Electronic Supplementary Material (ESI) for Materials Horizons.

Supporting Information

Monolayer hydrophilic MoS_2 with strong charge trapping for atomically thin neuromorphic vision systems

Yunxia Hu, [ab] Mingjin Dai, [ab] Wei Feng, [c] Xin Zhang, [b] Shichao Zhang, [b] Biying Tan, [b] Huiming Shang, [b] YongQing Fu,[d] PingAn Hu[*ab]

School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, P. R. China.

MOE Key Laboratory of Micro-Systems and Micro-Structures Manufacturing, Harbin Institute of Technology, Harbin 150001, P. R. China

College of Chemistry, Chemical Engineering and Resource Utilization, Northeast Forestry University, Harbin 150040, P. R. China

Faculty of Engineering & Environment, Northumbria University, Newcastle upon Tyne, NE1 8ST, UK

†Electronic Supplementary Information (ESI) available: See DOI: 10.1039/ x0xx00000x

*E-mail: hupa@hit.edu.cn

Experimental Section:

Preparation of precursor solution: 0.05 g $(NH_4)_2Mo_4O_{13}\cdot 2H_2O$ (Aladdin, 99%) and 0.84 g KOH were dissolved in 5 mL of deionized water in a small vial to obtain 10 $mg\cdot ml^{-1}$ precursor solutions. Then 20 μl precursor solutions were directly spin-coated on mica substrates at different speeds.

CVD growth of MoS_2: Mica substrates with precursor solution drops were used as the source substrates and growth substrates simultaneously. Several spin-coated substrates on a quartz plate were placed at the downstream heating center of a quartz tube, which was ramped up to 750 °C for 50 min. An alumina boat loaded with 200 mg of sulfur was placed at another upstream heating center in the tube. The S powder was heated to 250 °C when the substrates were heated to 750 °C. This reaction was carried out with 30 sccm flow of argon gas at an atmospheric pressure. Then the furnace was cooled down naturally to room temperature. When the mica substrate was placed on the 22 cm away from the sulfur powder and the growth time was 20 min with a spin speed lower than 3000 rpm, a monolayer MoS_2 film was obtained.

Characterization: The morphology of the MoS_2 was illustrated using an optical microscope (Nikon Eclipse LV100). Raman and PL spectra/maps were collected with a confocal Raman spectroscopy (LabRAM XploRA) using a 532 nm laser as the excitation source. Atomic force microscopy (Dimension Icon, Bruker) was carried out to probe the thickness of the samples. Transmission electron microscope (Tacnai-G2 F30) was performed to determine the structure of MoS_2. MoS_2 films were transferred on a copper grid for TEM characterization. XPS X-ray photoelectron spectroscopy (Thermo Scientific K-alpha XPS, using Al (Kα) radiation as a probe) was used to characterize the chemical composition of the as-grown MoS_2 sample with −OH.

Device fabrication and measurement: The MoS_2 optoelectronic memory device was fabricated by transferring monolayer MoS_2 single crystals from mica substrate to SiO_2 substrate using a polymethyl methacrylate (PMMA) assisted transfer approach. After the patterns of electrode contacts of the device were obtained using a standard photolithography process, Au (30 nm) source and drain electrodes were deposited by thermal evaporation. Then devices were treated by thermal annealing at 200 ℃ for 1 h in to minimize the effects of physisorbed oxygen and water on the MoS_2 surface. The optoelectronic memory performances were analyzed using a Keithley 4200-SCS Parameter Analyzer and a Cascade Microtech probe station. The laser applied to devices was carried out with a xenon lamp, monochromator (Zolix, Omni-λ300i), and the corresponding light power density was measured by a power meter (Thorlabs, PM100D). All measurements were performed at room temperature. To demonstrate the image memorization of our neuromorphic vision sensors, an optical mask with rose shape was applied. In each pixel, the length of the channel is 5 μm and the width of the channel is 10 μm in each device. The pixel was illuminated and measured one by one, and the pixel current was received sequentially.

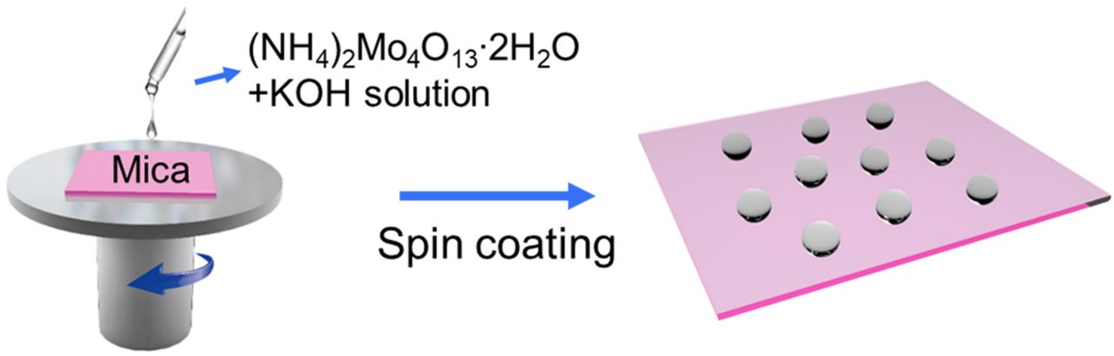
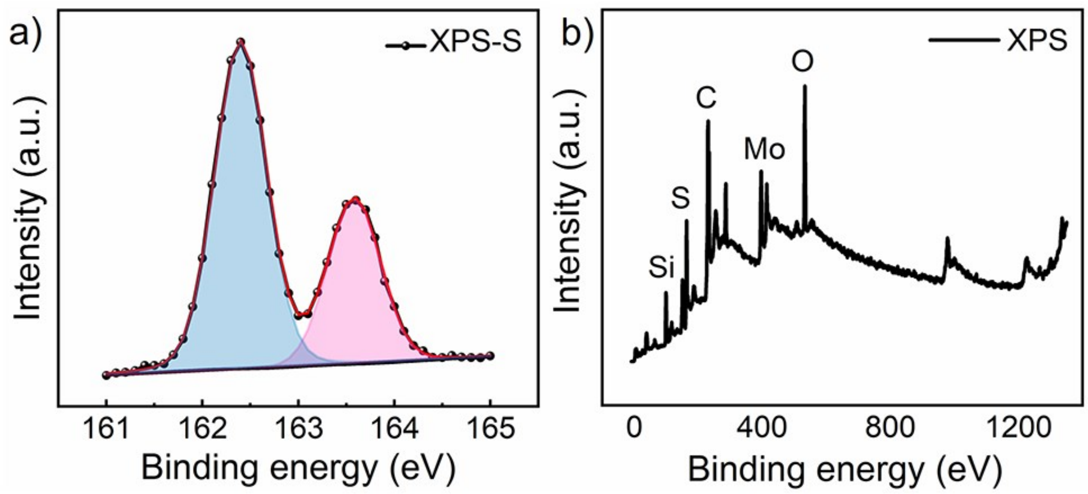


Fig. S1 Illustration of the synthesis process for MoS_2 on mica substrate by spin coating



precursor solution.

Fig. S2 XPS spectra of as-grown MoS_2 samples with KOH (a) S 2p and (b) full XPS spectra of MoS_2 on silicon substrate.

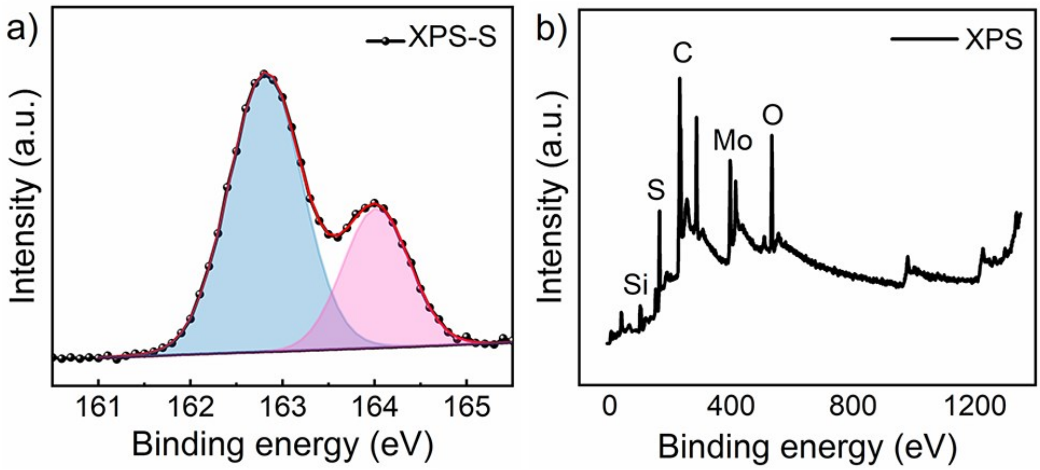
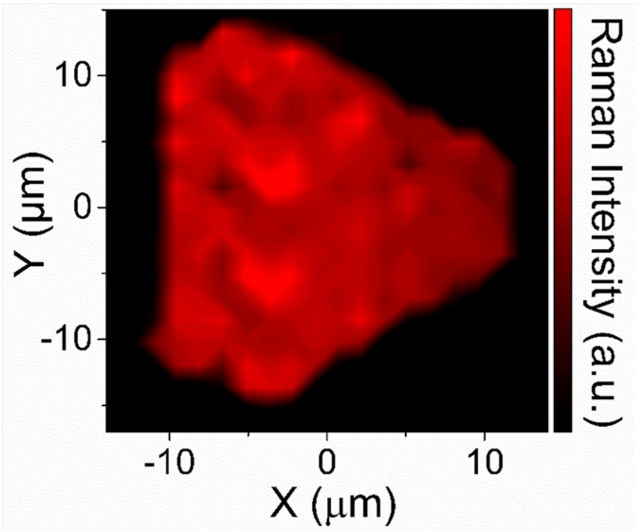


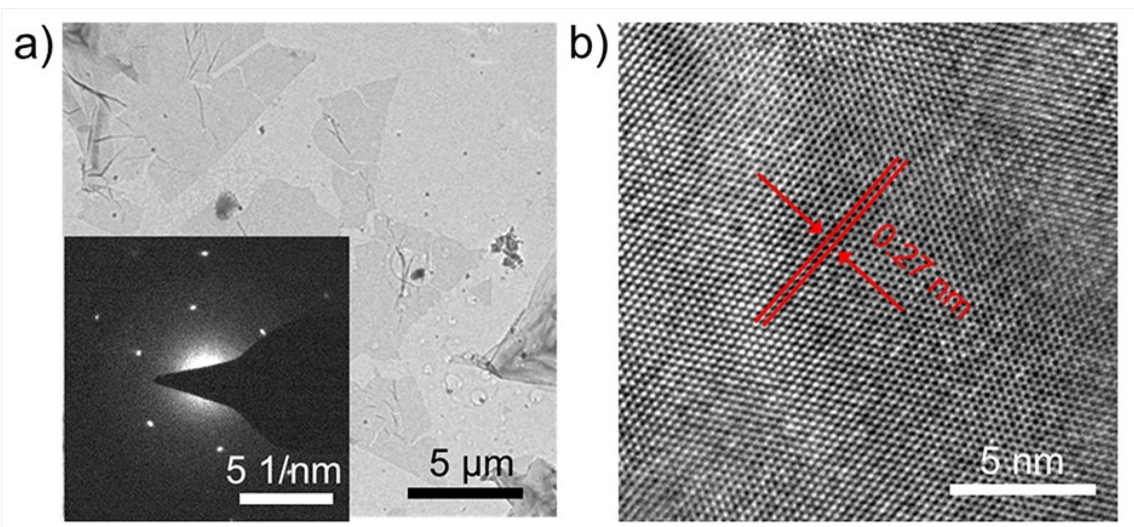
Fig. S3 XPS spectra of as-grown MoS_2 samples without KOH (a) S 2p and (b) full XPS spectra of

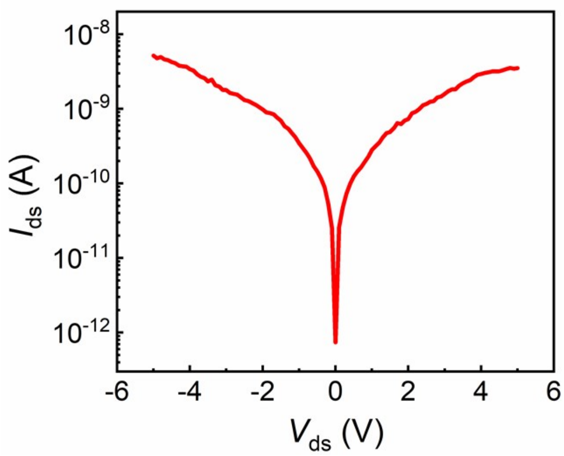


MoS_2 on silicon substrate.

Fig. S4 Raman mapping image of the E^1_{2g} peak in monolayer MoS_2 single crystals.

Fig. S5 (a) The TEM image at low magnification of monolayer MoS_2 with KOH. Inset: corresponding SAED pattern. (b) HRTEM image of monolayer MoS_2.





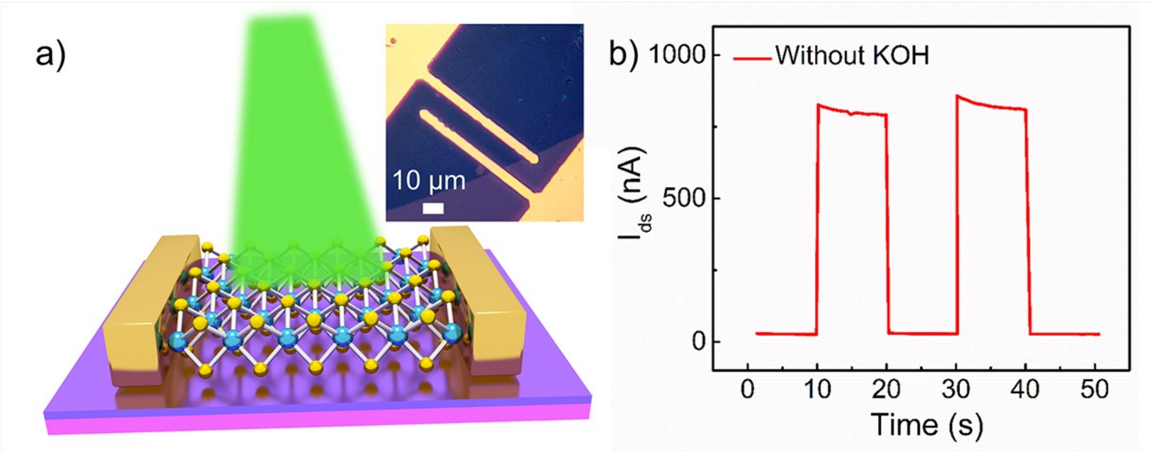


Fig. S6 The output curves of this hydrophilic MoS_2 FET.

Fig. S7 (a) Schematic of the fabricated based on MoS_2 without KOH. Inset: the optical image of the MoS_2 device. (b) Operating sequence and readout current of the synapse device at 20 V gate voltage. A gate pulse (Vg = 60 V, 2 s) and a source-drain readout bias (V_{ds} = 3 V, 10 s) were applied for the reset and readout operations, respectively. The green vertical column indicates the period when the device was illuminated with a 550 nm laser with a power of 1.04 mW/cm^2. The inset represents an enlarged view of the readout current of Off-state.

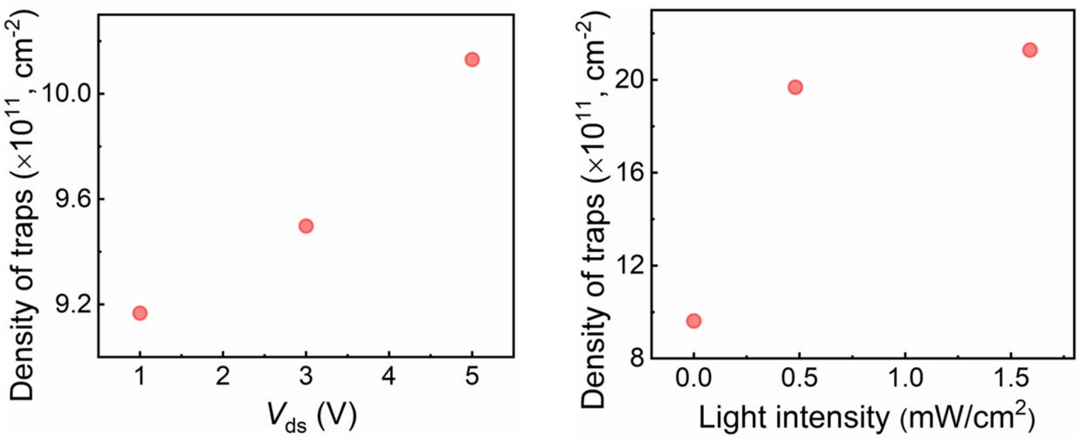
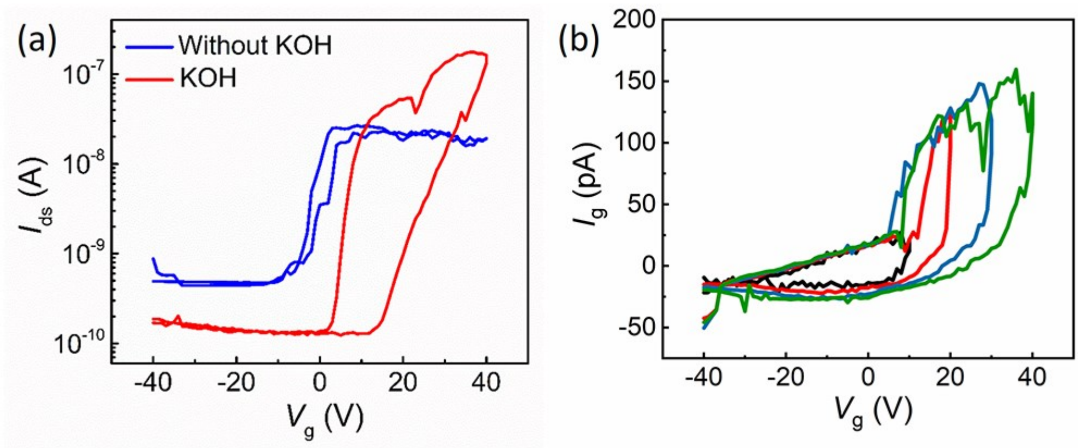


Fig. S8 (a) The density of trapped charges of hydrophilic MoS_2 FET under different V_{ds}. (b) The density of trapped charges of hydrophilic MoS_2 FET with different light intensity of 550 nm



laser at V_{ds}=3 V.

Fig. S9 (a) I_{ds}–V_g curves of the device based on MoS_2 with KOH (red) and without KOH (blue). (b) the I_g vs V_g curves with different positive V_g ranges of hydrophilic MoS_2 FET.

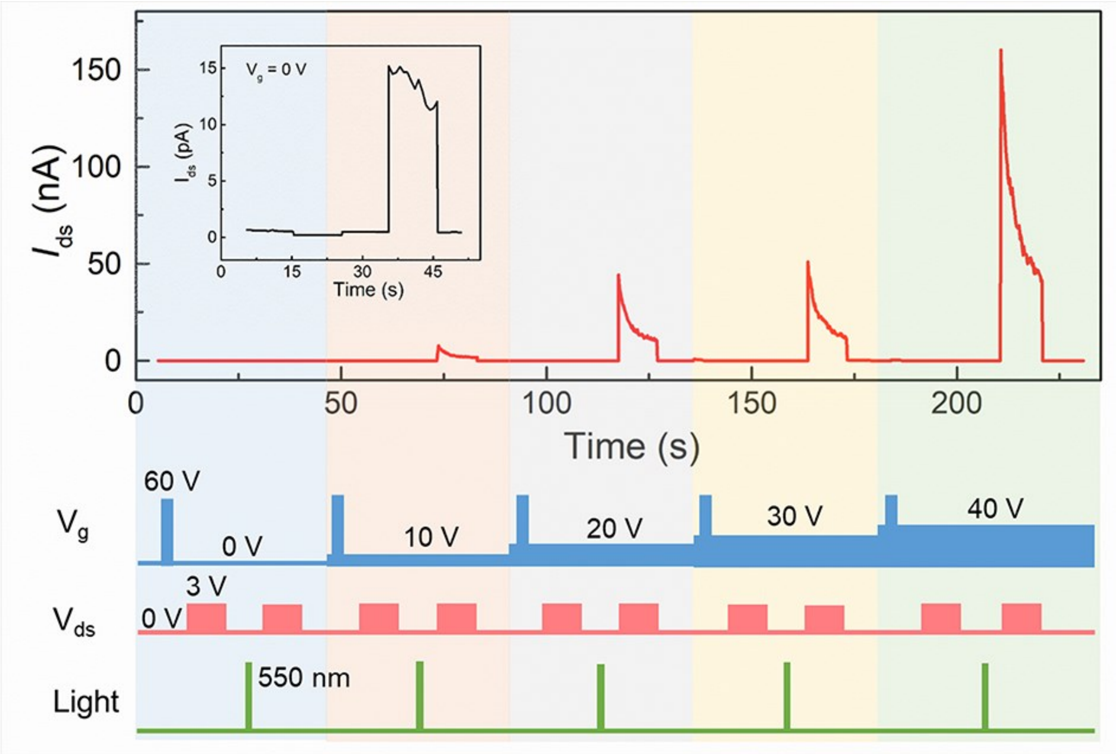


Fig. S10 Characterization of the optical memory with varying gate voltages. Operating sequence and readout current of the optoelectronic device at different gate voltages. A gate pulse (Vg = 60 V, 2 s) and a source-drain readout bias (Vsd = 3 V, 10 s) were applied for the reset and readout operations, respectively. The green vertical column indicates the period when the device was illuminated with a 550 nm laser with a power of 1.04 mW/cm^2. The inset represents an enlarged view of the readout current for different gate voltage.

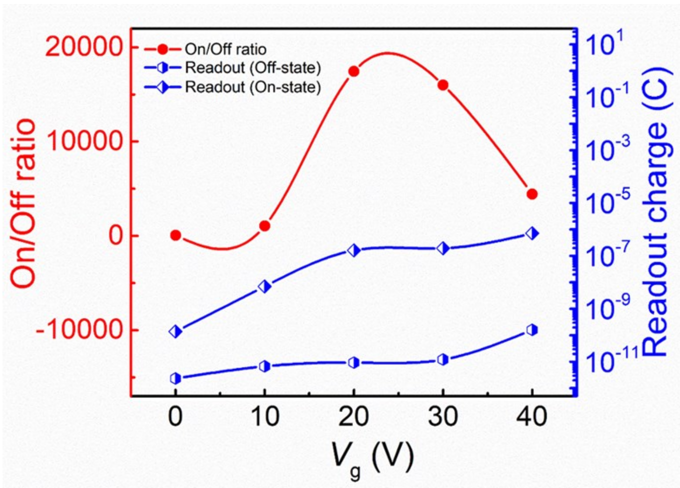
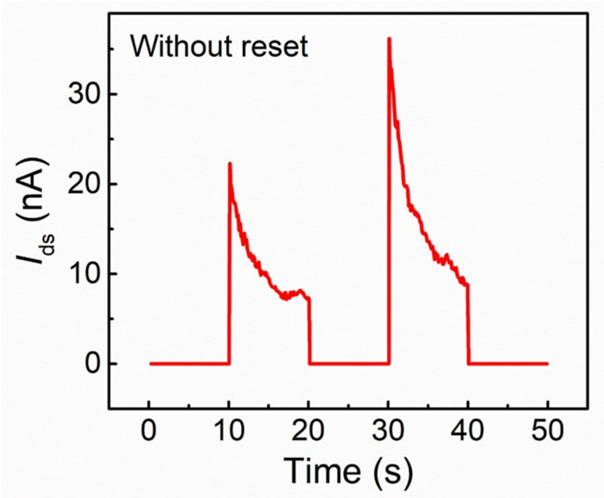


Fig. S11 Readout charge for the On/Off-states and On/Off ratio as a function of gate voltage.



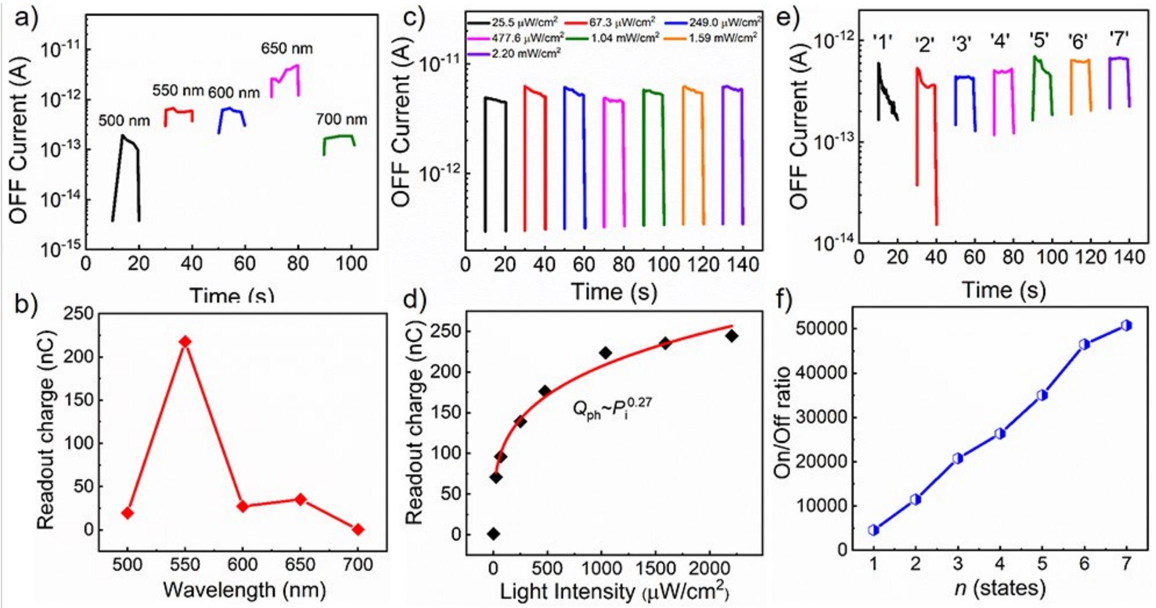


Fig. S12 Optical memory operation without reset operation.

Figure S13 (a) The Off-state current of hydrophilic MoS_2 FET under different light wavelengths. (b) The extracted readout charge obtained by integrating the readout current for 10 s with increasing light wavelength. (c) The Off-state current of hydrophilic MoS_2 FET with different light intensities. (d) The extracted readout charge obtained by integrating the readout current for 10 s with increasing light intensity. (e) The Off-state current of hydrophilic MoS_2 FET with different numbers of laser pulses. (f) The collected readout charge in the on-state recorded for 10 s. The line of best fit (red) indicates that the charge increases by 20 nC for each optical laser pulse.

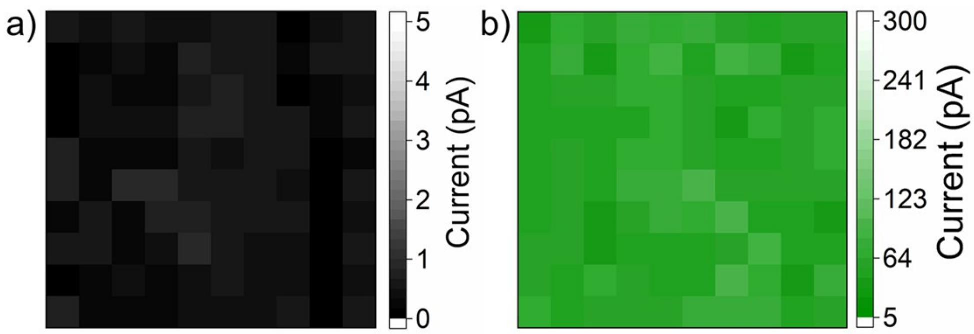


Figure S14 The uniformities of MoS_2 neuromorphic vision. (a) An illustration of the image of Off-state current at 100 mV without mask. (b) An illustration of the image memory under 550 nm laser with a power of 1.04 mW/cm^2 at 100 mV without mask. The light exposure time was 10 s, and the readout bias was applied after waiting time of 10 s.

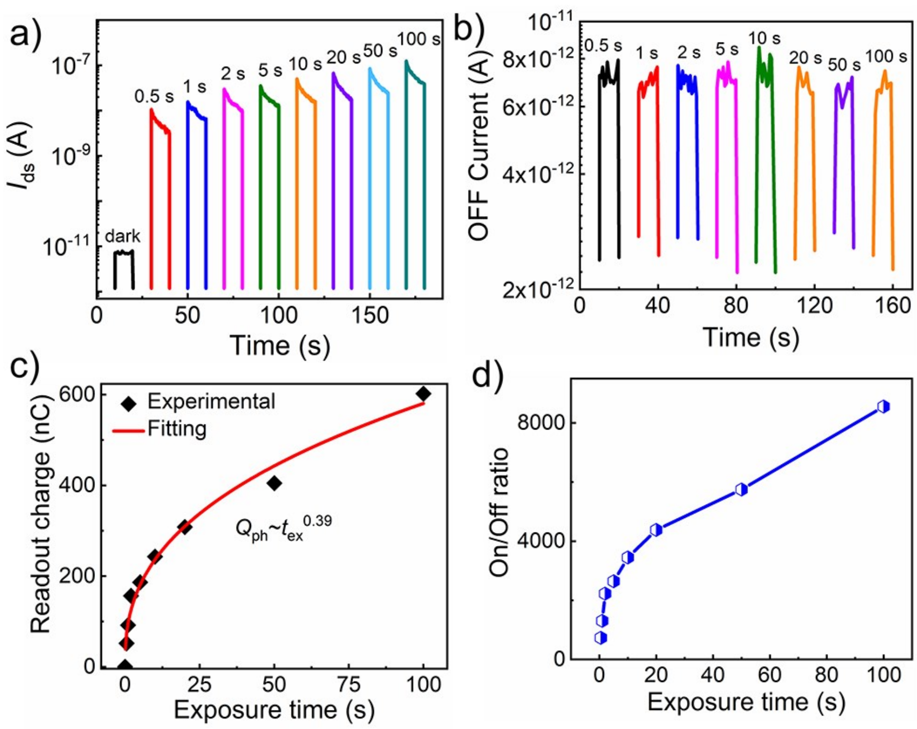


Fig. S15 Photoresponse characteristics for different exposure times. (a) The readout current as a function of light exposure times. The plots were separated with an interval of 10 s, regardless of the time that was measured initially. And the readout bias was applied after a 3 s waiting time. (b) The Off-state current of hydrophilic MoS_2 FET under different exposure time. (c) The extracted readout charge obtained by integrating the readout current for 10 s with increasing light exposure times. (d) The On/Off readout charge ratio as a function of light exposure times.

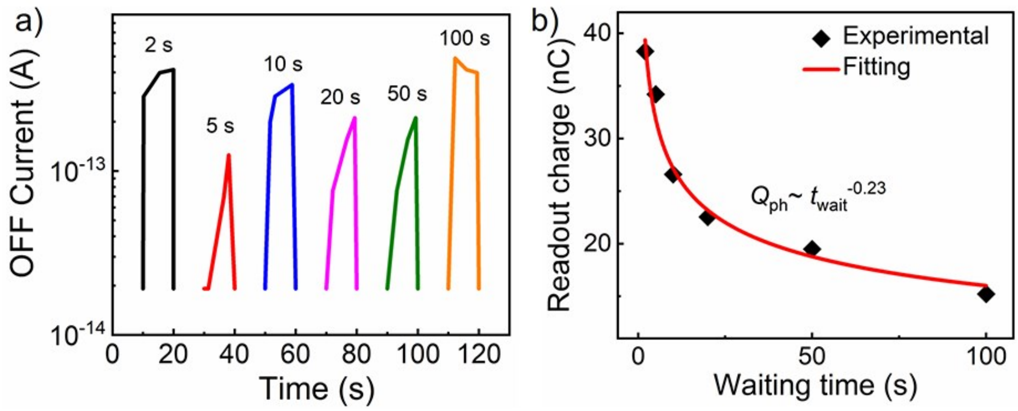


Figure S16 Photoresponse characteristics for different waiting time. (a) The Off-state current of hydrophilic MoS_2 FET under different waiting time. (b) The extracted readout charge obtained by integrating the readout current for 10 s with increasing waiting time.