and directions⁵. However, as shown in Tab. S1, these studies suffer from limited integration of three-terminal architecture^{6,8,9}, difficulty in achieving multiwavelength sensing and memory^{6,10}, and complexity of the fabrication process⁵.

To design this compact and reliable dual-mode neuromorphic vision system, high-performance materials and novel structures are needed. 0D/2D heterostructures offer a promising platform for building high performance and adjustable optoelectronic devices. 2D materials provide excellent favorable carrier mobility¹¹ and high photoelectric conversion efficiency¹², while 0D materials contribute unique modulation effects such as surface plasmon polaritons¹³, photogating¹⁴, and local regulation¹⁵. Currently, the integration of 0D/2D systems is typically achieved through non-localized methods such as drop-casting¹⁶, spin-coating¹⁵, or preparing and mixing 0D and 2D materials in solution¹⁷. These techniques lack spatial

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control, leading to on-chip contamination¹⁸ and preventing the precise, selective integration required for complex, multi-functional device arrays. Therefore, there is an urgent need for an advanced integration technique that enables the localized, in situ fabrication of 0D/2D heterostructures at the micro-scale.

Physical vapor deposition (PVD) is a cornerstone technique in semiconductor manufacturing, valued for its ability to produce high-quality, large-area materials^{19,20}. However, PVD methods are ill-suited for the micro-scale manufacturing of heterostructures. To achieve patterned deposition, the custom-fabricated shadow masks are often needed²¹, which complicates the process and introduces alignment challenges. Normally, physical deposition involves sputtering or ablating a macro-scale target to deposit material. This paradigm, however, also suggests a new approach: achieving localized and in situ micro-deposition by miniaturizing the material source (the “target”) and positioning it directly within the desired deposition zone.

Realizing ablation and deposition from a micro-scale target hinges on the controlled delivery of energy to the material with extreme spatial precision. The femtosecond (fs) laser is an ideal tool for this purpose. The ultrashort pulse duration enables “cold” ablation²², minimizing heat diffusion and collateral damage to adjacent sensitive materials²³, while its diffraction-limited focusing capability allows for energy delivery to a micrometer or even sub-micrometer spot²⁴. Some studies have shown that femtosecond (fs) laser can be used for the fabrication of nanoparticles²⁵, joining of nanowires²⁶, and localized modification of 2D materials^{27,28}. The unique advantages of fs laser processing therefore offer a promising pathway to realize the localized and micro-scale physical deposition by ablating a micro-target.

In this work, we propose a novel technique, Microzone Femtosecond Laser Deposition (M-FLD), to flexibly realize in situ dual-mode device preparation. We use a focused fs laser to ablate a micro-scale black phosphorus (BP) flake, serving as a localized solid-state target. This process enables the in situ deposition of BP nanoparticles (NPs) onto a MoS₂ channel with a feature deposition distance of approximately 16 μm . After deposition, the device exhibits two-order-of-magnitude enhanced electrical and optical sensing performance due to the charge injection from BP NPs. Additionally, the heterostructure can memorize light from UV to NIR (near infrared) via photogating effect of BP NPs. Interestingly, by leveraging the high precision of M-FLD in conjunction with a partial h-BN nanomask, we fabricated a spatially asymmetric 0D-BP/2D-MoS₂ heterostructure. This unique design allows the single, two-terminal device to be reversibly switched between a high-speed PD (up to 3030 Hz) and a low-power NVS (191.2 pJ per spike) simply by reversing the

voltage direction. The NVS can be used to mimic complex synaptic functions and achieve a high recognition accuracy of 96.20% in simulated handwritten digit recognition tasks. This work demonstrates the potential of M-FLD for the flexible and in situ construction of 0D/2D heterostructures and highly integrated humanoid vision systems.

Results

Design and fabrication of dual-mode optoelectronic device

In the human visual system, optical signals are perceived in the retina and converted into electrical signals, which are later transmitted via the optic nerve to the brain for further processing (Fig. 1a)²⁹. Throughout the processing of optical signals, the synapse is the basic unit for information sensing, memory, and processing³⁰. Therefore, for constructing a neuromorphic visual system, there have been considerable researches on the fabrication of neuromorphic vision sensor artificial optoelectronic synapses³¹. However, just as the human retina is unable to recognize coherent images of less than 24 frames per second (refresh rate $\approx$ 40 Hz), artificial optoelectronic synapses also have difficulty in real-time acquisition of fast, high-frequency image signals due to their slow optical response, limiting application scenarios (Fig. 1b)⁴. Therefore, the simultaneous realization of dual-mode conversion of a PD and NVS in a single device is essential for extending the application scenarios and improving the integration of the whole visual system.

0D/2D heterostructure is promising for the construction of dual-mode optoelectronic device. As shown in Fig. 1c, we process a novel way called M-FLD to fabricate 0D/2D heterostructure. The process features for 2D target and substrate, laser ablation of the target material to form a plasma, and deposition of new material (nanoparticle) onto the substrate. Due to the micro-area characteristics and absence of liquid involvement, M-FLD shows advantages over previous 0D/2D preparation techniques, including lower temperature and vacuum requirements, superior spatial resolution, in-situ preparation capability, broad material applicability, and clean operation (Fig. 1d). The evaluation criteria are shown in Tables S2 and S3.

Figure 1e demonstrates the fabrication process of the dual-mode optoelectronic device by M-FLD process. As shown in Fig. S1, MoS₂ was chosen as the channel material (substrate) due to its broadband optical sensitivity across ultraviolet (UV), visible, and near infrared light (NIR) spectral ranges³². BP was selected as a target material to prepare nanoparticles to modulate the photo response and recovery rate. Hexagonal boron nitride (h-BN), with its wide bandgap, was employed as a “nanoscale mask” to selectively protect regions of the MoS₂ channel and enabled precise control over the deposition area of BP nanoparticles during the M-FLD process. As shown in Fig. S2, all flakes were peeled from bulk crystals using



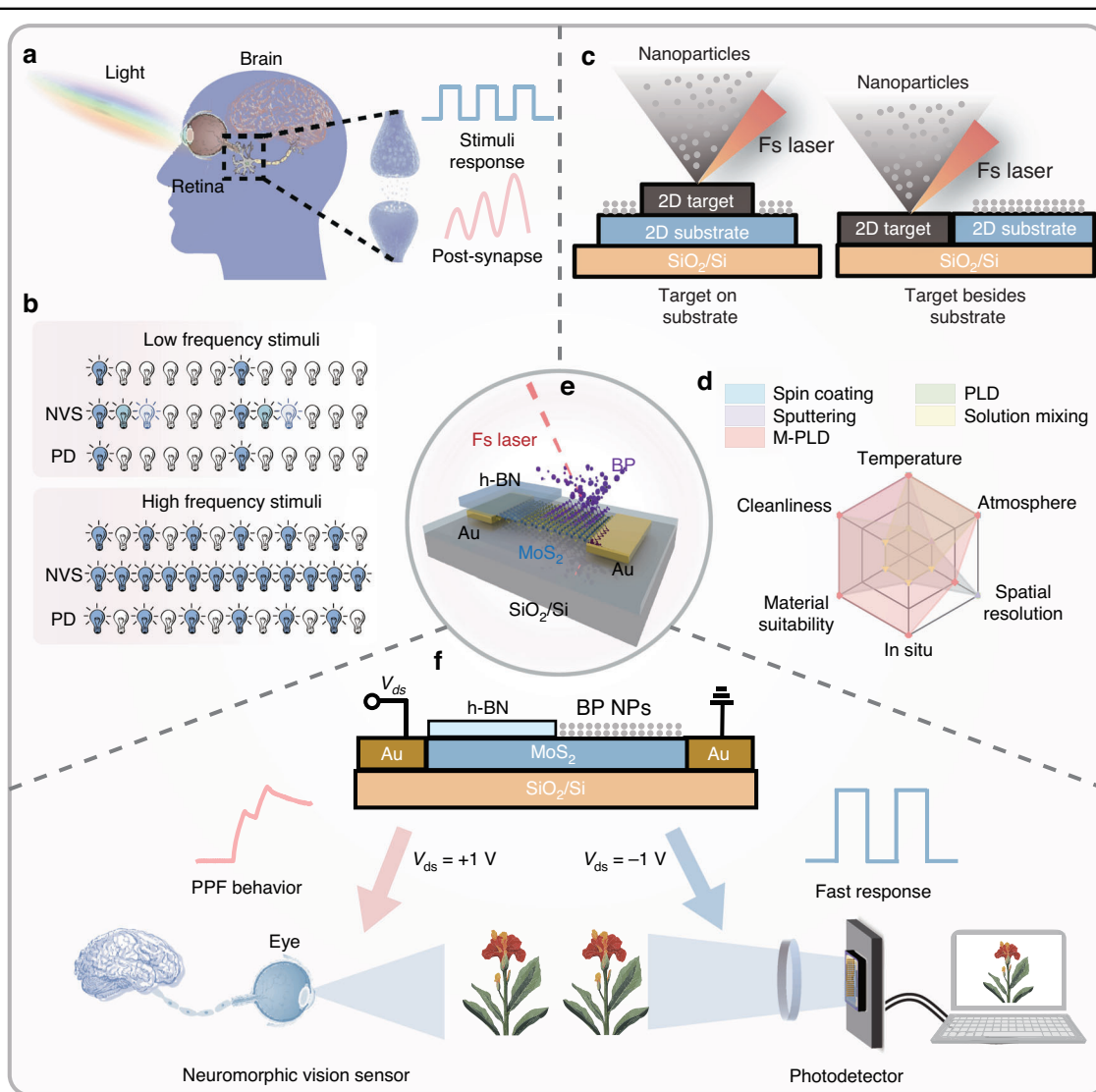


Fig. 1 Design and fabrication of dual-mode optoelectronic devices. **a** Schematic diagram of the human visual system. **b** Performance of the neuromorphic vision sensor (NVS) and photodetector (PD) under low- and high-frequency external light stimuli. The bulb indicates the external light source and highlights the device's sensing ability. **c** Schematic diagram of M-FLD. **d** Statistical graph of the fabrication process for 0D/2D heterostructure. **e** Fabrication of asymmetric device for dual-mode optoelectronic device by M-FLD. **f** Schematic diagram of dual-mode conversion between photodetector and NVS

adhesive tape and transferred to the chip substrate at a fixed position by dry transfer method. After that, the BP target was ablated using an fs laser. Under the action of the high-energy laser field, the BP flake underwent interlayer exfoliation and phase explosion processes and was formed into nanoparticles, which are later deposited back onto the bare MoS₂ surface in situ to form a 0D/2D heterostructure³³. In the h-BN mask-covered region, the MoS₂ was isolated from the BP NPs, thereby avoiding doping effects and interface modulation.

Hence, an asymmetric heterostructure was prepared and possessed asymmetric optoelectronic properties. As

demonstrated in Fig. 1f, at positive V_{ds} , the device exhibits significant paired-pulse facilitation (PPF) characteristics under light irradiation due to the photogating effect of BP NPs and the reduction of the Schottky barrier at the MoS₂/Au interface. This synaptic-like response enables the realization of NVS that integrates sensing and memory ability. At negative V_{ds} , the device can realize fast optical detection due to the isolation effect of h-BN, allowing its application as a photodetector for high-frequency optical signals. In this approach, the asymmetric 0D/2D heterostructure fabricated by M-FLD and h-BN mask can realize the dual-mode conversion of NVS

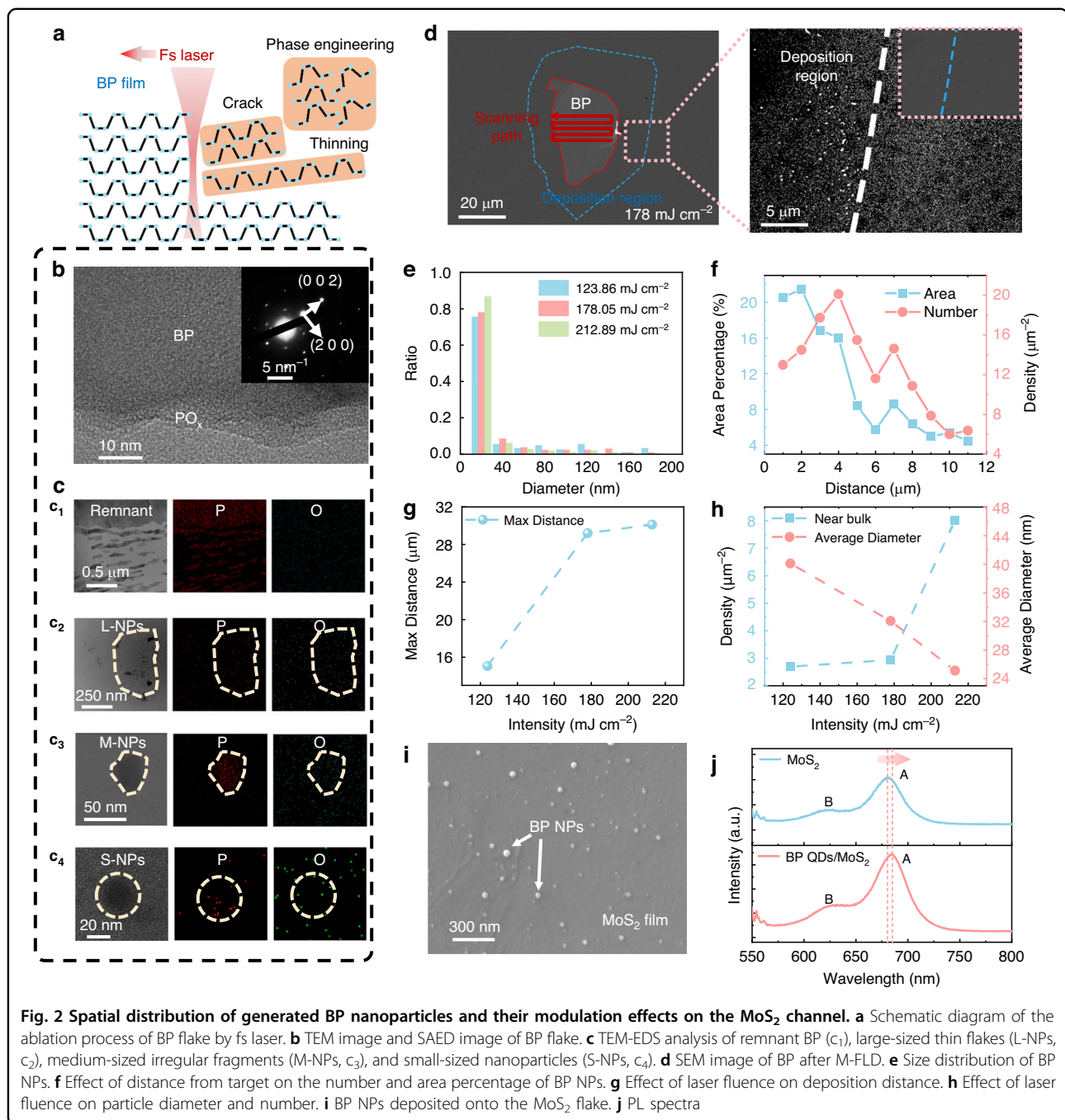


Fig. 2 Spatial distribution of generated BP nanoparticles and their modulation effects on the MoS₂ channel. **a** Schematic diagram of the ablation process of BP flake by fs laser. **b** TEM image and SAED image of BP flake. **c** TEM-EDS analysis of remnant BP (c₁), large-sized thin flakes (L-NPs, c₂), medium-sized irregular fragments (M-NPs, c₃), and small-sized nanoparticles (S-NPs, c₄). **d** SEM image of BP after M-FLD. **e** Size distribution of BP NPs. **f** Effect of distance from target on the number and area percentage of BP NPs. **g** Effect of laser fluence on deposition distance. **h** Effect of laser fluence on particle diameter and number. **i** BP NPs deposited onto the MoS₂ flake. **j** PL spectra

and photodetector. Additionally, comparing this work with current dual-mode devices (Fig. S3), it can be found that the asymmetric BP NPs/MoS₂ facilitates wide-band sensing and is favorable for high-density integration.

Controllable fabrication and morphological characterization of BP NPs via M-FLD

As shown in Fig. 2a, the fs laser ablation process of 2D-BP thin flakes involves phase explosion, layer by layer thinning, and crack extension^{33,34}. As a result,

nanoparticles of different sizes and morphologies, such as nanoparticles, irregular fragments, and thin flakes, are formed after ablation and deposition.

To characterize the crystal structure and morphology of the deposited BP NPs, BP flake was transferred to a SiC film carrier and ablated by fs laser for transmission electron microscope (TEM) observations. The TEM and high-resolution TEM (HRTEM) images of BP in Figs. 2b and S4 show the in-plane lattice with a space of 0.22 nm, corresponding to the (002) plane of BP. In



addition, the selected area electron diffraction (SAED) image of BP flake shows the (002), (202), and (200) crystal orientations of BP flake clearly, implying a single crystal structure. Four types of BP were observed under TEM in Fig. 2c: remnant BP (Fig. 2c₁), large-sized thin flakes (L-NPs, ~500 nm, Fig. 2c₂), medium-sized irregular fragments (M-NPs, ~70 nm, Fig. 2c₃), and small-sized nanoparticles (S-NPs, ~20 nm, Fig. 2c₄). The L-BPs, M-NPs, and S-NPs were individually induced by layer thinning, crack extension, and phase engineering. Compositional analysis by energy dispersive spectrometer (EDS) confirmed that the different BP regions consist predominantly of phosphorus, with no detectable oxygen, indicating minimal oxidation.

The core advantage of M-FLD lies in its excellent controllability and spatial locality. As shown in Fig. 2d, fs laser was utilized to process the BP flakes and the distribution range of the particles was evaluated under scanning electron microscope (SEM). To simultaneously resolve both nanoscale features and microscale spatial distribution of particles, high-resolution SEM images were taken with a resolution of 4.6 nm per pixel point. The region with deposited particles and the region without deposited particles can be clearly distinguished on the boundary. The influence of laser fluence on the deposition was tested (Fig. S5). As demonstrated in Fig. 2e, the size range of BP NPs is 0–200 nm in diameter, and more than 75% are less than 40 nm in diameter for different laser fluences. Within the deposition zone, the number and area percentage of NPs gradually decrease with distance from the ablation source (Fig. 2f). The deposition area (Fig. 2g) and number (Fig. 2h) exhibit a positive correlation with the laser fluence, which may be related to the fact that the higher the laser fluence, the higher the material removal rate (Fig. S7) and the higher the initial velocity of sputtering particles³⁵. The particle size (Fig. 2h) decreases with increasing laser fluence, which may be related to the intensification of phase explosion processes. The deposition range can be controlled to a minimum of 16 μm × 16 μm , demonstrating the high preparation accuracy of M-FLD.

After deposition onto the MoS₂ substrate (Fig. 2i), the BP NPs can modulate the properties of MoS₂. Raman and PL spectra of MoS₂ were measured before and after the deposition of BP NPs. The Raman spectrum in Fig. S8a shows an in-plane E_{2g}¹ mode near 383 cm⁻¹ and an out-of-plane A_{1g} mode near 409 cm⁻¹, indicating typical 2H-MoS₂ features³⁶. After the deposition of BP NPs, the full width at half maxima of the A_{1g}¹ peak increased from 7.5 cm⁻¹ to 8.4 cm⁻¹, implying the n-type doping of MoS₂ (Fig. S8b)³⁷. The E_{2g}¹ peak is nearly unchanged. In addition, three visible peaks appeared at 362 cm⁻¹, 468 cm⁻¹, and 437 cm⁻¹, representing the A_g¹, E_{2g}², and A_g² peaks of BP^{38,39}. The results of Raman spectra mapping shown in

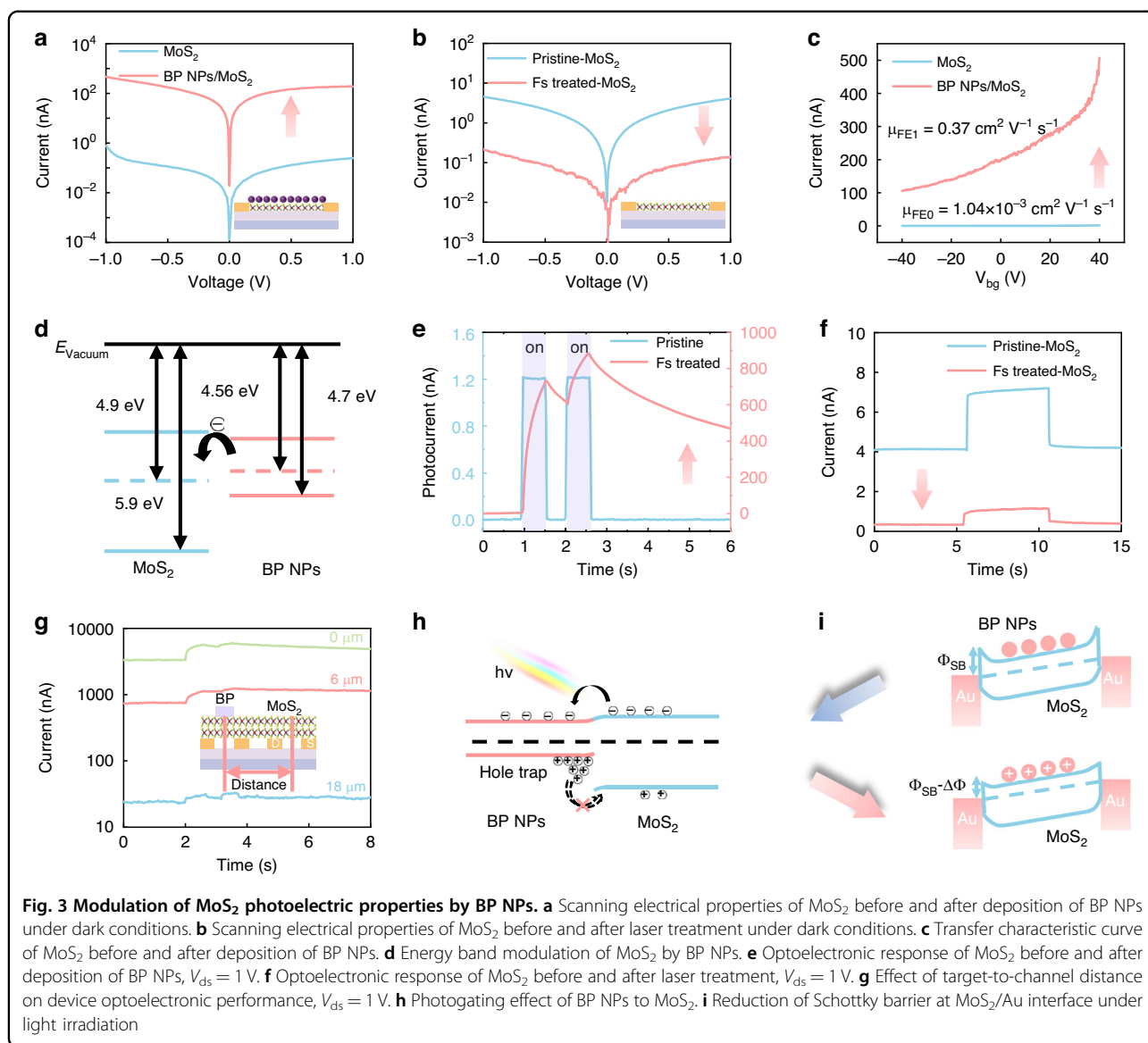
Fig. S9 also indicate that after deposition, the doping effect diminishes and the number of BP NPs gradually decreases with increasing distance from the BP target. In the PL spectra (Fig. 2j), the A exciton peak of MoS₂ showed red-shifted (from 680 nm to 685 nm) due to the increase in electron concentration⁴⁰.

The electron injection and photogating effect of BP NPs into MoS₂

Since the minimum ablation threshold of MoS₂ is tested to be about 150 mJ cm⁻² (Figs. S12 and S13, varies depending on thickness), which is lower than the maximum ablation threshold of BP (97 mJ cm⁻², Fig. S15, varies depending on thickness), selective ablation of BP flake can be performed without damaging the surrounding MoS₂. To ensure that MoS₂ is undamaged during the ablation of BP, the thickness is selected to be 8–30 nm (Fig. S16). Hence, as shown in Fig. S17, through precise thickness selection and differences in ablation thresholds, BP target can be placed besides or on the top of MoS₂ without damaging the MoS₂ substrate during M-FLD, enabling the preparation of in situ BP NPs/MoS₂ heterostructure. As demonstrated in Fig. 3a, the conductivity of the device is improved by two orders of magnitude after deposition of BP NPs, which is ascribed to the increase in electron concentration. As shown in Fig. 3b, the conductive ability of MoS₂ undergoes a reduction due to the narrowing of the conductive pathway, which excludes the influence of possible doping or phase transition effects caused by fs laser. Additionally, as shown in Fig. 3c, electron injection increases carrier mobility by two orders after deposition of BP NPs. Combined with PL, Raman, and energy band analyses, the improvement in the electrical properties of MoS₂ is attributed to n-type doping due to the electron injection effect of BP NPs into MoS₂. As demonstrated in Fig. 3d, the Fermi energy level of BP (-4.56 eV) is higher than that of MoS₂ (-4.9 eV)⁴⁰. When BP NPs and MoS₂ form heterostructures, the electrons in BP are injected into MoS₂ to realize the equilibrium of the Fermi energy level, leading to an increase in carrier concentration in MoS₂.

After M-FLD process, the device transitioned from subtle rapid light response and recovery to significant optical memory. As shown in Fig. S19a, the paired-pulse facilitation (PPF) index is often used to represent whether the device has synapse-like properties. PPF is 0 for photodetector while often greater than 0 for excitatory NVS due to the memory ability. As shown in Fig. 3e, the PPF index of the device increased from nearly 0 to about 30%, and photocurrent increased by two orders, representing the excellent memory effect for pulsed stimuli. The optical response of several devices is statistically characterized (Fig. S19b and c), demonstrating stability. Also, it is found that the photocurrent of ablated MoS₂





undergoes a reduction without memory effect due to the narrowing of the conductive pathway (Fig. 3f), which excludes the influence of possible doping or phase transition effects caused by fs laser.

The process of M-FLD also features for universality and flexibility. Various typical 2D materials were employed as target materials and MoS₂ were employed as substrate for the M-FLD process. The results indicate that the optoelectronic performance of the devices is improved after deposition of different kinds of nanoparticles. Among these, BP NPs exhibited the most pronounced regulatory effect on MoS₂ (Tab. S4). Additionally, the number of particles was found to have a significant effect on the photoresponse by adjusting the distance between the target and the channel (Fig. 3g), implying the flexible tunability of the M-FLD process to the photoresponse.

When placed on MoS₂, the BP NPs exhibits stronger performance regulation (Fig. S18). Hence, unless otherwise specified, all devices in this paper pertains to BP on MoS₂.

The excellent optical sensing and memory of the device come from the favorable light absorption of MoS₂ and the photo-gating effect of BP particles. As shown in Fig. 3d, the valence bands of the MoS₂ and the BP NPs are -5.9 eV¹⁴ and -4.7 eV⁴¹, respectively. When the heterostructure interface is irradiated by light, photogenerated electron-hole pairs are generated and separated under the built-in electric field. However, due to the bending of the valence band, the photogenerated holes are trapped in the BP particles, leading to strong positive charge accumulation (Fig. 3h)⁴². As a result, the positively charged BP NPs form a photogating effect on MoS₂, which reduces the

Schottky barrier between the MoS₂ channel and Au electrodes (Fig. 3i), resulting in an increasing (excitatory postsynaptic current) EPSC under light irradiation. Additionally, the large specific surface area of BP NPs provides a significant surface effect, creating an abundance of surface defect states⁴³. These defects are equivalent to a potential well for a certain charge, which can trap the inside charges and lead to long-lived net charge^{44,45}. Hence, when the outside light source is removed, photogenerated carriers are trapped in defect states, leading to a slow recombination rate of photo-generated holes and electrons. Additionally, the effects caused by strain resulting from contact between MoS₂ and the Au electrode are eliminated (Fig. S21 and S22).

Fabrication of dual-mode optoelectronic device by M-FLD

As demonstrated in Fig. 4a, the dual-mode conversion device with asymmetric response characteristics can be produced flexibly based on the M-FLD and h-BN nanomask. To enable in situ testing and comparative analysis of device performance, MoS₂ was transferred onto four Au electrodes. Electrodes 3 and 4 were covered with h-BN masks, while the central region between electrodes 1 and 2 was covered with a BP flake, serving as the source of BP particles. Afterward, fs laser is used to ablate the BP flake; the BP NPs will then be deposited on the MoS₂ surface, while h-BN hinders the contact of the particles with MoS₂ as shown in Fig. S23.

The fabricated devices in different regions show different optoelectronic responses (Fig. 4b). In the region of electrodes 1 and 2, the device exhibits good optical response and memory effects under both positive and negative conductance (Fig. 4b₁). The device shows typical fast photodetection capabilities in the region of electrodes 3 and 4, as the h-BN mask completely protects the MoS₂ (Fig. 4b₃). In the region of electrodes 2 and 3, the device exhibits an optical memory capability during positive conductivity and an apparent fast photodetection capability during negative conductivity, implying that the device can be converted to two modes of functionality by simply changing the direction of conductivity (Fig. 4b₂). Hence, the asymmetric device structure in the region of electrodes 2 and 3 can then be used as a dual-mode optoelectronic device for photodetector and NVS.

In addition, the *I-V* curves of different regions and the device before and after deposition of the particles are compared. As shown in Fig. 4c, after deposition of BP NPs, for completely exposed (electrode 1 and electrode 2) and completely protected (electrode 3 and electrode 4) regions, the *I-V* curves of the devices are symmetric at the center of the origin of the coordinate axes. The device shows rectification behavior with a rectification ratio of about 20. As demonstrated in Fig. 4d, the *I-V* curves of asymmetric region before and after deposition of BP NPs

are compared. The rectification ratio of the device before deposition of the particles is close to 1, which proves that the variation of the rectification ratio originates from the BP NPs and excludes the effect due to the shape factor of MoS₂. In addition, as demonstrated in Fig. 4e, this change in rectification ratio varies with the duration of outside light. As the history plus light time increases, the current of the device under dark conditions increases at +1 V, while remaining essentially unchanged at -1 V. The rectification ratio of the device can be adjusted from 40 to 66 by controlling the light irradiation time (Fig. S24).

The device exhibits certain environmental stability. As shown in Fig. S25, the device maintains stable performance during short-term testing. After being stored in air for 2 months, the device still retains the ability to memory light signals (Fig. S26a). After stored in an oxygen-free environment for 9 days, neither the forward NVS performance nor the reverse PD performance exhibited any noticeable deterioration (Fig. S26b-h). Hence, for future practical applications, it is necessary to isolate water and oxygen by encapsulation structures.

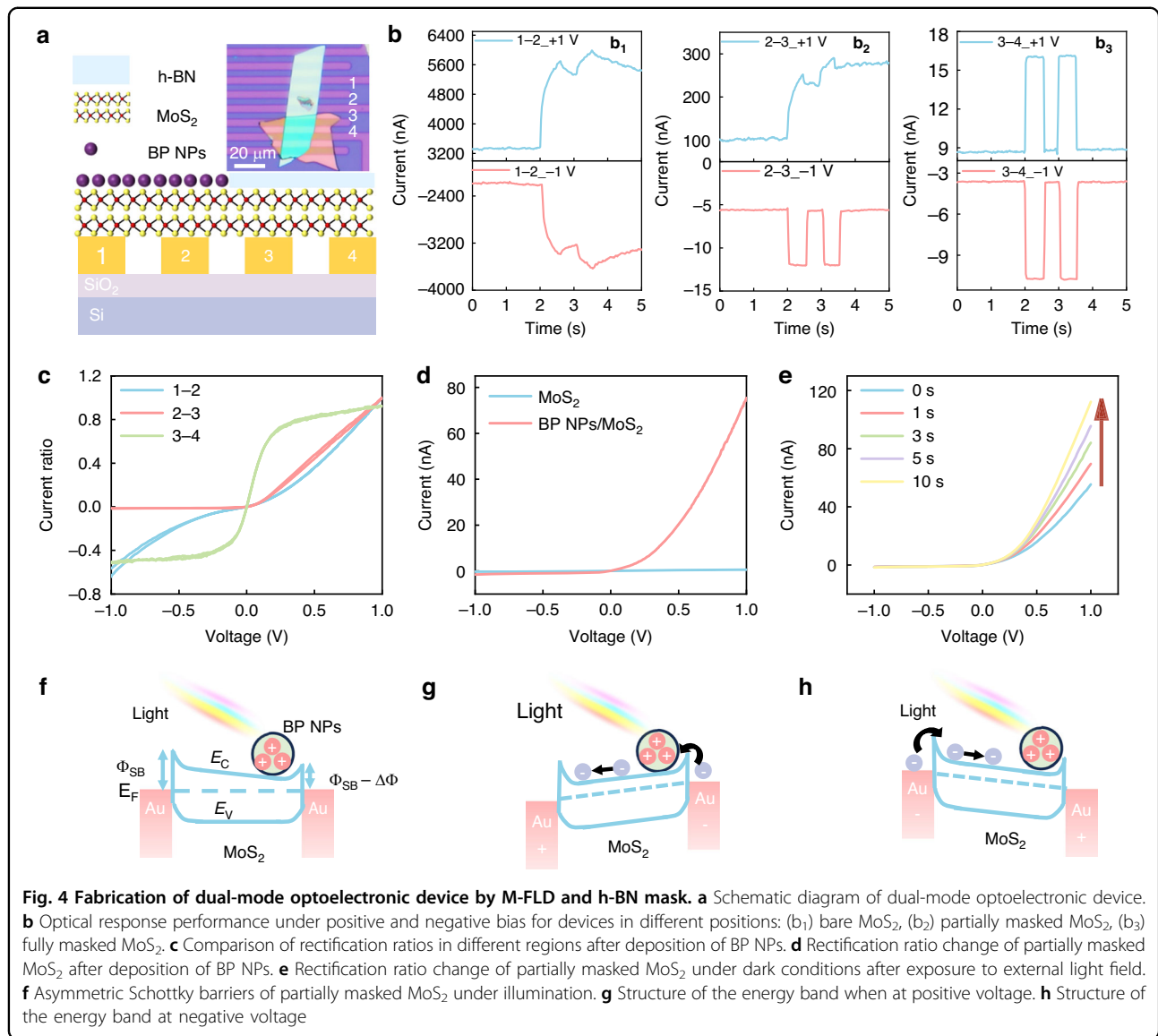
The inconsistency in dark current is caused by asymmetry doping. As shown in Fig. S27, during prolonged dark current testing, the current at $V_{ds} = +1$ V remains stable and is an order larger than that at $V_{ds} = -1$ V, suggesting that only the side of bared MoS₂ undergoes asymmetric permanent doping. Differences in light perception due to the direction of V_{ds} arise from structural asymmetry. As demonstrated in Fig. 4f, since BP NPs are deposited on only the bare side of the MoS₂, the Schottky barrier reduction due to the gating effect is present only on the exposed side. Therefore, when the exposed side is connected to the negative side of the electrode, the decrease in the Schottky barrier due to photogating causes a decrease in the barrier that impedes the movement of electrons, leading to an increase in current (Fig. 4g). When the bare side is connected to the positive pole of the electrode, the reduction of the Schottky barrier due to photogating has almost no effect on the conductivity due to the directionality of the electron motion (Fig. 4h). The photoelectric response was tested by irradiating different areas using a point light source, verifying the presence of a photogating effect only in the area where the BP particles are in contact with MoS₂ (Fig. S28).

Performance and application of the dual-mode device

Performance and application of the dual-mode device as a PD

As shown in Fig. 5a, the asymmetric device presents the ability of fast speed photo response at -1 V, providing the basis for the construction of the photodetector. As the intensity of external light increases, the photocurrent continues to increase (Fig. S29a). As shown in Fig. S29b, the incident optical power density dependence of the





photocurrent is fitted using the power law shown as:

$$I = A \cdot P_{\text{in}}^{\theta}$$

where A is constant, P_{in} is incident light power intensity, and θ is the power-law index. The fitted θ is extremely close to 1, indicating a good linear dependence property, which is favorable for decoding current data into light intensity data.

The dual-mode conversion device has a fast optical perception capability that exceeds the limit of the human eye at -1 V. As shown in Fig. S30a, the device exhibits clear separation at a fast optical switching frequency of 50 Hz. The photocurrent remains relatively constant during the test. Additionally, the rise time ($\tau_r = t_{90\%} - t_{10\%}$) and fall time of PD ($\tau_f = t_{90\%} - t_{10\%}$) were tested. As shown

in Fig. 5b and c, the τ_r is tested to be 210 μs and the τ_f was tested to be 120 μs . The theoretical maximum resolvable optical pulse frequency is about 3030 Hz, which has not yet been realized in this research due to the limits of the frequency of the light source (Fig. S30b). As shown in Tab. S5 and S6, the response speed of PD mode is comparable to that of mainstream works in the fields of MoS₂-based photodetector and dual-mode device.

The resolution of different light sources in fast photo-detection mode was tested (Fig. S32), and the D^* is about 1.02×10^{11} Jones at 525 nm. The photodetector has a similar sensitivity to light as the classic MoS₂ detector, being most sensitive to visible light while retaining the ability to detect UV and NIR. Based on the excellent photodetection capability and the ability to distinguish colors, the device can realize the logic “AND” and “OR” by



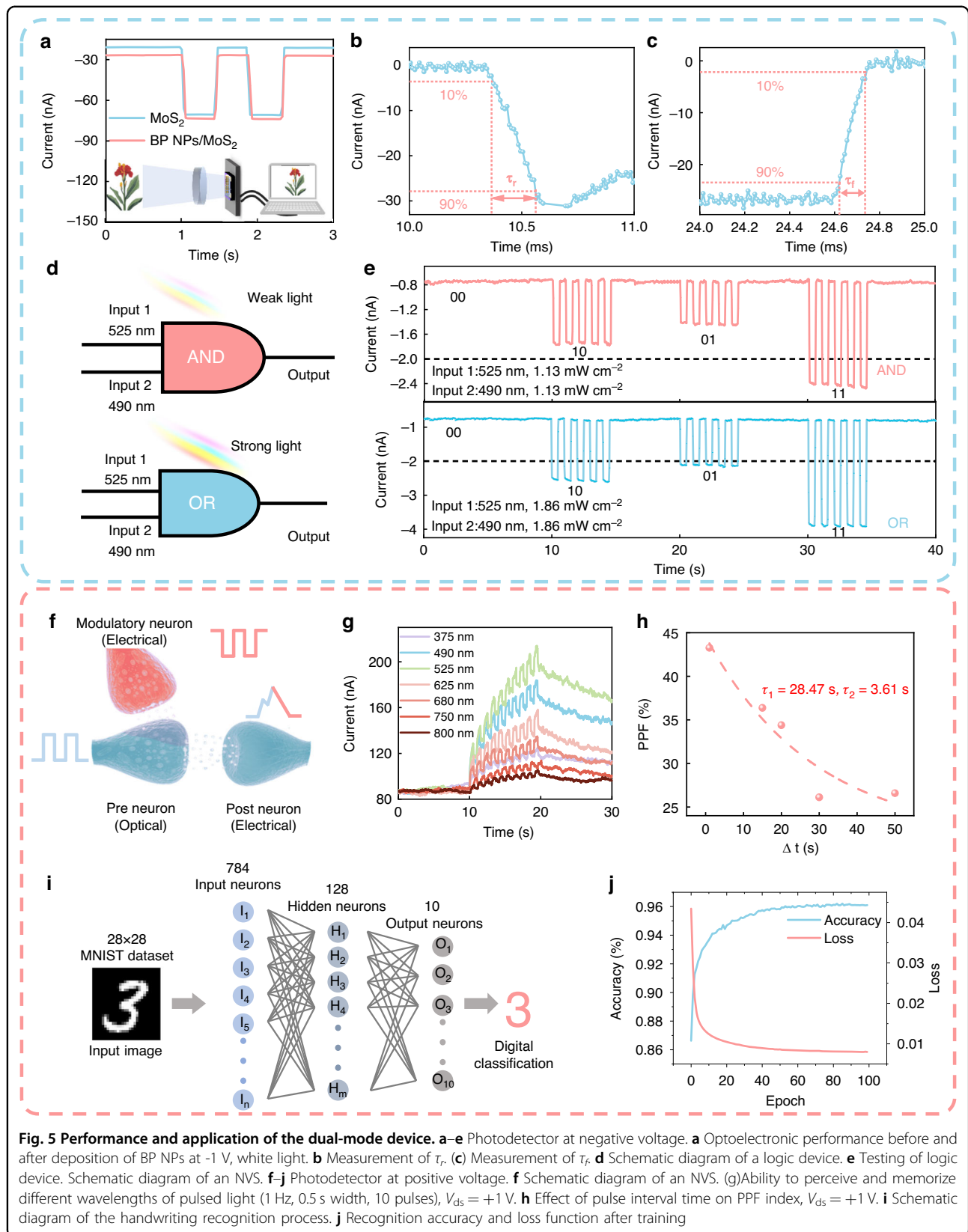


Fig. 5 Performance and application of the dual-mode device. **a–e** Photodetector at negative voltage. **a** Optoelectronic performance before and after deposition of BP NPs at -1 V, white light. **b** Measurement of τ_r . **c** Measurement of τ_f . **d** Schematic diagram of a logic device. **e** Testing of logic device. Schematic diagram of an NVS. **f–j** Photodetector at positive voltage. **f** Schematic diagram of an NVS. **g**) Ability to perceive and memorize different wavelengths of pulsed light (1 Hz, 0.5 s width, 10 pulses), $V_{ds} = +1$ V. **h** Effect of pulse interval time on PPF index, $V_{ds} = +1$ V. **i** Schematic diagram of the handwriting recognition process. **j** Recognition accuracy and loss function after training

controlling light intensity and wavelength (Fig. 5d), demonstrating its potential for use in multispectral optical logic devices. As shown in Fig. 5e, when the external light intensity is 1.13 mW cm^{-2} , the response current values of different colors are small. Only after the superposition of two colors of light, the size of the light response current exceeds the preset threshold, to achieve the “AND” logic. When the external light intensity is 1.86 mW cm^{-2} , monochromatic light irradiation can reach the threshold, realizing the “OR” logic.

Performance and application of the dual-mode device as an NVS

As shown in Fig. 5f, bio-like NVS device is realized by integrating optical memory effects with electrical modulation strategies. The window of optical sensing wavelength is shown in Fig. 5g. Based on the excellent absorption of visible light by MoS_2 , asymmetric BP NPs/ MoS_2 features favorable perception and memory of visible light, which is analogous to the perception of the human eye. Furthermore, the device also exhibits certain sensing capabilities for UV and NIR light, broadening the functionality of the eye-like visual system. The recognition effect of flower pattern demonstrates the device’s ability to discriminate colors and learning ability (Fig. S33).

Additionally, the ability to memorize the optical signals of the device is described. As shown in Fig. S34a, the natural relaxation process following the optical stimuli is fitted using the classical Kohlrausch stretched-exponential function³²:

$$I = I_{\infty} + (I_0 - I_{\infty}) \times e^{-\left(\frac{t-t_0}{\tau}\right)^{\beta}}$$

where τ and β represent the relaxation time constant and the stretching exponent, respectively. The relaxation time was fitted to be 35.58 s, indicating the nonvolatile ability. Subsequently, the variation of the PPF index with pulse time interval was fitted. (Fig. 5h). As the time between pulses increases, the PPF index tends to decrease, and the curve can be fitted as⁴⁶:

$$\text{PPF} = \frac{A_2 - A_1}{A_1} = C_1 \times e^{-\frac{\Delta t}{\tau_1}} + C_2 \times e^{-\frac{\Delta t}{\tau_2}}$$

where A_2 and A_1 represent the magnitude of the current values for the first and second impulse responses, C_1 and C_2 represent initial facilitation magnitudes, τ_1 and τ_2 represent characteristic relaxation time. τ_1 is one to two orders of magnitude larger than τ_2 , implying the presentation of properties similar to those of biological synapses⁴⁷.

The modulation of the device memory effect by optical pulse parameters was tested. As pulse intensity (Fig. S34b) and number of pulses (Fig. S34c) increase, the EPSC of the device is enhanced, leading to a transition of short-term

memory to long-term memory. The device can sense and memory light with low power intensity (0.04 mW cm^{-2}). As a result, the energy consumption of light irradiation can be reduced to 191.2 pJ, making it possible to fabricate an energy-efficient neuromorphic system. The max operating rate of NVS is about 50 Hz (Fig. S35), which is also close to the response speed of the human retina ($\sim 40 \text{ Hz}$).

In addition to facilitating excitation, depression of potentiation is also extremely important for realizing advanced functions in the neuromorphic system. Different kinds of neurotransmitters can be used in organisms to regulate excitation or inhibition of the post-synaptic neuron⁴⁸. As demonstrated in Fig. S36a, when increasing the scanning V_{ds} to $\pm 10 \text{ V}$, a significant change in the high resistance state (HRS) and low resistance state (LRS) is observed. The change in resistance due to the historical current states may be related to the sulfur vacancy defects inherent in the MoS_2 flake after exfoliation⁴⁹. Upon application of V_{ds} , the sulfur vacancies migrate and accumulate to the junction of MoS_2 and Au electrodes, causing the Schottky barrier to decrease, which results in a change in resistance from HRS to LRS⁵⁰. The negative voltage can rapidly reduce the magnitude of the response current after light stimulation in contrast to the natural forgetting process (Fig. S36b). Hence, the introduction of electrical stimuli provides the basis for the realization of advanced optoelectronic synaptic functions. The modulation of conductance by electrical stimulation was tested in detail in Fig. S36c-g.

As shown in Fig. 5i, to validate the feasibility of the artificial synapse fabricated by M-FLD for use in an artificial vision system, a three-layer spiking neural network (SNN) based on the impulse response data is used to recognize and discriminate handwritten digits from the Modified National Institute of Standards and Technology (MNIST). The image is segmented into 28×28 -pixel arrays, fed into 728 input neurons, passed through 128 hidden neurons, and output through 10 output neurons. The number of the input handwritten digits can then be determined by the values of the 10 output neurons. The test accuracy increases with the number of training epochs, as illustrated in Fig. 5j. The excellent visual recognition capability of the device is verified by optical potentiation, with the recognition accuracy up to 96.20% respectively after training. The loss curve and the confusion matrix shown in Fig. 5j and S36i also reflect the convergence and accuracy of the training process, implying the practicality of the device in artificial vision systems.

Application of the dual-mode device combined with PD and NVS

The PD mode and NVS mode can be operated simultaneously to enable real-time motion recording and noise



reduction at the same time. As shown in Fig. S37, under continuous light irradiation, the negative photocurrent remains stable, while the positive photocurrent exhibits a continuously accumulating memory effect. Therefore, by measuring the rate of increase in forward photocurrent under different light intensities ($I_1 = I_1(t, P)$) and the magnitude of negative photocurrent ($I_2 = I_2(P) = A \cdot P_{in}^\theta$), a single device can simultaneously achieve decoupling of light intensity and light exposure time instantly. Based on this, dynamic noise reduction for transient background noise can be achieved (Fig. S38). Assuming an arrayed device, the forward NVS function can remember and distinguish the historical motion state of an object as it moves. However, when transient noise is present, it interferes with the accuracy of motion recognition. At this point, subtracting the absolute value of the transient photocurrent recorded by the PD from the photocurrent of the NVS can be used for noise reduction.

Discussion

To conclude, we proposed a novel process for the in situ and flexible preparation of 0D/2D heterostructure based on the microzone pulsed laser deposition process. Compared with the previous preparation process for 0D/2D heterostructure, the M-FLD process features more precision ($\sim 16 \mu\text{m}$), simplicity, versatility, and universality, which is expected to be used to prepare large-scaled 0D/2D heterostructure arrays. Based on the M-FLD process and h-BN nanomask, we fabricated an environmentally stable asymmetric BP NPs/MoS₂ device which can realize the dual-mode conversion of photodetector and NVS. The device enables high-frequency optical detection at 3030 Hz at -1 V. Device enables low-energy neuromorphic activity at +1 V, consuming only 191.2 pJ for a single activity. We have realized various applications in various scenarios, such as logic judgment, colored pattern recognition, MNIST recognition training (with accuracy of up to 96.20%), real-time motion recording and noise reduction. Looking ahead, further control of the deposited particle size is also expected to be achieved by beam shaping and the use of monolayers in subsequent work. Overall, the excellent optoelectronic performance and diverse applications of this dual-mode device demonstrates the potential of the M-FLD process for the construction of high-performance and versatile vision systems.

Materials and methods

Device fabrication

A silicon wafer covered with a 300 nm SiO₂ layer as an insulating layer was used as the substrate. Cr/Au (5 nm/25 nm) electrodes were prepared on Si substrate by photolithography and magnetron sputtering. The MoS₂, BP, and h-BN flakes (bought from SixCarbon Technology

Shenzhen) were mechanically exfoliated from the bulk material using adhesive tape and picked by thermal-released polydimethylsiloxane (PDMS). As shown in Fig. S39, a typical thickness of MoS₂, BP, and h-BN is 22 nm, 35 nm, and 28 nm, respectively. Then, the PDMS with the material adhered was aligned to a specific location on the substrate using a micro-imaging system, and the material was released by heating (80 °C, 20 min).

M-FLD process

The samples were placed on a high-precision motorized stage (ProScan III, Prior Scientific, positioning accuracy of 0.01 μm) and imaged for observation using an optical microscope (Olympus, BX51) and CCD module. Fs laser (800 nm, 1000 Hz, 50 fs) produced by Ti: sapphire mode-locked oscillator was used as the high-energy physical field for material ablation. An electronic shutter was used to control the irradiation time of the fs laser. The optical path structure for the fs laser is shown in Fig. S40. A 20X objective lens was used to focus the laser beam into a spot approximately 4 μm in size. During the ablation deposition process, the sample was moved in the horizontal plane by the motorized stage at a speed of 20 $\mu\text{m s}^{-1}$.

Analytical characterization

Precise source meters (Keithley 2636B and Agilent B1500A) were used to test the electrical and optoelectronic characteristics. The Agilent B1500 WGFPU pulse generator is used as the power source to generate input signals and measure the output spikes more quickly. Materialistic and morphological characterization was carried out by Raman and PL spectroscopy (alpha 300 R), Scanning Electronic Microscopy (SEM, JEOL-7001F-77), Transmission Electron Microscope (TEM, JEM-2100F), and adsorption spectrum (PE-L950). The thickness of materials were carried out by Atomic Force Microscope (AFM, Dimension, Bruker Inc.)

Acknowledgements

This work was supported by National Natural Science Foundation of China (Grant Nos. 52275346 and 52475370) and Beijing Natural Science Foundation (Grant No. L234032 and L248003).

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Author contributions

Zehua Li, Guisheng Zou, Jinpeng Huo, and Lei Liu conceived the ideas and designed the experiments. Zehua Li fabricated the dual-mode device and performed electrical and optical measurement. Jin Peng, Tianming Sun, and Yu Xiao helped with the Raman, AFM, SEM characterization and analysis. Jiali Huo



performed the SNN simulation. Bin Feng provided valuable suggestions for the manuscript and participated in the data analysis. Zehua Li, Jinpeng Huo, and Lei Liu wrote the paper, and all authors revised the paper.

Data availability

The data that supports the findings of this study are available from the corresponding author upon reasonable request.

Conflict of interest

The authors declare no competing interests.

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41377-026-02195-8>.

Received: 23 July 2025 Revised: 20 December 2025 Accepted: 13 January 2026

Published online: 06 March 2026

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