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**Floating Gate Negative Capacitance MoS₂ Phototransistor
with High Photosensitivity**

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1 **Title** Floating Gate Negative Capacitance MoS₂ Phototransistor with High
2 Photosensitivity

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16 Keywords: MoS₂, Ferroelectric, Phototransistor, Photogating, Floating gate

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18
19 Monolayer MoS₂ exhibits interesting optoelectronic properties that have been utilized in
20 applications such as photodetectors and light emitting diodes. For image sensing applications,
21 improving the light sensitivity relies on achieving a low dark current that enables the
22 detection weak light signals. Although previous reports to improve the detectivity have been
23 explored with heterostructures and pn junction devices, some these approaches lack CMOS
24 compatibility processing and not sufficiently low dark current suppression. Steep slope
25 transistors that overcome the Boltzmann tyranny can further enhance the performance in
26 photodetectors by providing efficient extraction of photogenerated charges. Here, we report a
27 monolayer MoS₂ floating gate negative capacitance phototransistor with the integration of a
28 hafnium-zirconium oxide ferroelectric capacitor. In this study, a SS_{min} of 30 mV/dec, very
29 low dark currents 10⁻¹³~10⁻¹⁴A, and a high detectivity of 7.2x10¹⁵ cm Hz^{1/2} W⁻¹ was achieved
30 under weak light illumination due to an enhancement in the photogating effect. In addition, its
31 potential as an optical memory and as an optical synapse with excellent long-term potentiation
32 characteristics in an artificial neural network were also explored. Overall, this device structure
33 offers high photosensitivity of weak light signals for future low-powered optoelectronic
34 applications.

35

36 **1. Introduction**

37 Photodetectors are used in various applications such as image sensing, surveillance, and
 38 biomedical imaging^[1,2]. Research efforts have been focused towards exploring materials and
 39 device structures to improve the incident light absorption and conversion efficiency into
 40 current. 2D materials are particularly promising due to their interesting optoelectronic
 41 properties^[38,39,40] and thus have been applied to phototransistors where the built-in electric
 42 field generated can enable efficient photocarrier extraction. An n-type and p-type MoS₂
 43 phototransistor based on a PN junction achieved a high responsivity of 7x10⁴ A/W and a
 44 detectivity of 3.5x10¹⁴ Jones^[3]. Modifying the Schottky barrier in graphene/MoS₂
 45 heterojunctions was explored where a high responsivity and detectivity of 2.2x10⁵ A/W and
 46 3.5x10¹³ Jones respectively was demonstrated^[4]. Pb quantum dots integrated with MoS₂ have
 47 also been demonstrated to enhance the photosensitivity via interface engineering to improve
 48 the I_{on}/I_{off} ratio^[5]. Further improvement in the detectivity can enable the detection of weak
 49 light signals that could not be detected leading to better imaging resolution for applications
 50 like facial recognition and surveillance systems. In addition, for the practical integration of
 51 highly photosensitive detectors in imaging sensing technologies, it is important to have a
 52 complementary-metal-oxide-semiconductor (CMOS) compatible processing and low power
 53 operation.

54

55 Steep slope transistor device structures that overcome the Boltzmann tyranny have a
 56 promising potential in photodetection applications. The subthreshold swing (SS) is defined as
 57 the amount of gate voltage (V_G) required to increase the drain current (I_{DS}) by one decade.

$$58 \quad SS \equiv \frac{dV_G}{d\log_{10}[I_{DS}]} = \left[\frac{dV_G}{d\phi_s} \right] \left[\frac{d\phi_s}{d\log_{10}[I_{DS}]} \right] = \left[1 + \frac{C_s}{C} \right] \left[\frac{k_T}{q} \ln(10) \right] = m \times n, \text{ where } C_s \text{ is the}$$

59 semiconductor depletion capacitance, C is the gate insulator capacitance, and ϕ_s is the

60 channel's surface potential. The limiting factor in scaling SS in conventional field effect
61 transistors (FET) comes from the carrier transport (n) term. Here, the SS can be scaled down
62 to a minimum of 60 mV/dec due to the Boltzmann factor (kT/q) at room temperature
63 conditions. There are two approaches to reduce the SS which is either modifying the carrier
64 transport (n) in the channel region like tunneling FETs or by changing the gate capacitance
65 (m). Ferroelectrics are nonlinear insulators that have two remnant polarization states that can
66 be retained without the application of an applied electric field. Negative capacitance FETs
67 using ferroelectric insulators can reduce the body factor (m) to values less than 1 offering sub
68 60 mV/dec SS^[6]. An attractive feature of a negative capacitance device structure is that it
69 allows for the flexibility in changing the photoactive channel material and its structure does
70 not impact the carrier transport physics. A previous study explored the application of multi-
71 layered NC MoS₂ phototransistors using capacitance matched hafnium-zirconium oxide
72 (HZO)/Al₂O₃ gate stack where they achieved an impressive steep subthreshold slope 17.64
73 mV/dec and a detectivity of 4.75×10^{14} cm Hz^{1/2} W⁻¹ at an effective optical power of 2.7 nW
74 with the photogating effect^[7]. Although the photogating effect can offer higher photocurrents
75 due to its charge trapping mechanism, a limitation of its sensitivity arises from the fixed
76 density of oxide traps in the dielectric layer. Under the illumination of different light
77 intensities, the filling of oxide trap sites by photogenerated minority carriers will reach
78 saturation. This results in limited threshold voltage shifting which directly impacts its
79 photosensitivity. In order to further improve the light sensitivity with the photogating effect
80 via higher threshold voltage shifting, the use of materials with more charge trapping storage
81 capacity is needed.

82

83 In this work, we explore for the first time monolayer MoS₂ negative capacitance
84 floating gate (NCFG) phototransistors by using a series connection of an HZO ferroelectric
85 capacitor with a conventional ZrO₂ MoS₂ phototransistor. According to NC theory, during the

86 state-to-state transition from polarization switching, the ferroelectric capacitor provides a brief
87 transient negative capacitance that couples to the positive capacitance in the conventional
88 FET device which should provide a low SS and lower power consumption. However, due to
89 the inherent floating metal gate in this device structure, it can also function as a floating gate
90 memory device to further enhance the hole-trapping from the photogating effect. As a result,
91 we obtained an enhancement in the photogenerated hole-tunneling into both oxide traps in
92 ZrO_2 and the TiN floating gate. This combined effect provided strong threshold voltage
93 shifting for weak optical powers. The highest detectivity among various types of MoS_2
94 photodetectors was achieved with peak value of $7.2 \times 10^{15} \text{ cm Hz}^{1/2} \text{ W}^{-1}$. An SS_{min} of 30.2
95 mV/dec and low dark currents in the range of $10^{-13} \sim 10^{-14} \text{ A}$ were also obtained. Moreover, due
96 to the long carrier lifetime from hole de-trapping, optical memory functionality such as multi-
97 level state programming and charge retention was measured under weak optical power
98 densities. In addition, its potential as an optical synapse in an artificial neural network was
99 also evaluated. Overall, this NCFG device structure shows a promising practical option to
100 develop high sensitivity 2D photodetectors for low power and weak light detection.

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103 **2. Results and Discussion**

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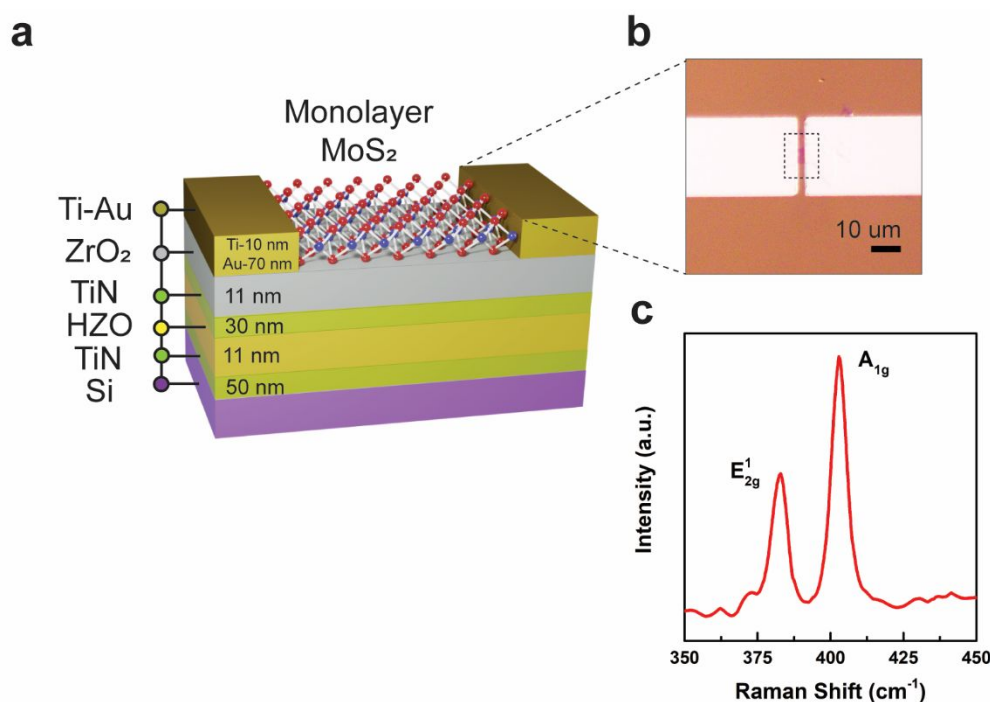
105 **2.1. Device Structure and MoS_2 Characterization**

106

107

108 **Figure 1a.** The NCFG structure was composed of HZO with top and bottoming capping
109 layers of TiN. The film thicknesses from bottom to top was TiN (50 nm)/HZO (11 nm)/TiN
110 (30 nm). TiN was deposited by DC sputtering, and HZO was deposited by atomic layer
111 deposition (ALD). After the TiN capping of HZO, a rapid thermal annealing (RTA) was
112 performed to produce ferroelectricity behavior. Next, 11 nm of ALD ZrO_2 was deposited as
113 the dielectric layer. Chemical vapor deposition (CVD) monolayer MoS_2 was transferred, and

114 then source-drain contacts of 10 nm Ti and 70 nm Au was deposited by e-beam evaporation.
 115 More details can be found in the Experimental Methods section. A close-up image of the
 116 channel region is in Figure 1b, where the dotted box emphasizes the location of the MoS₂
 117 flake. The channel length of the devices in this study were $\sim 2.5 \mu\text{m}$. A Raman
 118 characterization was performed to confirm the transferred monolayer film onto the ZrO₂
 119 dielectric layer. As seen in Figure 1c, the measured Raman shift was 19.8 cm^{-1} . As a
 120 reference, a Raman shift of around 19 cm^{-1} corresponds to monolayer MoS₂^[8].



121 **Figure 1.** a) Device structure of negative capacitance floating gate (NCFG) monolayer MoS₂
 122 phototransistor. b) Image of channel region where the dotted box shows the location of MoS₂.
 123 The scale bar is $10 \mu\text{m}$. c) Raman spectrum of transferred monolayer MoS₂ with a Raman
 124 shift of 19.8 cm^{-1} .
 125
 126
 127

128 2.2. Ferroelectricity and Negative Capacitance Effect

129
 130 The observation of ferroelectricity in HfO₂ films doped with elements such as Si^[9],
 131 Zr^[10], Y^[11], and Al^[12] has been reported. In comparison to other ferroelectric materials like
 132 lead based PZT^[13] and organic polymers like PVDF^[14], ALD doped-HfO₂ is a promising
 133 option for the development of ferroelectric devices due to its decent remnant polarization
 134 ($10\sim 45 \mu\text{C}/\text{cm}^2$) for small film thicknesses ($< 10 \text{ nm}$) and its CMOS compatible processing.

135 The driving force for ferroelectricity in doped-HfO₂ films has been attributed to the formation
 136 of the non-centrosymmetric orthorhombic phase (Pbc2₁) from tensile stresses induced from a
 137 capping layer^[9]. TiN capping with HZO is one of the most studied metal/ferroelectric/metal
 138 (MFM) structures and has been shown to offer high polarization values around a film
 139 thickness of 10 nm^[10,15,16]. In this study, a MFM capacitor structure as seen in **Figure 2a** was
 140 fabricated. To obtain a high remanent polarization (P_r), different post-metallization annealing
 141 (PMA) temperatures were analyzed. The best result was at a PMA of 700°C and its measured
 142 polarization as a function of applied electrical field (P-E) at 1 kHz is in Figure 2b. The P_r was
 143 22 μC/cm² and the coercive field (E_c) was 1.3 MV/cm. Its capacitance-voltage characteristics
 144 can be seen in supporting information fig. 1.

145 The negative capacitance concept is understood from the energy landscape of a
 146 ferroelectric capacitor that is modelled by a double well that has two energy minimums which
 147 represents the two polarization states (Figure 2c). From the Gibbs free energy for
 148 ferroelectrics:

$$149 \quad U = \alpha P^2 + \beta P^4 + \gamma P^6 + EP \quad (1)$$

150 U is the energy, P is the polarization, and E is the electric field. The static Landau coefficients
 151 of α , β , and γ are material-related parameters. For ferroelectric materials $\alpha < 0$.

152 The time-dependent Landau-Khalatnikov (LK) equation:

$$153 \quad dU/dP + \rho dP/dt = 0 \quad (2)$$

154 can be applied to obtain a relationship between the polarization and applied electric field
 155 where under steady-state conditions $dP/dt = 0$:

$$156 \quad E = 2\alpha P + 4\beta P^3 + 6\gamma P^5 + \rho dP/dt \quad (3)$$

157 A fitting of the LK model to the P-E measurement can be seen as the orange curve in Figure
 158 2b. The α , β , and γ coefficients from the fitting were -5.5×10^8 Vm/C, 4.8×10^9 Vm⁵/C³, and
 159 1.2×10^{10} Vm⁹/C⁵ respectively. The negative capacitance effect arises from the region in the

160 P-E characteristics where the slope (dP/dE) is negative as seen by the LK curve in the middle
 161 segment of the P-E measurement. This region occurs between the polarization state-to-state
 162 transition. The capacitance of a ferroelectric capacitor can be determined from equation 1:

$$163 \quad C_{FE} = [d^2U/dP^2]^{-1} \quad (4)$$

164 With the series connection of two positive capacitors, the equivalent capacitance is generally
 165 lower than the smallest capacitance value. A schematic of the negative capacitance transistor
 166 with its capacitors and node voltages are seen in Figure 2d. Although the negative capacitance
 167 state is unstable, it can be partially or fully stabilized by a series connection with a positive
 168 capacitor. Under the NC regime, C_{FE} is negative and C_{ox} is positive. As a result, the total gate
 169 capacitance becomes larger resulting in the internal amplification of the channel's surface
 170 potential for smaller V_G biasing^[6]. This NC effect was proposed to have the potential to
 171 achieve a steep SS with the attractive feature that the transport physics of the device remain
 172 unaltered.

173 To test the potential of the internal voltage amplification gain ($\delta V_{int}/\delta V_G$), an MFMIM
 174 structure (inset of Figure 2b) was measured. The measurement involved sweeping the gate
 175 voltage and measuring the floating potential of TiN under a zero current bias. The
 176 amplification gain factor for both the forward and reverse scans can be seen in Figure 2b. It
 177 displayed a transient amplification gain where the peaks of the forward and reverse gains
 178 were 3.79 and 2.31 respectively. The transfer characteristics of the ferroelectric driven NCFG
 179 phototransistor under the dark condition (no light illumination) can be found in Figure 2e at
 180 $V_{DS} = 100$ mV. This device displayed a small hysteresis window of 0.35V, a forward
 181 threshold voltage V_{TH} of 0.5 V, an I_{on}/I_{off} ratio of 6.6×10^6 , and a low I_{off} in the range of $\sim 10^{-14}$
 182 A. A sub 60 mV/dec SS was observed in the lower subthreshold region where the gate
 183 leakage current exceeds the drain current. This behavior of $I_G > I_D$ in this region can also be
 184 seen in a similar device structure using HfO_2 as the dielectric layer^[32]; however, they
 185 achieved a steep SS as I_D became larger than I_G . Although most NC devices have claimed sub

186 60 mV/dec SS over multiple current decades, they have not provided detailed discussion that
 187 the gate current contribution in this lower SS region can cause the drain current to increase
 188 rapidly, thus yielding misleading SS values. In order to evaluate the true SS_{\min} without the
 189 gate current contribution in this region, an approximation method found in supporting
 190 information fig. 2 was used. In Figure 2f, it shows the subthreshold swing as a function of the
 191 drain current where the forward SS_{\min} was 30.2 mV/dec. According to the quasi-static NC
 192 theory that was previously discussed, a stabilized NC effect from capacitance matching
 193 should produce hysteresis-free I-V. However, for this device, hysteretic behavior was
 194 observed and the sub-60 mV/dec SS occurred only in the forward sweep which aligns with the
 195 forward scan transient voltage amplification gain. The transfer characteristics showed a
 196 clockwise hysteresis loop indicating the presence of electron trap sites near the conduction
 197 band edge of MoS_2 most likely due to presence of ambient gases and absorbates from the
 198 exposed channel. As a result, the reverse curve does not coincide with the reverse
 199 amplification gain, thus resulting in a higher SS. It is most likely that this device displays
 200 transient NC effects due to the transition in polarization switching in the ferroelectric layer.

201

202 A feature of NCFETs is the observation of negative differential resistance (NDR) in
 203 the output characteristics due to the coupling of the drain voltage (V_D) and the internal gate
 204 voltage (V_{int}). Drain-Induced Barrier Lowering (DIBL) is an undesirable short-channel effect
 205 where at larger V_D , the barrier height between the source and drain gets lowered where both
 206 the gate and drain voltages control the channel. For NCFETs, the internal gate voltage is a
 207 function of the gate and drain voltage:

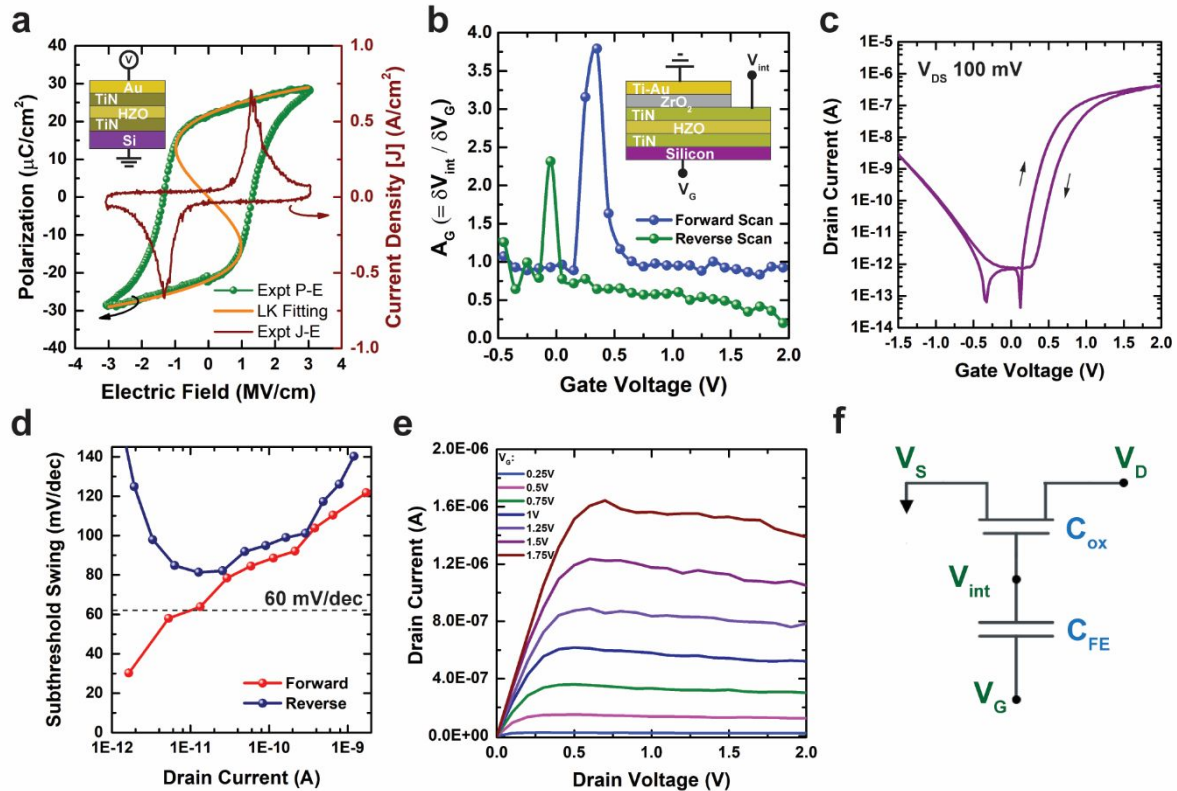
$$208 \quad V_{int} = \frac{|C_{FE}| \times V_G}{|C_{FE}| - C_{MOS}} + \frac{-C_{GD} \times V_D}{|C_{FE}| - C_{MOS}} = A_G \times V_G + [-A_D \times V_D] \quad (5)$$

209 where C_{FE} is the ferroelectric capacitance, C_{MOS} is the total underlying transistor capacitance,
 210 A_G is the voltage amplification gain, and A_D is the drain coupling factor. Although V_G is held

211 constant, V_{int} changes due to the gate-to-drain capacitance coupling. Initially, V_{int} is high
212 when $V_{\text{D}} = 0\text{V}$; however, as V_{D} increases, V_{int} drops resulting in a reduction of the gate
213 charge^[17,18]. In the saturation region of the output characteristics where V_{G} is constant and V_{D}
214 is increasing, a decrease in the channel current can be observed. The output characteristics in
215 Figure 2g displayed a peaking of the drain current followed by a decrease with increasing V_{D} .
216 At lower V_{D} as seen in supporting information 3, linear output curves were measured
217 indicating ohmic-like contacts to MoS_2 . In supporting information fig. 4, $I_{\text{D}}-V_{\text{D}}$ was measured
218 under different voltage sweeps with another device which also displayed this NDR effect. For
219 comparison, conventional FET device structures of back-gated Si-ZrO_2 and Si-TiN-ZrO_2 were
220 also measured where its output characteristics can be found in supporting information fig. 5
221 and fig. 6, respectively. Their output characteristics did not display this NDR effect. The
222 transfer characteristics of the Si-ZrO_2 and Si-TiN-ZrO_2 are seen in supporting information fig.
223 7 and 8 respectively. The high gate current leakage in the Si-ZrO_2 can be attributed to the
224 preferred tetragonal phase crystal growth on the silicon back gate in comparison to the TiN-
225 ZrO_2 (supporting information fig. 9).

226

227



228
 229 **Figure 2.** a) Structure of metal-ferroelectric-metal (MFM) capacitor (inset image). P-E
 230 measurement of MFM capacitor at 700°C annealing at 1kHz (green line). L-K fitting to the
 231 measurement is plotted in orange. Current density is the red plot (right side). b) Voltage
 232 amplification of the gate stack. c) Transfer characteristics of the NCFG phototransistor under
 233 dark condition. d) Subthreshold swing as a function of drain current. e) Output characteristics
 234 under dark condition. f) Device schematic of negative capacitance floating gate (NCFG)
 235 phototransistor with capacitors and voltage nodes.

236
 237

238 2.3. Photoresponse

239
 240

241 The photoresponse of the NCFG phototransistor was evaluated with green light
 242 illumination ($\lambda = 515 \text{ nm}$) and under ambient conditions. As previously mentioned, low light
 243 intensities in the range of $P_{\text{eff}} = 1.4 \text{ fW}$ to 6.3 pW for the effective light powers was studied to
 244 characterize its sensitivity to low light detection. **Figure 3a** shows the photoresponse of the
 245 NCFG device biased under $V_{DS} = 100 \text{ mV}$. The strong parallel shifting of the illumination
 246 curves indicates that the dominating photocurrent generation comes from the photogating
 247 effect. The photogating effect relies on the trapping of the photogenerated holes at the
 248 dielectric/semiconductor interface, which results in developing a local built-in electric field
 that shifts the Fermi level to induce more electrons.^[19,20,42] In addition, to evaluate the

249 enhancement in photodetection with the NCFG device structure, a conventional back-gate
250 TiN-ZrO₂ and Si-ZrO₂ device were measured and their photoresponse results can be found in
251 Figure 3b,c respectively. The Si-ZrO₂ device structure showed higher gate leakage current
252 due to the polycrystalline structure from the as-deposited ZrO₂ resulting in a higher I_{off}. In
253 addition, its light curves showed weak detection of low optical power densities. With the TiN-
254 ZrO₂ device, it showed lower gate leakage current due to the suppression of the tetragonal
255 (111) crystal phase in comparison to the Si-ZrO₂ device structure leading to a lower I_{off}. The
256 incorporation of the TiN metal back gate provided an improvement in detecting the same
257 optical power densities. The NCFG device was able to better detect the lowest optical power
258 density of 1.19 μW/cm² in comparison to the other device structures which either could not
259 detect (Si-ZrO₂) or could detect weakly (TiN-ZrO₂). In Figure 3d, it shows the change in the
260 threshold voltage from light illumination under varying optical power densities where $\Delta V_{TH} =$
261 $V_{LIGHT} - V_{DARK}$. The observation of threshold voltage shifting is a method of confirmation of
262 the presence of photogating. This plot shows the result from all 3 device structures where the
263 NCFG device showed overall the greatest amount of threshold voltage shifting. The
264 maximum change in the threshold voltage shifting was -0.92 V for 51.8 μW/cm² which is
265 quite a low light intensity in comparison to other MoS₂ phototransistors that have investigated
266 photogating at higher optical powers.^[7,20,21,22]

267

268 The photodetection mechanism for this device relies on the photogating effect. We
269 have previously studied high-k metal oxide dielectric HfO₂ with multi-layered MoS₂ and
270 found the photogating effect was a dominating photocurrent generation mechanism.^[30] This
271 was due to the valence band maximum (VBM) of MoS₂ having good band edge alignment
272 with the intrinsic oxygen vacancies of HfO₂. For ZrO₂, it also has oxygen vacancy point
273 defects located within its bandgap which serve charge-trapping sites^[23,24]. Even without the
274 presence of light irradiation, electrical stress biasing can also induce charging effects into

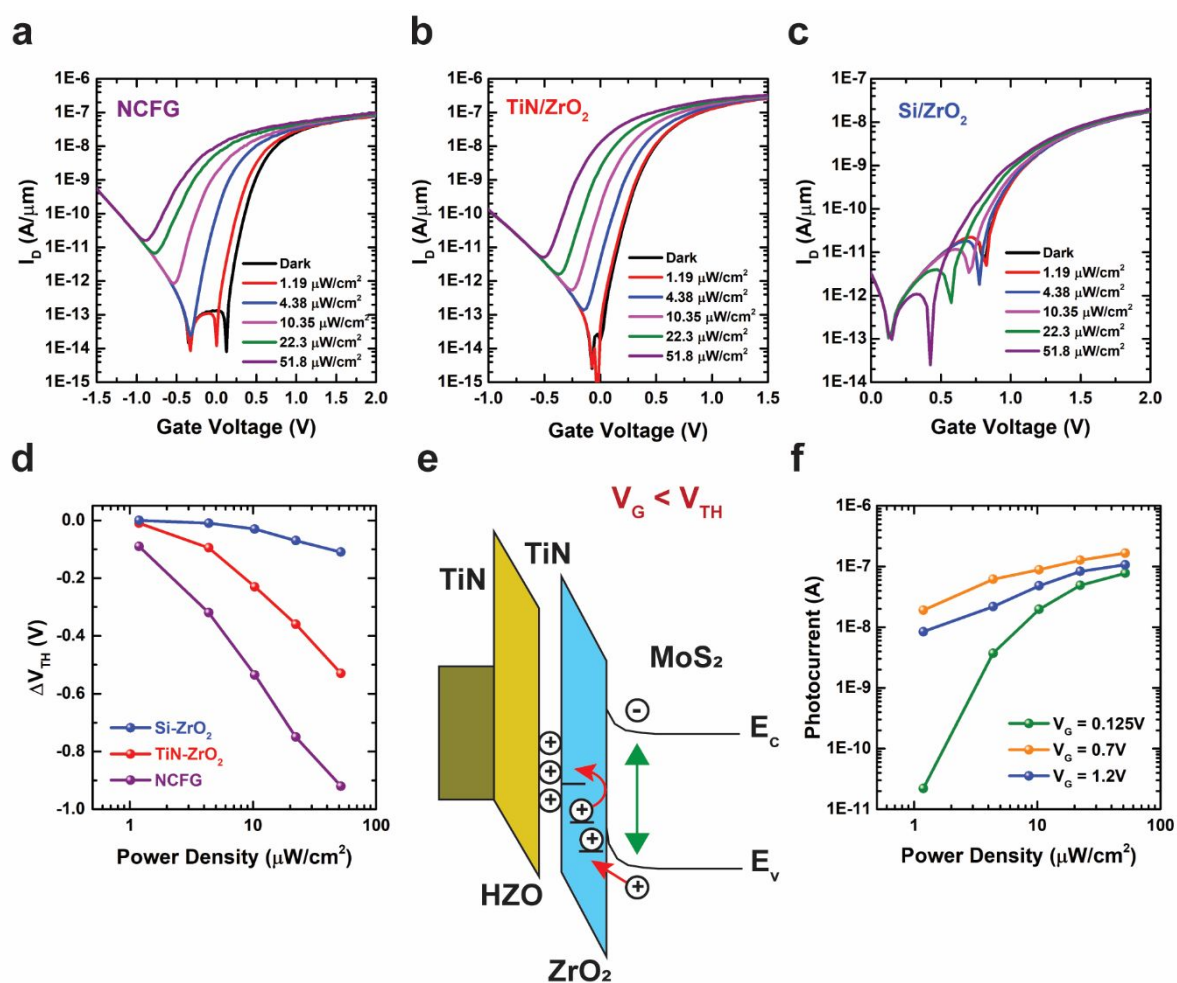
275 ZrO_2 where holes can be injected. ^[25] The photogenerated hole trapping at the $\text{MoS}_2/\text{ZrO}_2$
276 interface most likely occurs via charge tunneling from the VBM alignment of MoS_2 to these
277 oxide defect energy levels. In addition, the presence of ambient gases and adsorbates that are
278 known to deplete the MoS_2 channel can also serve as an additional source of charge-trapping
279 sites^[26,41].

280

281 With the integration of a ferroelectric capacitor to a high-k 2D FET, it contains a
282 floating gate (TiN) that can serve as an additional charge-trapping storage layer. During the
283 photogenerated carrier extraction process in MoS_2 , the photogenerated holes can tunnel into
284 ZrO_2 via oxygen vacancy point defects due to its VBM alignment. Due to the intrinsic fixed
285 defect density in ZrO_2 , additional photogenerated holes can tunnel into the TiN floating gate
286 due to the enhancement of the electric field generated from the transient voltage
287 amplification. This enhanced photogating effect was more pronounced in the NCFG device
288 structure in comparison to the conventional Si- ZrO_2 and TiN- ZrO_2 devices which represents
289 the case of a non-floating gate/ferroelectric device. An overview of this photogating
290 mechanism process can be seen in Figure 3e which shows the movement of the
291 photogenerated hole carriers. Evidence of hole-trapping can be indirectly observed in the gate
292 current where an enhancement in the electron leakage current results from a barrier lowering
293 effect from the hole-trapping into an oxide layer^[34]. In supplementary figure 10, it shows the
294 gate current in ZrO_2 as a function of the gate voltage for the NCFG and TiN- ZrO_2 devices
295 under the dark condition and illumination. The hump observed in the illumination curves
296 corresponds to the enhanced electron tunneling due to the photogenerated hole-trapping in
297 ZrO_2 . Overall, the NCFG structure showed more pronounced humps indicating there is more
298 hole-trapping in this device structure under illumination.

299

300 The NCFG photocurrent generation was also evaluated. The photocurrent as a function
 301 of V_G can be seen in supplementary figure 11. At $V_G = 0.7$ V which is 0.2 V above its dark
 302 V_{TH} , this device displayed peak photocurrents ranging from 1.8×10^{-8} A to 1.6×10^{-7} A. The
 303 I_{PH}/I_{DARK} ratio was as high as 2×10^6 for $V_G = 0.125$ V at optical power density $51.8 \mu\text{W}/\text{cm}^2$.
 304 Lastly, the relationship between the photocurrent and optical power density can be seen in
 305 Figure 3f under different gate biasing conditions: 0.125 V for depletion mode, 0.7 V slightly
 306 above V_{TH} and around the peak photocurrent generation, and 1.2 V for accumulation mode
 307 operation. All of these curves showed a non-linear photocurrent dependence on the optical
 308 power density indicating that there is a presence of traps and complicated photocarrier
 309 generation processes^[27].



310 **Figure 3.** a) Photoresponse of NCFG phototransistor under green light illumination and V_{DS}
 311 at 100 mV. b) Photoresponse of monolayer TiN-ZrO₂ (conventional FET) phototransistor. c)
 312 Photoresponse of monolayer Si-ZrO₂ (conventional FET) phototransistor. d) Threshold
 313 voltage shift from illumination at different optical power densities for NCFG (purple), TiN-
 314

315 ZrO₂ (red), and Si-ZrO₂ (blue). e) Mechanism of the photogating effect with the
 316 photogenerated hole-trapping into ZrO₂ and in the TiN floating gate. f) Photocurrent as a
 317 function of optical power density at different gate voltage biasing showing sublinear
 318 dependence (non-photoconductive).

319

320 **2.4. Photodetection Metrics and Optical Memory application**

321

322 To characterize the NCFG phototransistor's performance as a light detector, the

323 responsivity and detectivity were evaluated. Responsivity is a measure of the current

324 generated from the incident optical power and can be determined by the equation:

$$325 \quad R = I_{\text{ph}}/P_{\text{opt}} \quad (1)$$

326 where I_{ph} is the photocurrent ($I_{\text{LIGHT}}-I_{\text{DARK}}$) and P_{opt} is the effective optical power. In **Figure**

327 **4a**, the responsivity is shown at different gate biasing voltages. The highest responsivity

328 achieved was 1.3×10^5 A/W for the minimum optical power density of $1.19 \mu\text{W}/\text{cm}^2$. The trend

329 for the responsivity decreases with increasing light intensities; however, for the lower gate

330 bias, the responsivity started at a low value, peaked, and then decreased. This inverted

331 parabolic trend arises from the lower photocurrent generated in comparison to the other higher

332 gate biases. The detectivity describes the light sensor's sensitivity and can be determined

333 from:

$$334 \quad D^* = \frac{R\sqrt{A}}{\sqrt{2qI_{\text{dark}}}} \quad (1)$$

335 where R is the responsivity, A is the area of the channel, q is the electronic charge, and I_{dark} is

336 the dark current. The detectivity at varying optical power densities under different gate biases

337 can be seen in Figure 4b. A peak detectivity of $7.2 \times 10^{15} \text{ cm Hz}^{1/2} \text{ W}^{-1}$ was obtained with the

338 device biased at $V_G = 0.175 \text{ V}$ (depletion mode). This bias point was selected, since $I_D > I_G$ at

339 this operation point. At $V_G < V_{\text{TH}}$, the device is operated in the subthreshold region where the

340 NC effect is supposed to occur. In the lower SS region, the dark current was as low as $\sim \text{fA}$

341 range. A comparison of detectivity among various device structures of MoS₂

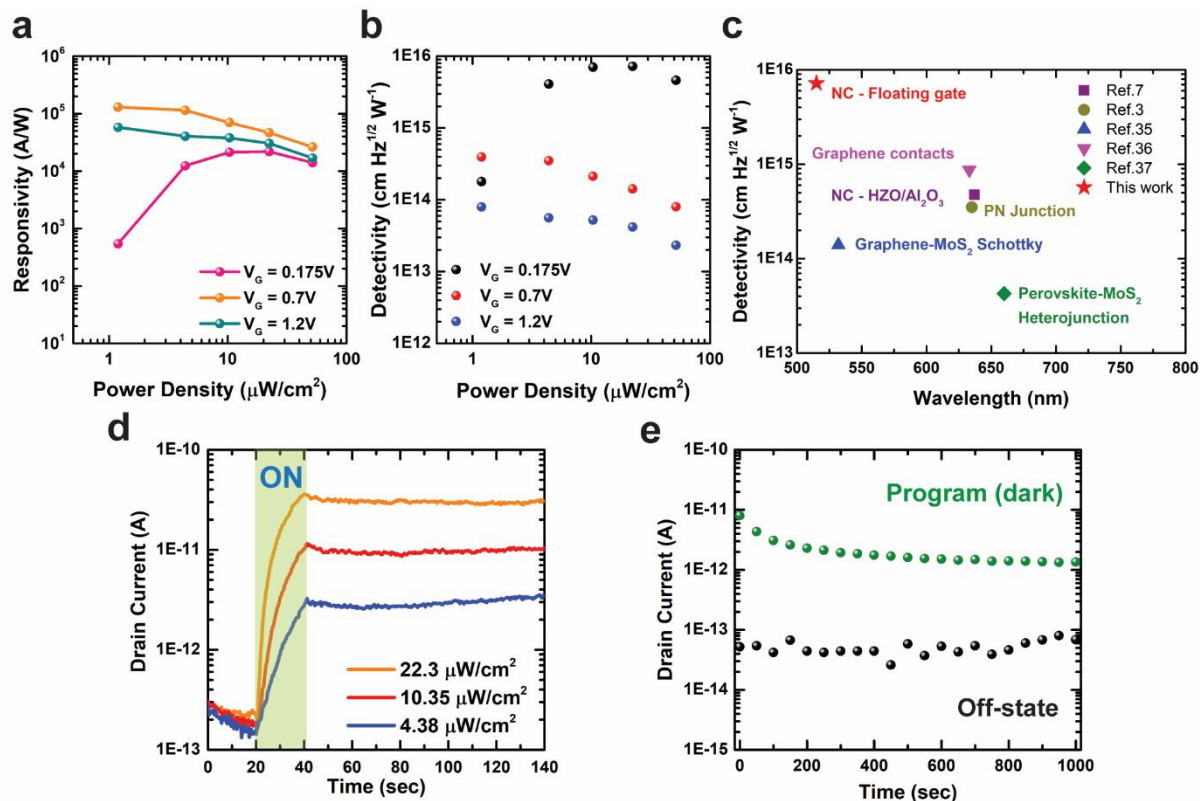
342 photodetectors^[3,7,35,36,37] is summarized in Figure 4c where this study offers the highest

343 detectivity. Although the detectivity and responsivity are competing parameters, this NCFG

344 device structure offers an opportunity to further suppress the dark current and enhances the
345 photogating effect to increase its light sensitivity.

346 Optical memory functionality was explored with the NCFG monolayer MoS₂
347 phototransistor. Since the photocurrent generation mechanism process relies on the charge-
348 trapping of the photogenerated holes tunneling into ZrO₂, the time response displays a long
349 relaxation process which is known as the persistent photocurrent effect^[28,29]. Although the
350 charge-trapping process results in a slow time response, this feature can be useful for
351 applications such as neuromorphic optical synapses or non-volatile optical memories. To
352 explore the potential of this device as an optical memory, we tested multi-state programming
353 and charge retention characteristics using weak light intensities since it can allow for a low-
354 powered application. In Figure 4d, it shows the multi-state programming of three
355 photocurrents generated from 3 different optical power densities. After the light pulse
356 programming, the photocurrent remained stable for 100 seconds after light exposure. Next, its
357 retention characteristics for 1000 second duration is found in Figure 4e where after the light
358 pulse programming, the programmed photocurrent and the dark current (off-state) was
359 measured. At the beginning, the programmed and off-state had a $\sim 10^2$ order of magnitude
360 difference. After 1000 seconds, the difference decreased to ~ 10 . Although it is common to
361 introduce additional dielectric layers to improve the charge retention^[30,31], this addition may
362 impact the internal voltage amplification, so an appropriate design is needed. As for its use as
363 a photodetector, the time response can be improved by applying a reset pulse to release the
364 trapped charges as seen in supplementary figure 12 and 13. An interesting direction to further
365 explore is the use of steep slope photodetectors capable of tuning between positive and
366 negative photocurrent generation. 2D heterostructures seem to be a promising photoactive
367 channel material to observe negative photocurrents, since the photogenerated charge transfer
368 process can occur between the layers^[43].

369



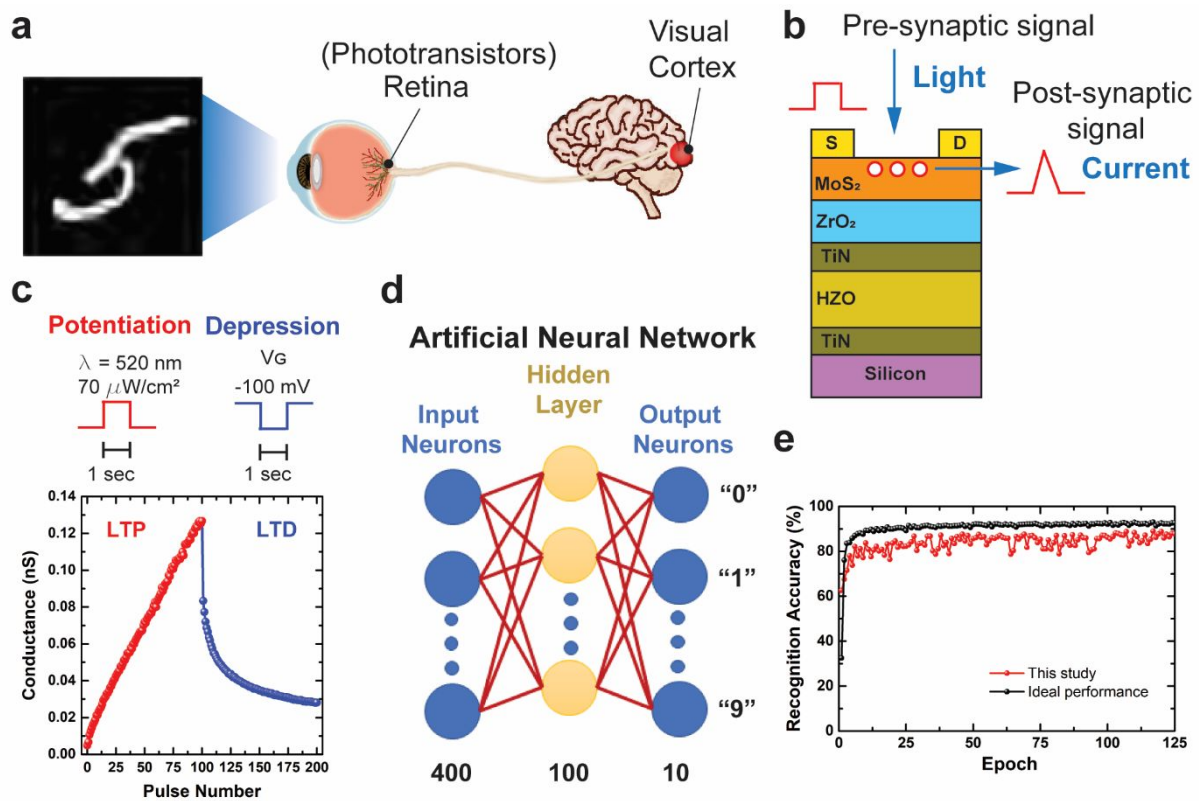
370
 371 **Figure 4.** a) Responsivity vs. optical power density for different gate biases. b) Detectivity vs. optical power density for different gate biases. c) Benchmark of detectivity for different MoS₂
 372 device structures. d) Multi-level photocurrent states after 20 second light pulse programming with V_{DS} = 100 mV and V_G = -150 mV. d) Optical memory retention of light current after
 373 light programming (green line) and the dark off-state (black line) with V_{DS} = 250 mV and V_G
 374 = -150 mV.
 375
 376
 377

378 2.5. Optical Synapse and Artificial Neural Network

379

380 Neuromorphic computing systems are considered to be a possible future candidate to
 381 replace the conventional Von Neumann model utilized in current computing architectures. An
 382 efficient approach to build this system is the use of devices with built-in synaptic
 383 functionalities where one device represents a synapse. In particular, optical synaptic
 384 transistors can be used towards the development of neuromorphic vision systems where it can
 385 mimic the light sensing and initial processing that occur in the retina. A schematic overview
 386 can be seen in **Figure 5a** where phototransistors can be used to act as a transducer to convert
 387 light into an electronic signal. Neurons transmit information from the pre and post neurons via
 388 the release of neurotransmitters from the pre-neuron and its binding to the receptors on the

389 post neuron. For optical synapse operation (Figure 5b), the pre-synaptic signal is provided by
390 a short light pulse acting as an optical spike which generates the post-synaptic signal called
391 the post-synaptic current (PSC). The synaptic weight can be determined from the connection
392 strength between the pre and post neuron that can be modified via learning. The synaptic
393 strength can be enhanced by long-term potentiation (LTP) and weakened by long-term
394 depression (LTD). The LTP/LTD characteristics in Figure 5c shows the LTP by optical
395 stimulation with green light pulses with an optical power density of $70 \mu\text{W}/\text{cm}^2$, $V_G = 0\text{V}$, and
396 $V_{DS} = 150 \text{mV}$. For LTD, electrical stimulation was used with $V_G = -100 \text{mV}$ and $V_{DS} = 150$
397 mV for the read pulse since it is difficult to demonstrate optical depression (generating
398 negative photocurrent). The LTP behavior showed linear-like conductance modulation which
399 can be attributed to the PPC effect. Next, a NCFG artificial neural network (ANN) was
400 simulated and its pattern recognition accuracy was tested using the MNIST handwriting
401 dataset. The ANN^[33] shown in Figure 5d consists of 400 input neurons, 100 hidden layer
402 neurons, and 10 output neurons. Its recognition accuracy after 125 epochs in Figure 5e
403 showed an accuracy of 87.8%. In comparison, the ideal performance had an accuracy of
404 92.6%.



405
 406 **Figure 5.** a) Schematic of neuromorphic vision system where phototransistor is used to mimic
 407 the functions of the retina. b) Device operation as an optical synapse. c) Long-term
 408 potentiation (LTP) by optical stimulation with $V_G = 0\text{V}$ and $V_{DS} = 150 \text{ mV}$. Long-term
 409 depression (LTD) by electrical stimulation with $V_G = -100\text{mV}$ and $V_{DS} = 150 \text{ mV}$ for the read
 410 pulse. d) Schematic of the artificial neural network (ANN) used in the simulation for image
 411 recognition of MNIST handwriting dataset. The ANN consists of 400 input neurons, 100
 412 hidden layer neurons, and 10 output neurons. e) ANN simulation results of the handwriting
 413 recognition accuracy. After 125 epochs, the NCFG neural network showed an accuracy of
 414 87.8% and the ideal case was 92.6%.

415

416 3. Conclusion

417

418 In conclusion, a monolayer MoS₂ negative capacitance floating gate (NCFG) phototransistor
 419 device structure was investigated. This device structure contains a ferroelectric capacitor
 420 integrated with a conventional 2D FET which has potential for negative capacitance operation
 421 and also contains a floating gate. A strong photogating effect was observed that produced
 422 large threshold voltage shifting for weak light illumination. As a result, the highest detectivity
 423 among various MoS₂ photodetectors was achieved with a peak value of $7.2 \times 10^{15} \text{ cm Hz}^{1/2} \text{ W}^{-1}$
 424 due to its enhancement in weak light detection in comparison to a conventional FET device
 425 structure. This device structure offers flexibility to transfer other kinds of 2D materials as
 426 photoactive channels to benefit from this enhanced photogating behavior. In addition, optical

427 memory functionality of multi-state programming and charge retention was demonstrated for
428 a duration of 1000 seconds. This device was also explored as an optical synapse where it
429 displayed excellent long-term potentiation characteristics and was simulated in an artificial
430 neural network (ANN). This NCFG ANN displayed an image pattern recognition accuracy of
431 87.8% after 125 epochs. Overall, the enhancement of photogenerated hole-trapping with this
432 NCFG device structure allows for improved light sensitivity for weak light illumination.

433

434 **4. Experimental Methods**

435 *Device Fabrication:*

436 Heavily doped n-type Silicon was used as the substrate for the back gate. Next, DC sputtering
437 of 50 nm of TiN was deposited and was followed by 11 nm of ALD deposited $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$
438 and then DC sputtering again with 30 nm of TiN. RTA post-metallization annealing at 700 °C
439 was performed to achieve ferroelectricity. Next, ALD deposition of ZrO_2 at 300 °C with
440 thickness of 11 nm. CVD monolayer MoS_2 purchased from SixCarbon Technology and was
441 transferred onto ZrO_2 layer. Source/drain contacts were patterned using a photolithography
442 process with channel length of 2.5 μm . Metal contacts of 10 nm Ti/ 70 nm Au was deposited
443 by e-beam evaporation. Aluminum was deposited on the backside of Silicon to provide better
444 electrical contact for the back gate, and then lift-off was performed.

445 *Device Measurements:*

446 All measurements were performed at room temperature and under ambient conditions with a
447 dark curtain. A laser diode purchased from Thorlabs with wavelength of 515 nm was used for
448 the photoresponse and optical memory measurements. A commercial LED with wavelength
449 of 520 nm was used for the optical synapse measurements. Device measurements were
450 performed using Cascade probe system (Form Factor) and Agilent 4156C Precision
451 Semiconductor Parameter Analyzer. For a steady state condition for illumination
452 measurements, the laser diode was on for ~1 minute before the measurement was taken.
453 Agilent 33500B series waveform generator to provide the light pulse waveforms.

454

455 **Supporting Information**

456

457 Additional measurements (PDF).

458

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467

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