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## Article

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# Tunable Intermediate-Logic Ternary Circuits based on MoSe<sub>2</sub>-WSe<sub>2</sub> Heterojunction

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## Abstract

Van der Waals (vdW) heterojunctions, which consist of p-type and n-type semiconductors, have provided new features for transition metal dichalcogenides (TMDs). In this work, a negative differential transconductance (NDT) transistor based on a MoSe<sub>2</sub>-WSe<sub>2</sub> heterojunction (MoSe<sub>2</sub>-WSe<sub>2</sub> H-TR) is proposed. The MoSe<sub>2</sub>-WSe<sub>2</sub> H-TR provides desirable device characteristics for ternary circuit operation with a switching behavior of off-state / p-type turn-on / NDT region / p-type turn-on. As a result, a 100% output voltage ( $V_{OUT}$ ) swing inverter can be achieved in a ternary inverter, which consists of the proposed MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR and a MoS<sub>2</sub> floating-gate transistor. Furthermore, a tunable intermediate-logic ternary circuit operation is demonstrated by controlling the threshold voltage ( $V_{TH}$ ) in a pull-down n-type MoS<sub>2</sub> floating-gate transistor. We also investigated that a light-induced operation on the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR offers control of the  $V_{OUT}$  amplitude at the intermediate-logic state. Based on the proposed MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR, this work suggests a strategy to obtain a tunable ternary circuit, thus providing a new concept of heterojunction electronics using layered TMDs.

## Introduction

Two-dimensional (2D) materials, especially Transition metal dichalcogenides (TMDs) have recently attracted attention due to their great potential in various applications such as field-effect transistors and optoelectronics.<sup>1–10</sup> In addition to their intrinsic properties of the TMDs,<sup>11–14</sup> van der Waals heterostructures (vdW-H), which are constructed by stacking different TMDs materials, have also attracted significant attention in recent years. The ability to form different TMDs material combinations (*e.g.*, MoS<sub>2</sub>-WS<sub>2</sub>,<sup>15</sup> MoS<sub>2</sub>-WSe<sub>2</sub>,<sup>16</sup> MoS<sub>2</sub>-MoTe<sub>2</sub>,<sup>17</sup> and MoS<sub>2</sub>-BP<sup>18</sup>) offers the possibility of achieving new electronic characteristics. One of the emerging electronic functions arising from the vdW-H is a new switching behavior, which includes negative differential resistance (NDR) or negative differential transconductance (NDT) characteristics.<sup>19–21</sup>

This new switching characteristic does not behave monotonically, *i.e.*, the current does not continuously increase as the applied voltage increases. There is NDT regions, where the current decreases when the applied voltage is in the middle of its range. Due to this unique switching behavior, a new circuit scheme for ternary logic has been proposed. In the complementary circuit consisting of two serially connected transistors, where one transistor is a vdW-H transistor with NDT, the charging pull-up transistor has a comparable resistance region with the discharging pull-down transistor.<sup>22,23</sup> As a result, the vdW-H-based circuit offers an additional output voltage ( $V_{OUT}$ ) state at which both transistors have a comparable resistance. Thus, ternary logic operation can be achieved. As the ternary circuit has an extra logic state, more efficient logic computing is potentially provided.

However, the transition from binary to ternary logic circuit results in the reduction of the circuit's noise margin. Specifically, the maximum noise margin of a ternary logic circuit is 33% of the  $V_{DD}$  ( $V_{DD}$  is divided by three logic states), while the maximum noise margin of a binary logic circuit is 50% of the  $V_{DD}$  ( $V_{DD}$  is divided by two logic states).<sup>24</sup> As a result, it is

very important to accurately control each logic state margin circuit as the number of logic level states increases. However, previously reported vdW-H-based ternary circuits include: (1) incomplete output voltage swing<sup>25–27</sup> and (2) narrow margin for the intermediate-logic state.<sup>28–30</sup> These issues must be addressed to make the ternary circuit suitable to practical applications. Therefore, it is necessary to investigate how to implement vdW-H-based logic more systematically.

Here, we report a new combination of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H, providing desirable electrical properties for ternary circuit operation. As MoSe<sub>2</sub> and WSe<sub>2</sub> have n-type dominant and p-type dominant ambipolar charge transport characteristics, respectively, the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H provides desirable switching behavior as follows: off-state / p-type turn-on / NDT region / p-type turn-on. Using a MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H, a 100% swing ternary inverter circuit with tunable intermediate-logic state combined with MoS<sub>2</sub> flash memory is demonstrated. The proposed ternary inverter enables its intermediate-logic state to be adjustable by controlling  $V_{TH}$  of the pull-down MoS<sub>2</sub> floating-gate transistor. Furthermore, the intermediate  $V_{OUT}$  state can be controlled by light irradiation to the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H in the ternary inverter. As the excess current generated by the light irradiation increases  $I_D$  in the NDT region, the intermediate  $V_{OUT}$  state is enhanced. Through this study, it is noted that the state and margin of the  $V_{OUT}$  state can be adjusted. This opens up new opportunities for a new concept TMDs-based vdW-H electronics.

## Results and Discussion

First, the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H was fabricated using a polydimethylsiloxane (PDMS) stamp method.<sup>31</sup> WSe<sub>2</sub> was deposited on the top of MoSe<sub>2</sub> to form a partial overlap in the middle of the channel between the source and drain electrode. As a result, the one contact electrode makes contact with the MoSe<sub>2</sub> only, while the other contact electrode makes contact with the WSe<sub>2</sub> only (Figure 1a and 1b). Details of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H fabrication steps are given in the Experimental Section.

To investigate the chemical and morphological structure of the fabricated MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H, a comprehensive analysis including scanning electron microscopy (SEM), Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), and atomic force microscopy (AFM) was performed. Figure 1c shows the SEM and EDS mapping images of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H, indicating the coexistence of MoSe<sub>2</sub> and WSe<sub>2</sub> region. The measured Raman spectra show the peaks of WSe<sub>2</sub> (Red curve) at 246.3 and 253.8 cm<sup>-1</sup> and the peak of MoSe<sub>2</sub> (blue curve) at 239.9 cm<sup>-1</sup>.<sup>32-34</sup> The details of the Raman spectroscopy are explained in the Supporting Information (Figure S1). The coexistence of MoSe<sub>2</sub> and WSe<sub>2</sub> is also supported by the binding energy peaks of Mo 3d, W 4f, and Se 3d in the measured X-ray photoelectron spectroscopy (XPS) spectra (Figure S2).<sup>35-37</sup> The measured AFM shows the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H formation structure in the middle of the transistor channel region (Figure S3).

We investigated the electrical characteristics of the fabricated MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR. We prepared a baseline MoSe<sub>2</sub> and WSe<sub>2</sub> transistors were prepared as baseline n-type and p-type devices, respectively (Figure S4). The measured transfer characteristics of the baseline MoSe<sub>2</sub> transistor exhibit n-type dominant ambipolar characteristics with an effective electron and effective hole mobility (Figure 1d and Table S1). In contrast, the baseline WSe<sub>2</sub> transistor exhibits p-type dominant ambipolar characteristics with an effective electron and effective hole mobility (Figure 1d and Table S1). These n-type dominant and p-type dominant ambipolar

characteristics are also supported by the measured output curves, which exhibit a linear current increase due to counterflow (*i.e.*, hole current in the n-type operation and electron current in the p-type operation) (Figure S5 and S6). A total of 40 baseline transistors were fabricated to confirm the uniformity and reliability of the MoSe<sub>2</sub> transistor exhibiting n-type dominant ambipolar characteristics and the WSe<sub>2</sub> transistor exhibiting p-type dominant ambipolar characteristics. The 20 baseline MoSe<sub>2</sub> and 20 baseline WSe<sub>2</sub> transistors were fabricated with a similar thickness to the sample used for MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR, and their electrical properties ( $V_{TH}$ ,  $I_{ON}$ , and  $I_{OFF}$ ) were observed similarly for the most of them (Figure S7-S9).

Based on the baseline MoSe<sub>2</sub> and WSe<sub>2</sub> transistors, the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR was characterized. It was observed that the device does not switch on monotonically as a function of the applied gate voltage bias ( $V_G$ ). Instead, the device exhibits an abnormal switching behavior as follows: off-state / p-type turn-on / NDT region / p-type turn-on (Figure 1e). Similar behaviors are also observed in the other MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TRs (Figure S10). In the  $V_G = 40$  V to  $-7$  V range, the WSe<sub>2</sub> p-channel begins to accumulate hole carriers, contributing to  $I_D$  increase. In about the middle of the range ( $V_G = -7$  V), both MoSe<sub>2</sub> and WSe<sub>2</sub> accumulate hole and electron carriers, contributing to  $I_D$  increase. Accordingly, the peak of  $I_D$  (the highest  $I_D$  value) was observed in this voltage range. In the  $-47$  V  $< V_G < -7$  V range,  $I_D$  decreases, since the MoSe<sub>2</sub> *n*-channel becomes depleted by the applied negative bias  $V_G$ . As the negative  $V_G$  increases further, the turn-on of the MoSe<sub>2</sub> *p*-channel contributes to  $I_D$  increase due to the ambipolar characteristics of the MoSe<sub>2</sub>. It is worth noting that  $I_D$  increases again, when further negative bias is applied ( $V_G < -47$  V). This is because the MoSe<sub>2</sub> contributes to electron carrier accumulation due to its ambipolar charge transport properties (Figure 1d, Figure S5). It can be observed that the decrease of  $I_D$  (NDT) depends on the applied drain voltage bias ( $V_D$ ) (Figure 1e and 1f). More negative  $V_D$  (*e.g.*,  $V_D = -40$  V) results in a larger electron accumulation in the MoSe<sub>2</sub> region of the drain node. Thus, the peak current  $I_D$  increases by 9.2 times to 78  $\mu$ A, the

NDT voltage range widens by 2.1 times to 45 V, and the maximum NDT value increases by 6 times to  $-1.27$  S at  $V_D = -40$  V compared to the baseline values at  $V_D = -10$  V (Figure 1g–i). It is emphasized that the maximum NDT and voltage range values obtained in the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR are larger than in previously reported TMDs-based heterojunction transistors (Table S2).

Next, the ternary circuit was characterized using the proposed MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR. The proposed inverter circuit consists of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR and n-type MoS<sub>2</sub> floating-gate transistor in series (Figure 2a and 2b). The inverter circuit was fabricated by externally connecting the two transistors in different substrates. Details of the floating-gate transistor fabrication process and circuit measurement are given in Experimental Section. Unlike previously reported ternary inverters,<sup>23,26,38,39</sup> the floating-gate transistor is used as a pull-down transistor in this work. The MoS<sub>2</sub> transistor without the floating-gate layer was showed no hysteresis in the transfer characteristics curve (Figure S11). However, the MoS<sub>2</sub> floating-gate transistor was observed a memory window due to the charge traps in the floating gate.<sup>40,41</sup> As shown in Figure 2c, a memory window was originated from a large amount of charge stored in the PEDOT:PSS charge trap layer. When  $V_{Erase}$  is swept toward a positive bias, the electron carriers enter the PEDOT: PSS floating-gate through the TDL Al<sub>2</sub>O<sub>3</sub> layer by the mechanism of Fowler-Nordheim tunneling (FNT)<sup>42</sup> and trap-assisted tunneling (TAT)<sup>43</sup>, resulting in a positive shift of  $V_{TH}$ . However, when  $V_{Program}$  is swept from the positive to the negative direction, the trapped electrons pass through the energy barrier inside the floating gate, causing a negative shift of  $V_{TH}$ . Since the MoS<sub>2</sub> floating-gate transistor provides a controllable threshold voltage ( $V_{TH}$ ), which depends on the applied programming/erasing voltage bias, tunable inverter characteristics can be obtained. The fabricated MoS<sub>2</sub> floating-gate transistor exhibits a shift of  $V_{TH}$  from  $-20$  V to  $-47$  V depending on the applied erasing voltage bias

( $V_{Erase} = -20, -30, -40, -50$  V) (Figure 2c and 2d). Details of the MoS<sub>2</sub> floating-gate transistor fabrication process and structure are given in Figure S12.

Depending on the programmed/erased states ( $V_{Programmed} = 30$  V,  $V_{Erase} = -20$  V), the proposed inverter exhibits a transition from binary switching to ternary switching (Figure 2e and 2f). When  $V_{TH}$  shifts toward positive values (after  $V_{Programmed} = 30$  V), there is no region where pull-up and pull-down resistances can be matched as shown in Figure S13. However, as  $V_{TH}$  shifts toward negative values, the inverter has an intermediate-logic state, where the pull-up and pull-down resistances are matched, resulting in three distinct  $V_{OUT}$  states: Logic 1 at  $V_{OUT} = 20$  V, Logic 0.5 at  $V_{OUT} = 3$  V, Logic 0 at  $V_{OUT} = 0$  V. We emphasize that the proposed inverter combining with the  $V_{TH}$ -controllable floating-gate transistor enables its circuit operation to have binary and ternary logic behavior selectively. While few previous reports control the state of the pull-up transistor though  $V_{DD}$  or light irradiation to convert from binary to ternary,<sup>18, 24, 26, 44-46</sup> we believe that this is the first demonstration of selective circuit operation between binary and ternary logic by using the state of the pull-down n-type MoS<sub>2</sub> floating gate transistor.

The proposed inverter exhibits a stable operation with the appropriate ternary logic states ( $V_{OUT} = 20, 5$ , and 0 V) in the three-input pulsed measurement ( $V_{IN} = 20, 0$ , and  $-20$  V) for  $V_{DD} = 20$  V (Figure 2g and 2h).  $V_{IN}$ - $V_{OUT}$  transfer characteristics were also measured for different  $V_{DD}$  values ( $V_{DD} = 10, 20, 30, 40$ , and  $50$  V). The measured  $V_{OUT}$  value at the intermediate-logic state increases from 0.6 to 6.7 V as the applied  $V_{DD}$  (Figure 2i and 2j) increases. It must be emphasized that the proposed inverter exhibits 100%  $V_{OUT}$  swing with  $V_{DD}$ -to-GND (Figure 2k). Since the proposed MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR has a well-defined on-state (as shown in Figure 1d), the inverter performs a full pull-up operation, providing 100%  $V_{OUT}$  swing. Furthermore, it is demonstrated that the intermediate-logic region can be controlled by controlling the  $V_{TH}$  shift in the MoS<sub>2</sub> floating-gate transistor. When a larger  $V_{Erase}$

is applied,  $V_{TH}$  shifts to negative values. Therefore, the inverter has a wider region where the pull-up and pull-down resistances are matched (*i.e.*, the intermediate-logic voltage region). As shown in Figure 2l and 2m, the intermediate-logic voltage region increases by 1.7 times from 19 V to 33 V as the applied  $V_{Erase}$  decreases from -25 V to -50 V.

We investigated optoelectronic properties of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H. As a pre-step, the transfer characteristics of the baseline MoSe<sub>2</sub> and WSe<sub>2</sub> transistors as well as the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H under light illumination and  $V_D = -20$  V (Figure 3a-3c) were measured. These characteristics were measured as a function of the applied  $V_G$  at a fixed excitation wavelength ( $\lambda_{ex}$ ) of 638 nm for different incident power densities ( $P_{inc}$ ). In the measurement of the baseline MoSe<sub>2</sub> and WSe<sub>2</sub> transistors, an  $I_D$  increase was observed due to photocurrent generation as the applied  $P_{inc}$  increased. Furthermore, we extracted a photosensitivity, which is defined by the ratio of the photocurrent to dark current ( $S_{ph} = \frac{I_{ph}}{I_{dark}}$ ) (signal-to-noise ratio).<sup>47-49</sup> Figures 3d and 3e show the  $S_{ph}$  of the baseline WSe<sub>2</sub> transistor and baseline MoSe<sub>2</sub> transistor, indicating a peak value  $S_{ph}$  at the off-state region in both of the WSe<sub>2</sub> and MoSe<sub>2</sub> transistors. In the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR, the two peaks of  $S_{ph}$  were obtained (Figure 3f), resulting from a superposition form of photosensitivity peaks of the baseline MoSe<sub>2</sub> and WSe<sub>2</sub> transistors. Although the  $S_{ph}$  of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR is similar to the superposition form of the  $S_{ph}$  of the baseline device, the sensitivity values are different because the dark currents (*i.e.*, noise) of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR and baseline devices are different.

In the NDT region,  $I_D$  increases as the applied  $P_{inc}$  increases. This is shown in the plots of  $I_{peak}$  and  $I_{valley}$ , which depend on the illumination  $P_{inc}$  (Figure 3g and 3h). In the NDT region, an enhancement in  $I_D$  under illumination was observed. The NDT results from the depletion of the MoSe<sub>2</sub> channel (as described in Figure 1) and the photocurrent generation in the MoSe<sub>2</sub> by the light illumination lead to an increase in  $I_D$  in this region of the NDT. The peak-to-valley

ratio and maximum negative  $g_m$  decrease as the applied  $P_{inc}$  increases. This is due to the increase in  $I_D$  in the NDT region by light illumination (Figure 3i and 3j). Additional measurements and their analysis of the photoresponsive WSe<sub>2</sub>, MoSe<sub>2</sub>, MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR are given in Figure S14-S16. Figure S14a and S14b shows the transfer curves of baseline WSe<sub>2</sub> and MoSe<sub>2</sub> transistors under illumination light. These characteristics were measured under the same conditions in Figure 3a and 3b except for  $V_D$  of -1 V, which are performed to compare photoresponsive behaviors of the baseline devices depending on  $V_D$  of -1 V and -20 V. Figure S14c, S14d, and Figure S15a, S15b show the extracted photoresponsivity ( $R_{ph}$ ) of the baseline WSe<sub>2</sub> and MoSe<sub>2</sub> transistors as a function of  $V_G$  under  $V_D$  of -1 V and -20 V, respectively.  $R_{ph}$  can be calculated using the following equation:

$$R_{ph} = \frac{I_{ph}}{P_{inc} \times A_{ch}} \text{ [AW}^{-1}]$$

Where,  $I_{ph}$  is the photocurrent ( $I_{ph} = I_{total} - I_{dark}$ ) and  $A_{ch}$  is the channel area of the device.<sup>50</sup> A maximum  $R_{ph}$  of 366 AW<sup>-1</sup> was obtained for the MoSe<sub>2</sub> transistor, which is 3 times larger than the  $R_{ph}$  of the WSe<sub>2</sub> transistor for  $P_{inc} = 0.4 \text{ mW cm}^{-2}$ . This indicates that the MoSe<sub>2</sub> transistor exhibits more light absorption and photocurrent generation than those of the WSe<sub>2</sub> transistor.

The optoelectronic properties of the proposed MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR (Figure S15c) were also characterized. The transfer characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR under illumination light at a fixed excitation wavelength ( $\lambda_{ex}$ ) of 638 nm and  $V_D = -20 \text{ V}$  were measured. These are the same measurement conditions as those applied in the baseline WSe<sub>2</sub> and MoSe<sub>2</sub> transistors characterizations. It was observed that a maximum  $R_{ph}$  of 40 AW<sup>-1</sup> was obtained in the middle of the  $V_G$  range ( $V_G = -20 \text{ V}$ ).  $R_{ph}$  of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H transistor shows the two peaks at  $V_G = -47 \text{ V}$  and -24 V. It is indicated that the  $R_{ph}$  peaks of MoSe<sub>2</sub>-WSe<sub>2</sub> are formed by superposition of  $R_{ph}$  of WSe<sub>2</sub> and MoSe<sub>2</sub> transistors. The  $R_{ph}$  peak at  $V_G = -47 \text{ V}$  is lower than the  $R_{ph}$  peak at  $V_G = -24 \text{ V}$  due to the  $R_{ph}$  of WSe<sub>2</sub> lower than that of MoSe<sub>2</sub>. When  $V_G$  is  $V_G < -47 \text{ V}$ , the  $R_{ph}$  of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H transistor gradually decreased because  $R_{ph}$

of MoSe<sub>2</sub> in heterostructure converge to zero. Also, under -24 V < V<sub>G</sub>, the R<sub>ph</sub> of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H transistor gradually decreased because R<sub>ph</sub> of WSe<sub>2</sub> in heterostructure converge to zero. Further details of the photoresponsivity properties are given in Figure S16a. The specific detectivity (D\*), such as R<sub>ph</sub>, is the most representative figures of merit for the evaluation of the optical properties of semiconducting materials, D\* can be calculated using the following equation:  $D^* = \frac{\sqrt{A \times \Delta f}}{NEP} = \frac{R_{ph}\sqrt{A}}{S_n}$  (A: area, Δf: bandwidth, NEP: noise equivalent power, S<sub>n</sub>: noise spectral density). The calculated D\* of the WSe<sub>2</sub>, MoSe<sub>2</sub>, and MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR is shown in Figure S16b, which clearly indicates that the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR exhibits the highest value. This is because it is comparable regardless of the area of the devices.<sup>51</sup> Moreover, the higher D\* of the MoSe<sub>2</sub> transistor than that of the WSe<sub>2</sub> transistor demonstrates again that the MoSe<sub>2</sub> transistor exhibits superior optical properties than the WSe<sub>2</sub> transistor.

The I<sub>D</sub> increase in the NDT region by the observed photoelectric effect allowed the implementation of a controllable intermediate-logic V<sub>OUT</sub> ternary inverter based on the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR and the MoS<sub>2</sub> floating-gate transistors. The ternary inverter was measured by illuminating the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR at  $\lambda_{ex} = 638$  nm (Figure 4a). Figure 4b shows the V<sub>OUT</sub>-V<sub>IN</sub> characteristics of the ternary inverter with and without illumination light. The illuminated vdW-H pull-up transistor leads to a V<sub>OUT</sub> increase in the intermediate-logic of the ternary circuit. In the three-V<sub>IN</sub> pulsed measurements, the inverter showed that the intermediate-logic V<sub>OUT</sub> increases by 1.5 times to 7.2 V from 4.8 V as the illumination P<sub>inc</sub> increases (Figure 4c). In the NDT region, which determines the V<sub>OUT</sub> value of the intermediate-logic, the resistance ratio between the illuminated vdW-H pull-up transistor and the pull-down floating-gate transistor can be adjusted according to the illumination P<sub>inc</sub>. This results in the V<sub>OUT</sub> variation of the intermediate-logic state. In summary, we proposed a P-memory that combines a layered MoS<sub>2</sub> thin-film transistor structure with a PEDOT:PSS floating gate. Owing to the

virtues of the MoS<sub>2</sub> and PEDOT:PSS combined structure, the P-memory exhibited high performance including a high switching current ratio  $> 10^7$ , large memory window  $> 50$  V, excellent endurance  $> 1,000$  cycles, and long retention time  $> 2,000$  s. These were confirmed by multiple device fabrications and characterizations to evaluate their reproducibility and uniformity ( $>30$  devices). We also proposed a new performance metric, the photo programming responsivity, representing the amount of storage of optical information, which is determined by the light intensity exposed during the programming operation. We also achieved a photo-activated memory operation that combines photodiode and memory functions. Depending on whether the light was illuminated, the P-memory provided a different degree of programming, and thus the degree of light exposure could be stored directly in the device. Furthermore, we demonstrated a flexible P-memory by manufacturing the device on a flexible PI substrate. This indicated that the device maintained its multifunctional memory behaviors even after the 1,000 bending cycles. Through comprehensive experiments, this study showed not only the memory characteristics of the conventional floating-gate-based flash memory but also new versatility such as mechanical flexibility and photoactive memory behaviors. These reported results are expected to be useful guidelines in the development of new electronic device features using new nanomaterials.

In summary, tunable intermediate-logic ternary circuits based on the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H were demonstrated. By combining the illuminated MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H as the pull-up device and the MoS<sub>2</sub> flash memory as the pull-down device, tunable intermediate-logic ternary circuits were implemented for the first time. Specifically: (1) a tunable intermediate-logic  $V_{OUT}$  region and (2) a tunable intermediate-logic  $V_{OUT}$  amplitude were achieved. To the best of our knowledge, this is the first demonstration of the intermediate-logic tunability in both voltage region and amplitude (Table S3). In the transition from the conventional binary to multi-state circuit, the accurate control of  $V_{OUT}$  is essential. We believe that this work would provide

helpful guidelines for the development of ternary circuits using TMDs-based vdW-H technology.

## Methods

*Device Fabrication:* The MoSe<sub>2</sub> and WSe<sub>2</sub> layers were mechanically exfoliated and transferred onto Si/SiO<sub>2</sub> substrate from bulk crystals using polydimethylsiloxane (PDMS) stamp method. To fabricate the p-n heterojunction consisting of WSe<sub>2</sub> (p-type) and MoSe<sub>2</sub> (n-type), the solution of polypropylene carbonate (PPC) in chloroform was spin-coated on Si/SiO<sub>2</sub> substrate. The PDMS was attached on PPC coated WSe<sub>2</sub> flake, and then it was immersed in the DI water for 10 min. The PDMS/PPC/WSe<sub>2</sub> sample was loaded on the MoSe<sub>2</sub> flake on heavily p-doped Si wafer covered with 300 nm-thick SiO<sub>2</sub> substrate. The PDMS/PPC/WSe<sub>2</sub>/MoSe<sub>2</sub> sample was heated at 90 °C for 1 min. With cooling at room temperature, the PDMS was slowly disengaged, and the PPC film was removed by acetone cleaning. To fabricate the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H, the WSe<sub>2</sub> flake was partially overlapped with the form of head to tail hybrid on the surface of MoSe<sub>2</sub> flake. Next, Ti/Au (5/30 nm) as the source and drain (S/D) electrodes were deposited and patterned by e-beam evaporation and photolithography techniques. The length ( $L_{ch}$ ) and width ( $W_{ch}$ ) of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H channel were 24.1 μm and 13.8 μm, respectively. Finally, MoSe<sub>2</sub>/WSe<sub>2</sub> vdW-H-TR were annealed 250 °C in H<sub>2</sub>/Ar condition to reduce the contact resistance between contact electrode and semiconductors.

In order to fabricate the MoS<sub>2</sub> floating gate transistor, PEDOT:PSS as the floating gate was spin-coated on Si/SiO<sub>2</sub> substrate at 4000 rpm for 1 min and annealed at 150 °C for 15 min. 80 nm-thick Al<sub>2</sub>O<sub>3</sub> was then deposited by atomic layered deposition (ALD) at 200 °C for 4 h. After mechanically exfoliated MoS<sub>2</sub> were transferred onto Al<sub>2</sub>O<sub>3</sub> floating gate insulator, Ti/Au (20/100 nm) were deposited by e-beam evaporator system. Then, S/D electrodes were patterned by standard photolithography and etching techniques.  $L_{ch}$  and  $W_{ch}$  of the MoS<sub>2</sub> channel in floating-gate transistor is 9.4 and 10.2 μm, respectively. Finally, 20 nm-thick Al<sub>2</sub>O<sub>3</sub> passivation

layer on fabricated the device were deposited by ALD to prevent the effects of O<sub>2</sub> and H<sub>2</sub>O. To implement the inverter circuit, we externally connected the drain and gate nodes of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H and the MoS<sub>2</sub> floating-gate transistors, respectively.

*Characterization:* A laser confocal microscope (LCM) Raman was conducted with a He-Ar laser ( $\lambda_{ex} = 532$  nm). The spot diameter of the focused laser beam in the LCM system was approximately 1  $\mu\text{m}$ . AFM image and thickness of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR were measured by an atomic force microscope (XE7, Park Systems) with non-contact mode. SEM images and EDS mapping of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR were obtained by a secondary electron emission from the sample with a 15.0 kV acceleration voltage on top view. The electrical characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR were measured using a keithley 4200-SCS semiconductor characterization system connected with a probe station at room temperature in the air under dark condition. The photoresponsive behaviors of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR were investigated by a multichannel fiber-coupled laser source (MCLS1, Thorlabs) equipped with the single mode fiber optical patch cables (SM600).

### **Data availability**

All data during this project are included in this article and supplementary information.

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## **Acknowledgment**

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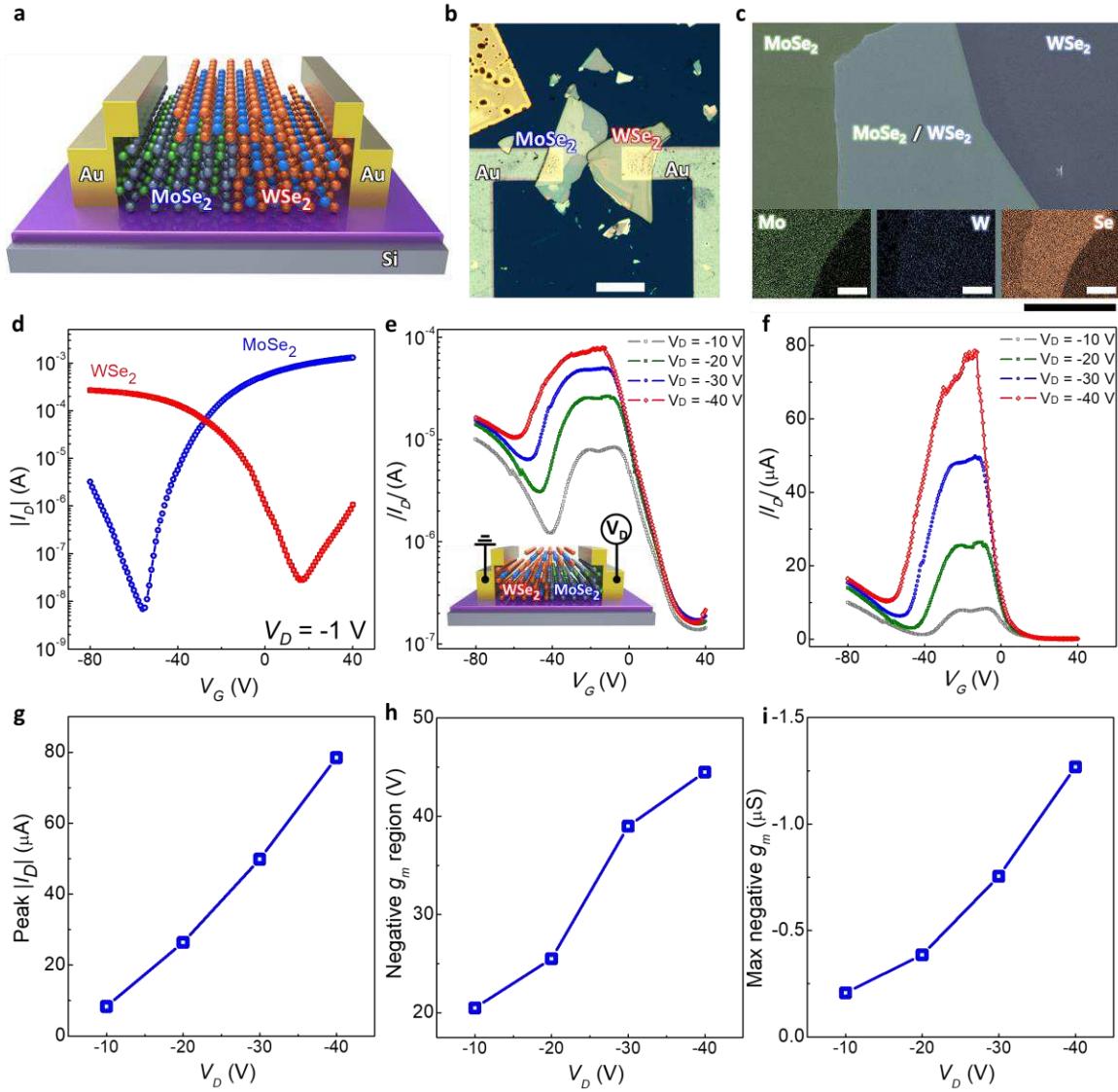
## **Author contributions**

S.H., H.J.P., and H.Y. performed all experiments. H.W.C. and J.P. contributed to the experiments. H.Y. and S.K. supervised the project. S.H., H.P., H.Y., and S.K. wrote the manuscript. All authors have given approval to the final version of the manuscript.

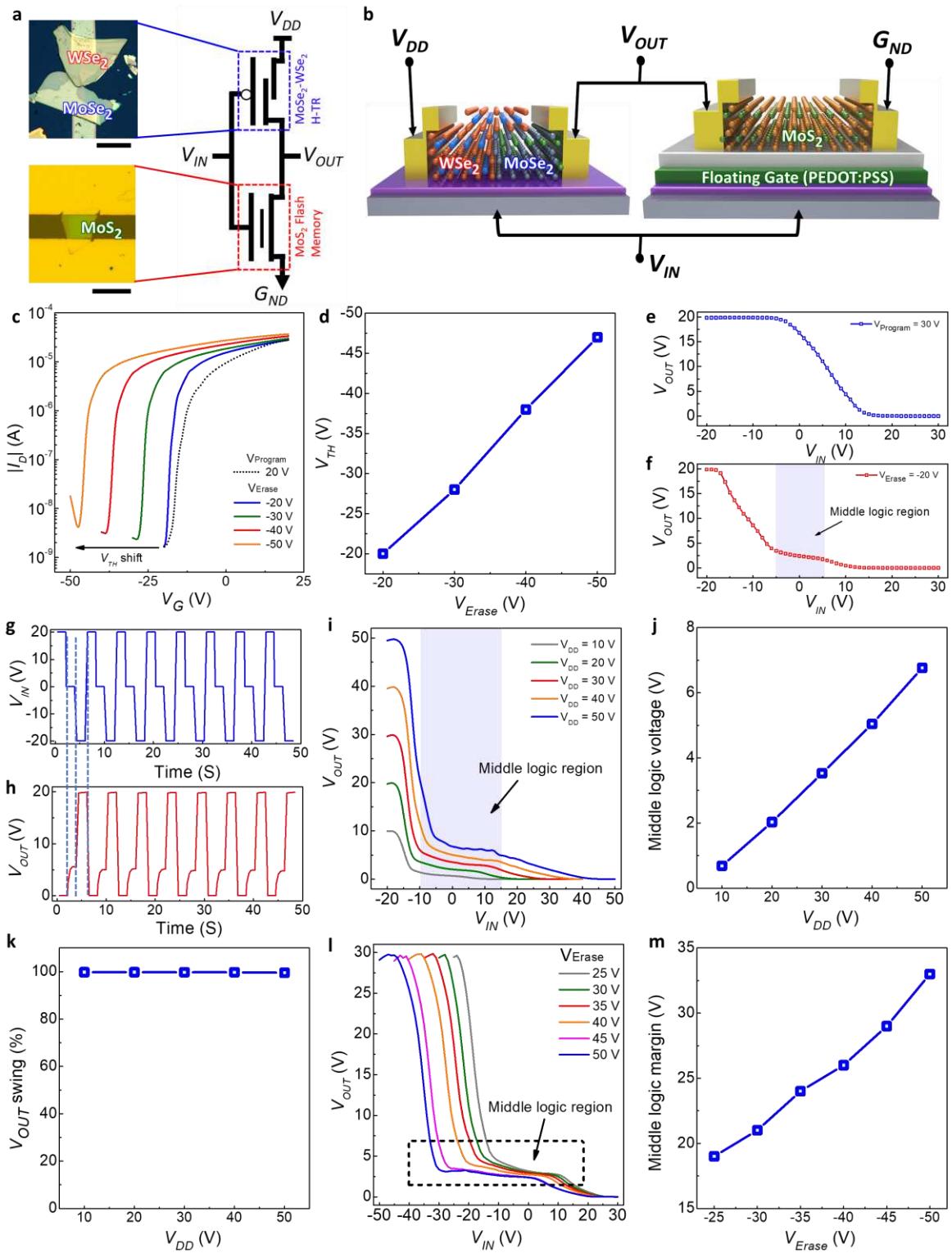
## **Competing interests**

The authors declare no competing financial interest.

## Figures and captions

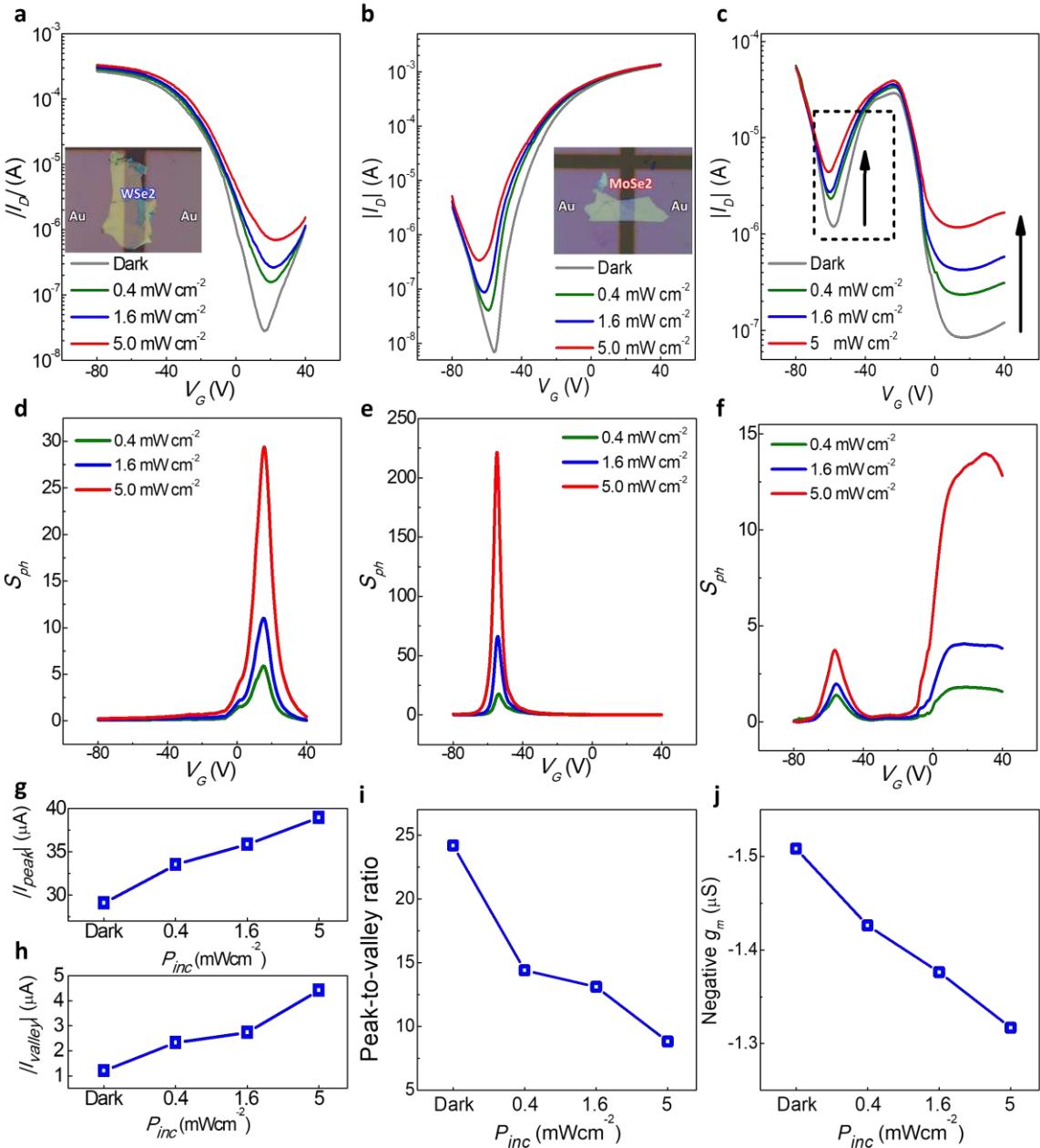


**Figure 1.** Investigation of the electrical characteristics of the MoSe<sub>2</sub>–WSe<sub>2</sub> vdW-H-TR. a) 3D Schematic structure of the MoSe<sub>2</sub>–WSe<sub>2</sub> vdW-H-TR. b) Optical microscopy image of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR. The scale bar is in 20  $\mu\text{m}$ . c) False-colored SEM image of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H. EDS mapping of the Mo (green dot), W (blue dot), and Se (orange dot) distributions for MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H. The length of the scale bar is 5  $\mu\text{m}$ . d) Transfer characteristics of the baseline MoSe<sub>2</sub> (blue curve) and baseline WSe<sub>2</sub> (red curve) transistors at  $V_D = -1$  V. e) Transfer characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR for  $V_D = -10, -20, -30, -40$  V. f) Transfer characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR for different values of  $V_D$  in linear scale. Plot of g) peak  $I_D$ , h) negative  $g_m$  region, and i) maximum negative  $g_m$  values as a function of  $V_D$  for the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR.

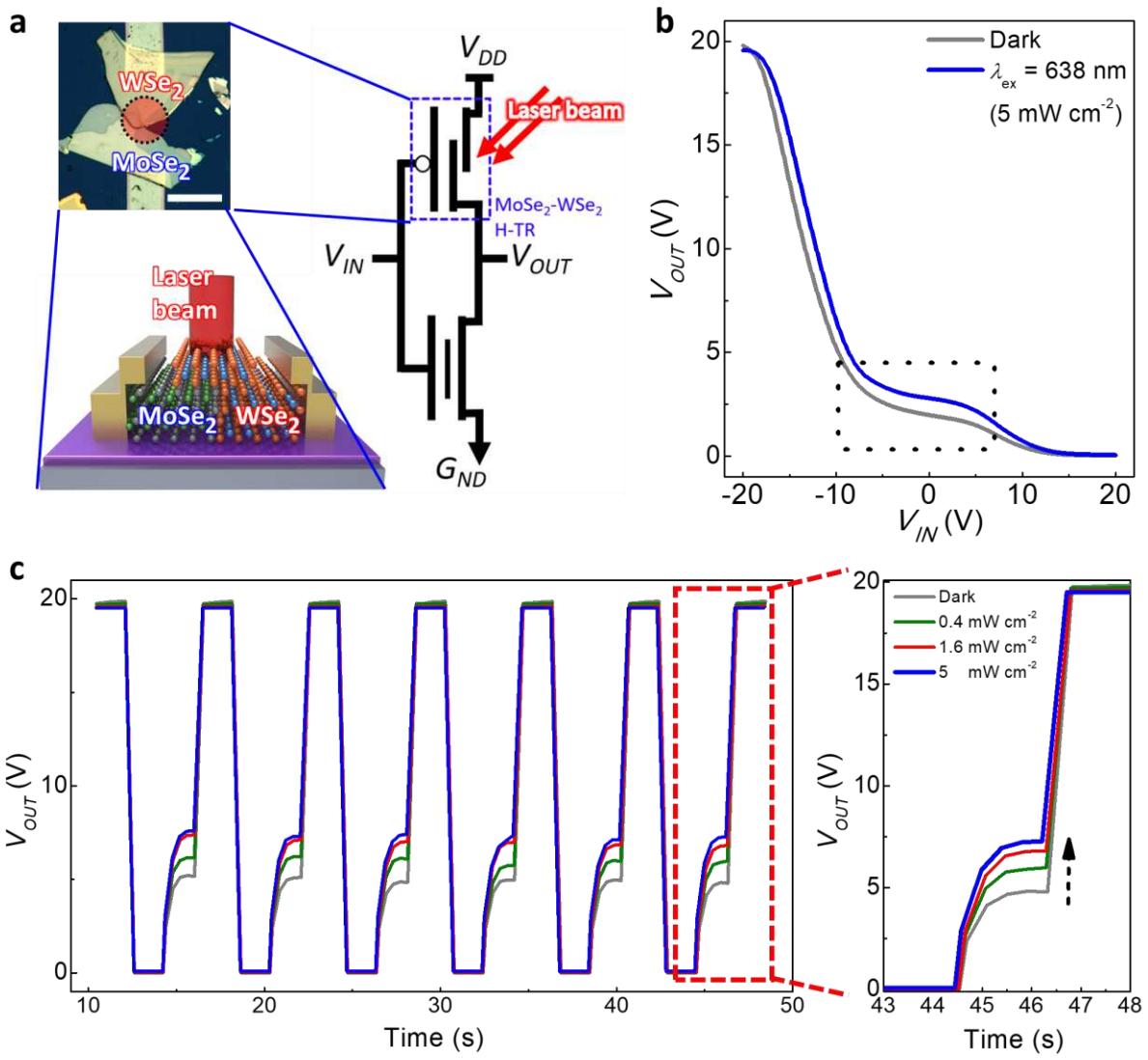


**Figure 2.** Ternary circuit using the proposed MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-TR and MoS<sub>2</sub> floating-gate transistor  
**a)** Schematic symbol of the ternary inverter using the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR and the n-type floating-gate transistor.  
**b)** 3D schematic structure of the ternary inverter.  
**c)** Transfer characteristics of the MoS<sub>2</sub> floating-gate transistor depending on the applied erasing voltage  
**d)** Threshold voltage shift versus erasing voltage.  
**e)** Output voltage versus input voltage for programming.  
**f)** Output voltage versus input voltage for erasing.  
**g)** Drain current versus time.  
**h)** Output voltage versus time.  
**i)** Output voltage versus input voltage for different supply voltages.  
**j)** Middle logic voltage versus supply voltage.  
**k)** Output swing percentage versus supply voltage.  
**l)** Output voltage versus input voltage for different erasing voltages.  
**m)** Middle logic margin versus erasing voltage.

bias ( $V_{Erase} = -20, -30, -40$ , and  $-50$  V). d)  $V_{TH}$  variation as a function of  $V_{Erase}$  for the MoS<sub>2</sub> floating-gate transistor. e) Measured binary inverter characteristics ( $V_{OUT} - V_{IN}$ ) for  $V_{Program} = 30$  V. f) Measured ternary inverter characteristics ( $V_{OUT} - V_{IN}$ ) for  $V_{Erase} = -20$  V. g, h) Transient measurement with respect to the  $V_{IN}$  ternary signal. i) Measured ternary inverter characteristics ( $V_{OUT} - V_{IN}$ ) for different  $V_{DD}$  values. j) Middle logic voltage as a function of  $V_{DD}$  for the ternary inverter. k) Plot showing 100%  $V_{OUT}$  swing with  $V_{DD}$ -to-GND. l) Measured ternary inverter characteristics ( $V_{OUT} - V_{IN}$ ) for different  $V_{Erase}$  values. m) Middle logic margin as a function of  $V_{Erase}$  for the ternary inverter.

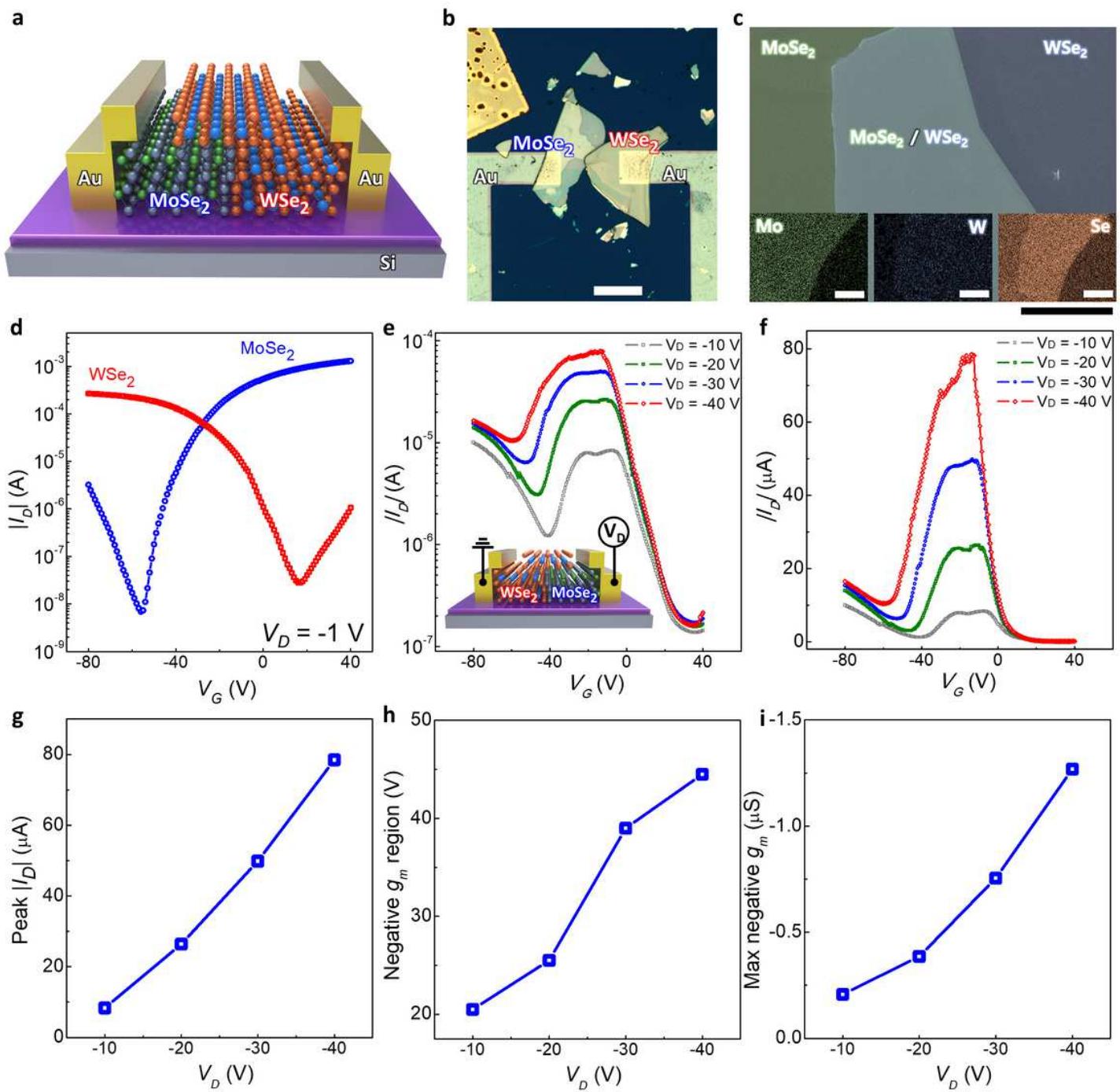


**Figure 3.** Investigation of the electrical characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR under dark and light illumination ( $\lambda_{ex} = 638$  nm) conditions. Transfer characteristics of a) baseline WSe<sub>2</sub> transistor b) baseline MoSe<sub>2</sub> transistor c) MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR for different incident power densities ( $P_{inc}$ ). Photosensitivity ( $S_{ph}$ ) of d) baseline WSe<sub>2</sub>, e) baseline MoSe<sub>2</sub>, and f) MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H as a function of  $V_G$ . Plot of g) peak  $I_D$ , h) valley  $I_D$ , i) peak-to-valley  $I_D$  ratio j) negative transconductance values ( $g_m$ ) as a function of  $P_{inc}$  for the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR.



**Figure 4.** Transient measurement of the ternary inverter under dark and light illumination ( $\lambda_{\text{ex}} = 638 \text{ nm}$ ) conditions. a) Schematic symbol of the ternary inverter using MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR with light illumination and n-type floating-gate transistor without illumination. b) Measured ternary inverter characteristics ( $V_{\text{OUT}} - V_{\text{IN}}$ ) under dark and light illumination conditions ( $\lambda_{\text{ex}} = 638 \text{ nm}$ ,  $P_{\text{inc}} = 5 \text{ mW cm}^{-2}$ ). c) Transient measurement with respect to the  $V_{\text{IN}}$  ternary signal for different incident power densities ( $P_{\text{inc}}$ ).

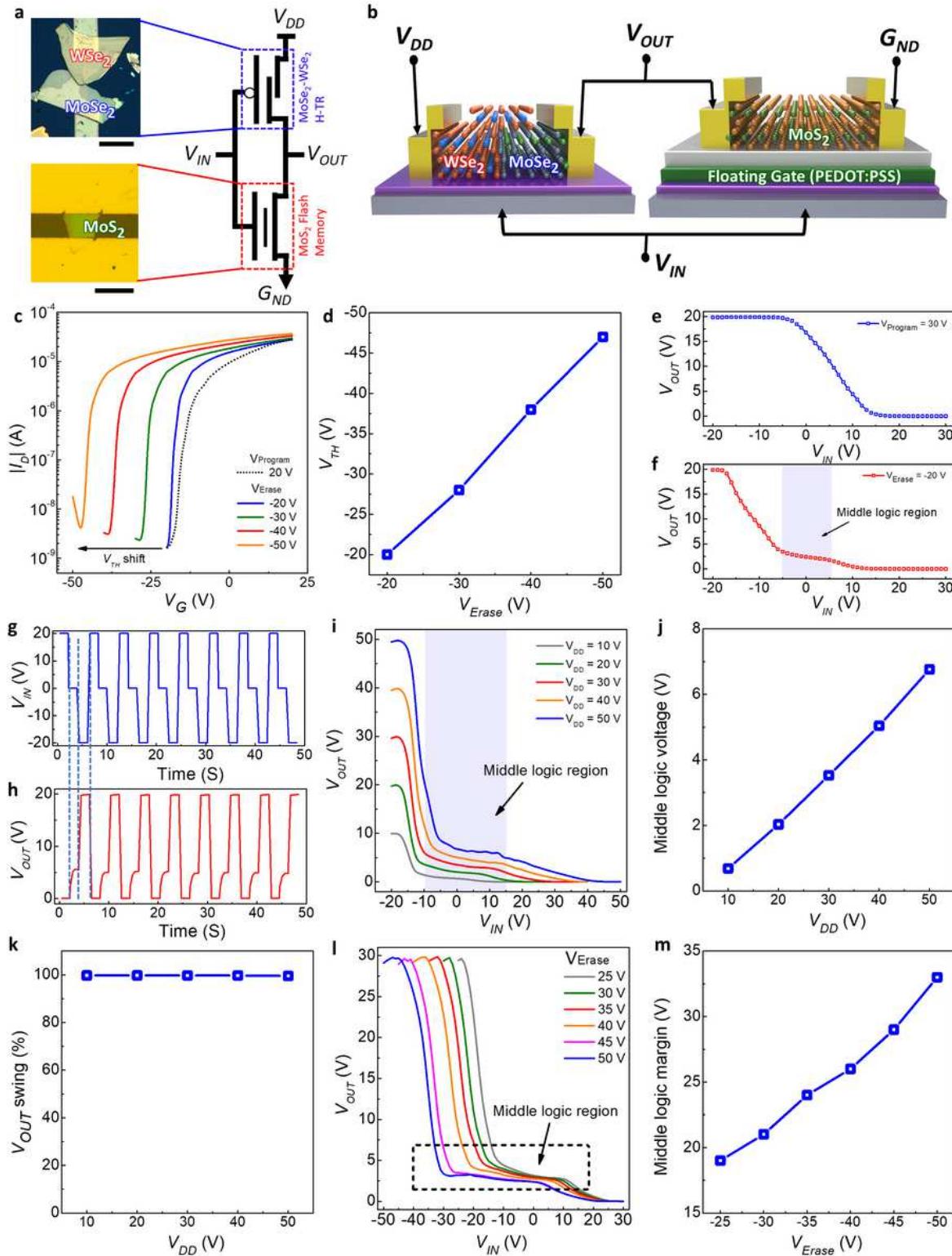
# Figures



**Figure 1**

Investigation of the electrical characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR. a) 3D Schematic structure of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR. b) Optical microscopy image of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR. The scale bar is in 20  $\mu$ m. c) False-colored SEM image of MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H. EDS mapping of the Mo (green dot), W (blue dot), and Se (orange dot) distributions for MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H. The length of the scale bar is 5  $\mu$ m. d) Transfer characteristics of the baseline MoSe<sub>2</sub> (blue curve) and baseline WSe<sub>2</sub> (red curve) transistors at  $V_D = -1$  V. e) Transfer characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR for  $V_D = -10$ ,  $-20$ ,  $-30$ , and  $-40$  V. f) Transfer characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR for  $V_D = -10$ ,  $-20$ ,  $-30$ , and  $-40$  V. g) Peak  $|I_D|$  ( $\mu$ A) as a function of  $V_D$  (V) for the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR. h) Negative  $g_m$  region (V) as a function of  $V_D$  (V) for the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR. i) Max negative  $g_m$  ( $\mu$ S) as a function of  $V_D$  (V) for the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR.

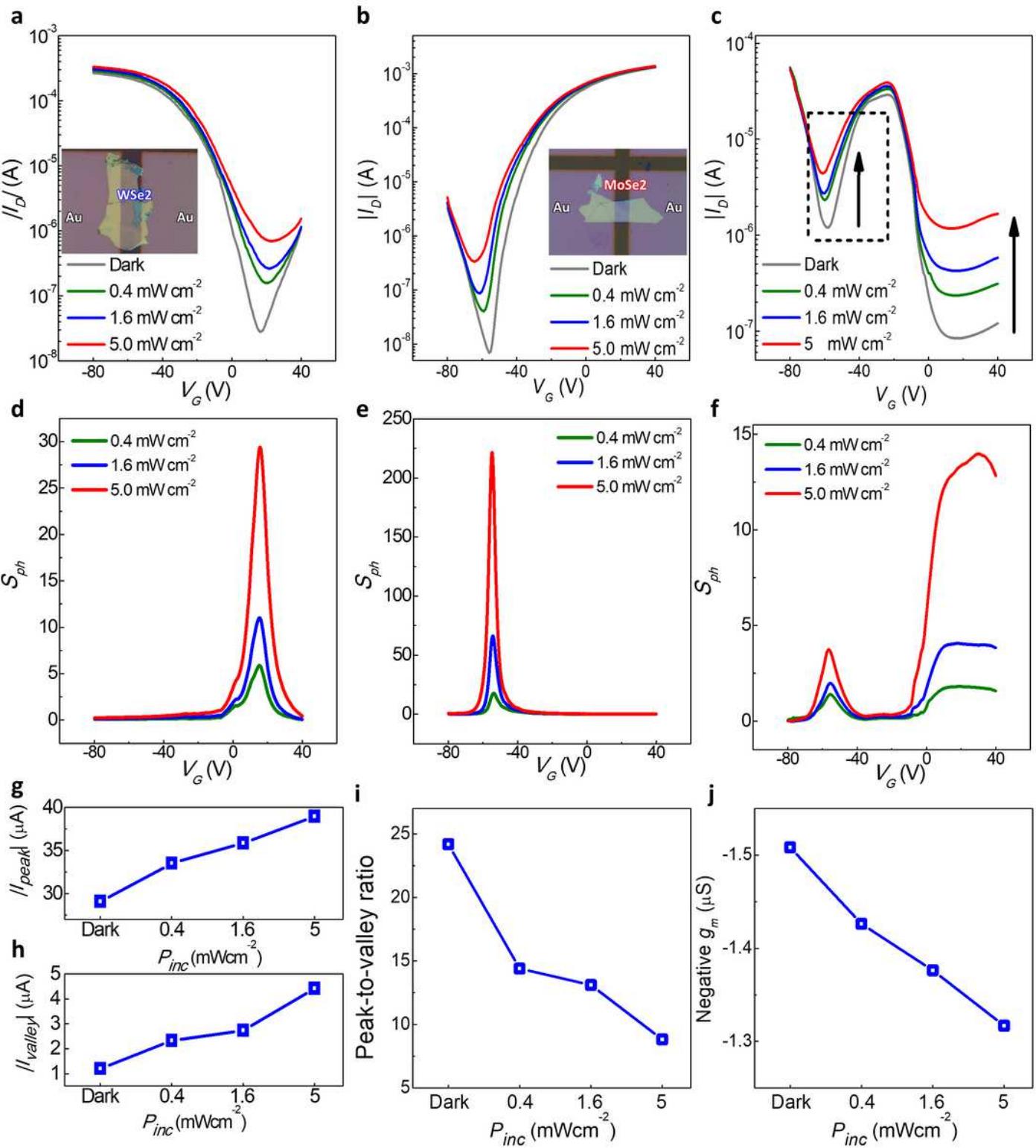
-40 V. f) Transfer characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR for different values of VD in linear scale. Plot of g) peak ID, h) negative gm region, and i) maximum negative gm values as a function of VD for the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR.



**Figure 2**

Ternary circuit using the proposed MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR and MoS<sub>2</sub> floating-gate transistor a) Schematic symbol of the ternary inverter using the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR and the n-type floating-gate transistor.

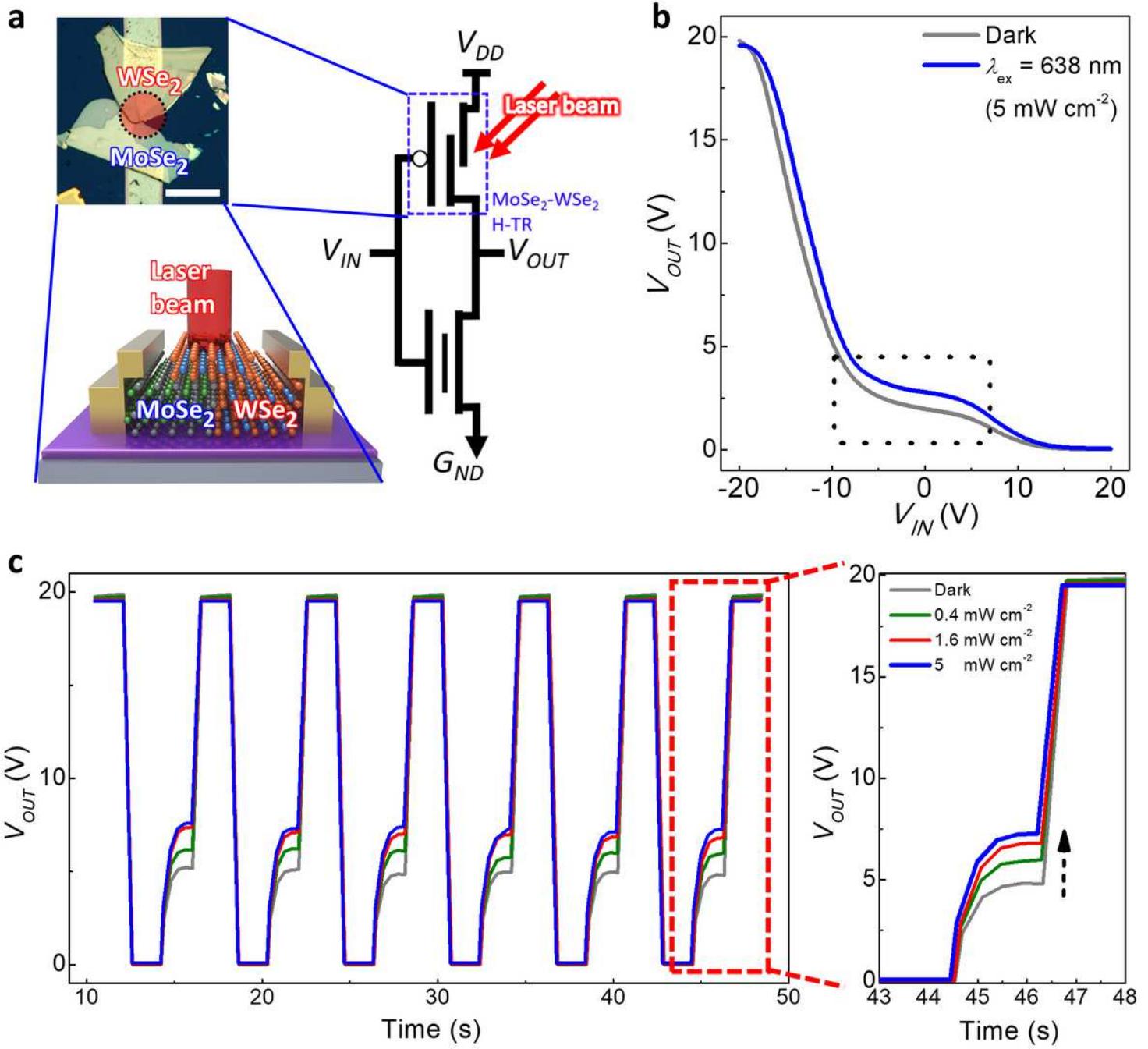
b) 3D schematic structure of the ternary inverter. c) Transfer characteristics of the MoS<sub>2</sub> floating-gate transistor depending on the applied erasing voltage bias ( $V_{Erase} = -20, -30, -40, \text{ and } -50 \text{ V}$ ). d)  $V_{TH}$  variation as a function of  $V_{Erase}$  for the MoS<sub>2</sub> floating-gate transistor. e) Measured binary inverter characteristics ( $V_{OUT} - V_{IN}$ ) for  $V_{Program} = 30 \text{ V}$ . f) Measured ternary inverter characteristics ( $V_{OUT} - V_{IN}$ ) for  $V_{Erase} = -20 \text{ V}$ . g, h) Transient measurement with respect to the  $V_{IN}$  ternary signal. i) Measured ternary inverter characteristics ( $V_{OUT} - V_{IN}$ ) for different  $V_{DD}$  values. j) Middle logic voltage as a function of  $V_{DD}$  for the ternary inverter. k) Plot showing 100%  $V_{OUT}$  swing with  $V_{DD}$ -to-GND. l) Measured ternary inverter characteristics ( $V_{OUT} - V_{IN}$ ) for different  $V_{Erase}$  values. m) Middle logic margin as a function of  $V_{Erase}$  for the ternary inverter.



**Figure 3**

Investigation of the electrical characteristics of the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR under dark and light illumination ( $\lambda_{ex} = 638$  nm) conditions. Transfer characteristics of a) baseline WSe<sub>2</sub> transistor b) baseline MoSe<sub>2</sub> transistor c) MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR for different incident power densities ( $P_{inc}$ ). Photosensitivity ( $S_{ph}$ ) of d) baseline WSe<sub>2</sub>, e) baseline MoSe<sub>2</sub>, and f) MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H as a function

of VG. Plot of g) peak ID, h) valley ID, i) peak-to-valley ID ratio j) negative transconductance values ( $g_m$ ) as a function of  $P_{inc}$  for the MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR.



**Figure 4**

Transient measurement of the ternary inverter under dark and light illumination ( $\lambda_{ex} = 638$  nm) conditions. a) Schematic symbol of the ternary inverter using MoSe<sub>2</sub>-WSe<sub>2</sub> vdW-H-TR with light illumination and n-type floating-gate transistor without illumination. b) Measured ternary inverter characteristics ( $V_{OUT}$  –  $V_{IN}$ ) under dark and light illumination conditions ( $\lambda_{ex} = 638$  nm,  $P_{inc} = 5 \text{ mW cm}^{-2}$ ). c) Transient measurement with respect to the  $V_{IN}$  ternary signal for different incident power densities ( $P_{inc}$ ).

## Supplementary Files

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