

## Supplementary Information

### Photo-induced oxygen vacancy modulation in solution-processed $\text{TiO}_2/\text{ZnFe}_2\text{O}_4$ heterointerface for all-oxide dual-mode neuromorphic logic memory

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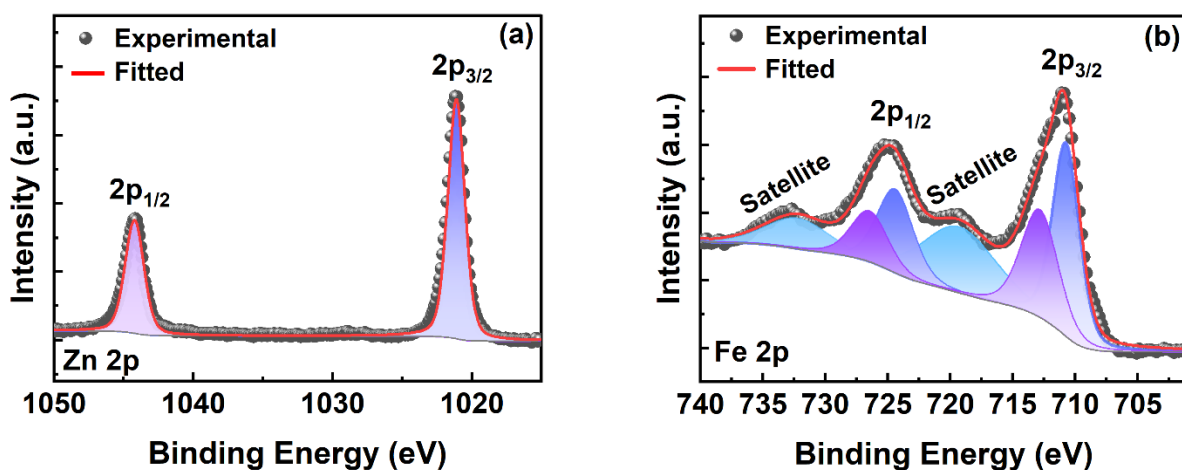


Fig. S1. XPS narrow scan spectra for (a) Zn 2p, and (b) Fe 2p.

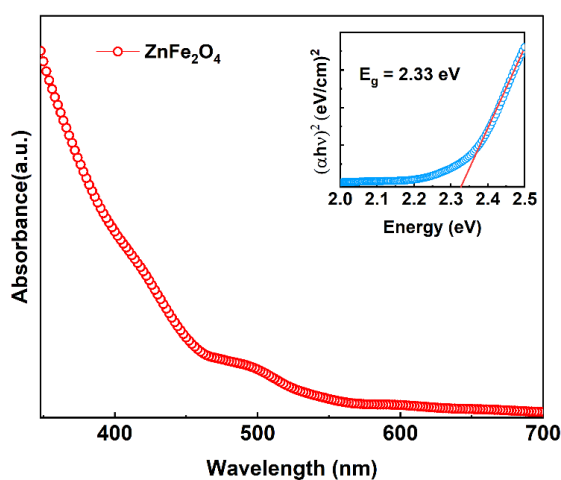


Fig. S2. UV-vis absorption spectrum and corresponding Tauc plot (inset) for ZFO film.

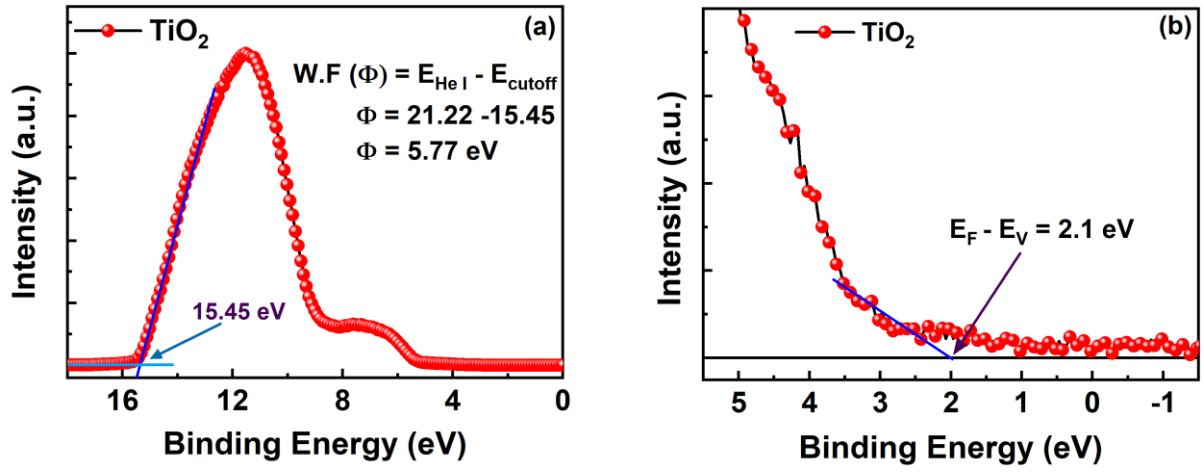


Fig. S3. (a) UPS spectrum for  $\text{TiO}_2$ , (b) valence band position of  $\text{TiO}_2$  thin film.

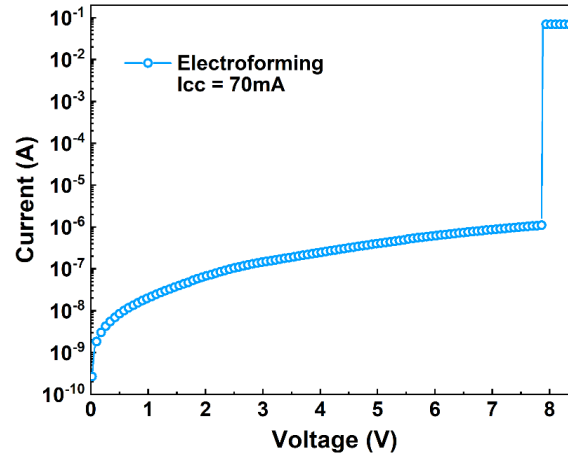


Fig. S4. Semi-logarithmic  $I$ - $V$  plot for the electroforming process in the  $\text{FTO}/\text{TiO}_2/\text{ZFO}/\text{Ag}$  device.

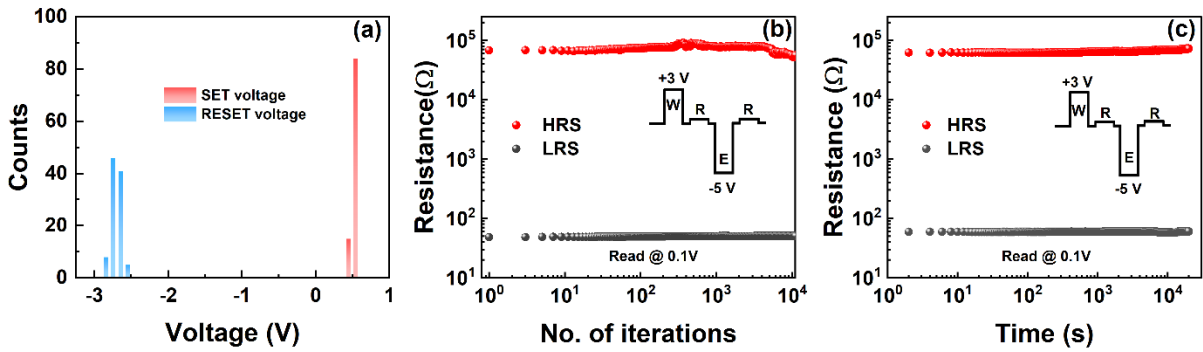
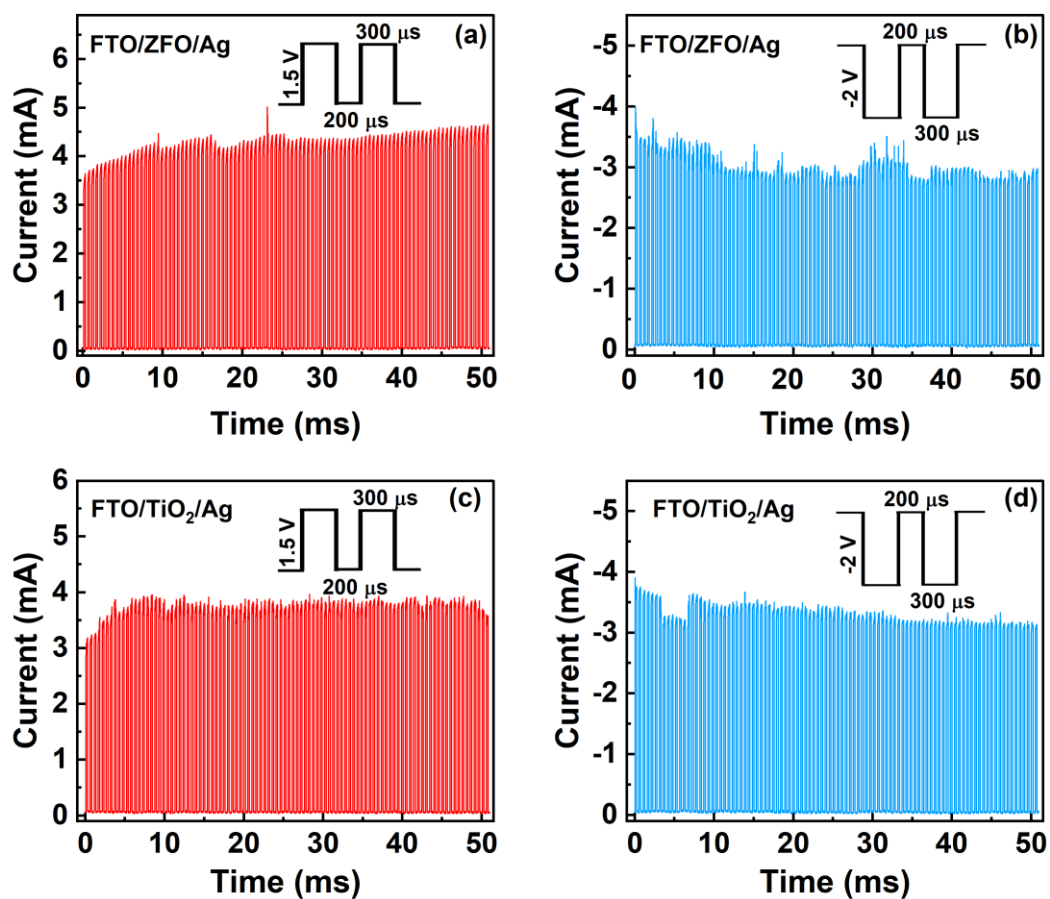
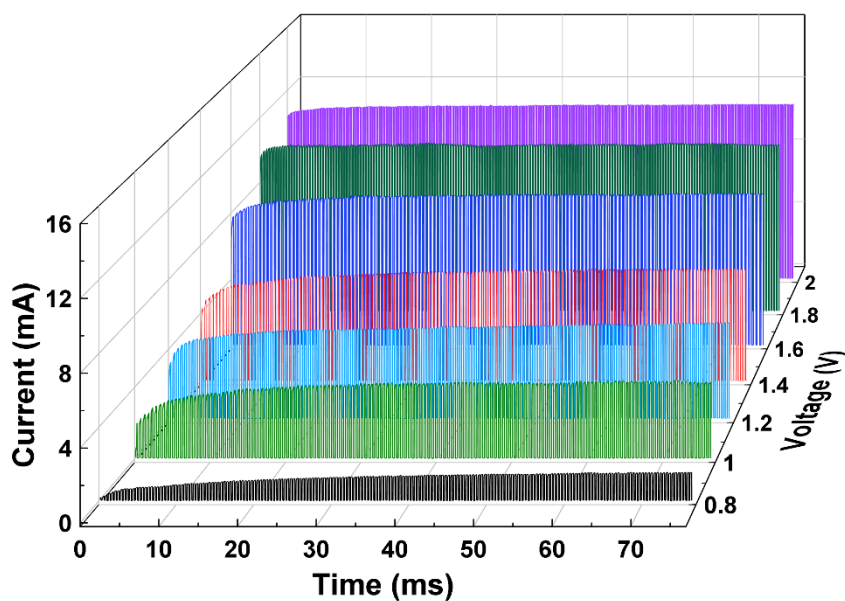


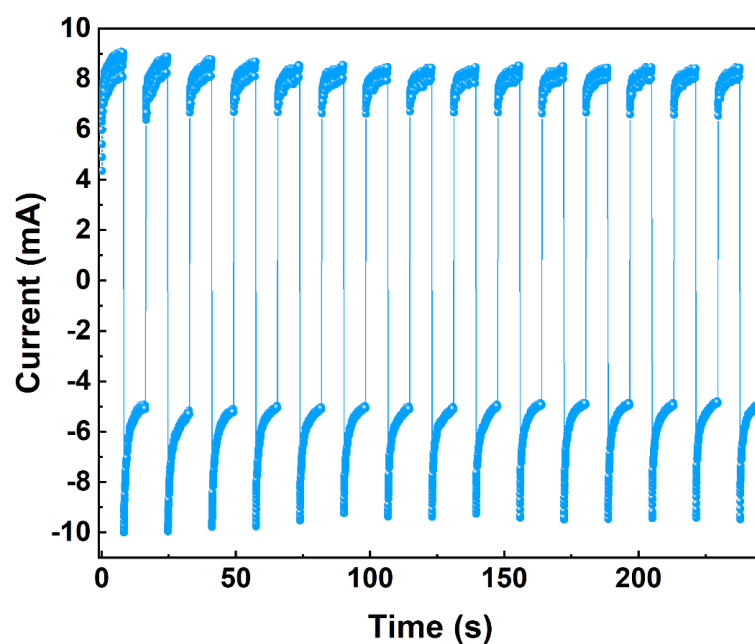
Fig. S5. (a) Distribution of SET-RESET voltages for the initial 100 cycles. (b) Endurance, and (c) retention data plot for the  $\text{FTO}/\text{TiO}_2/\text{ZFO}/\text{Ag}$  device.



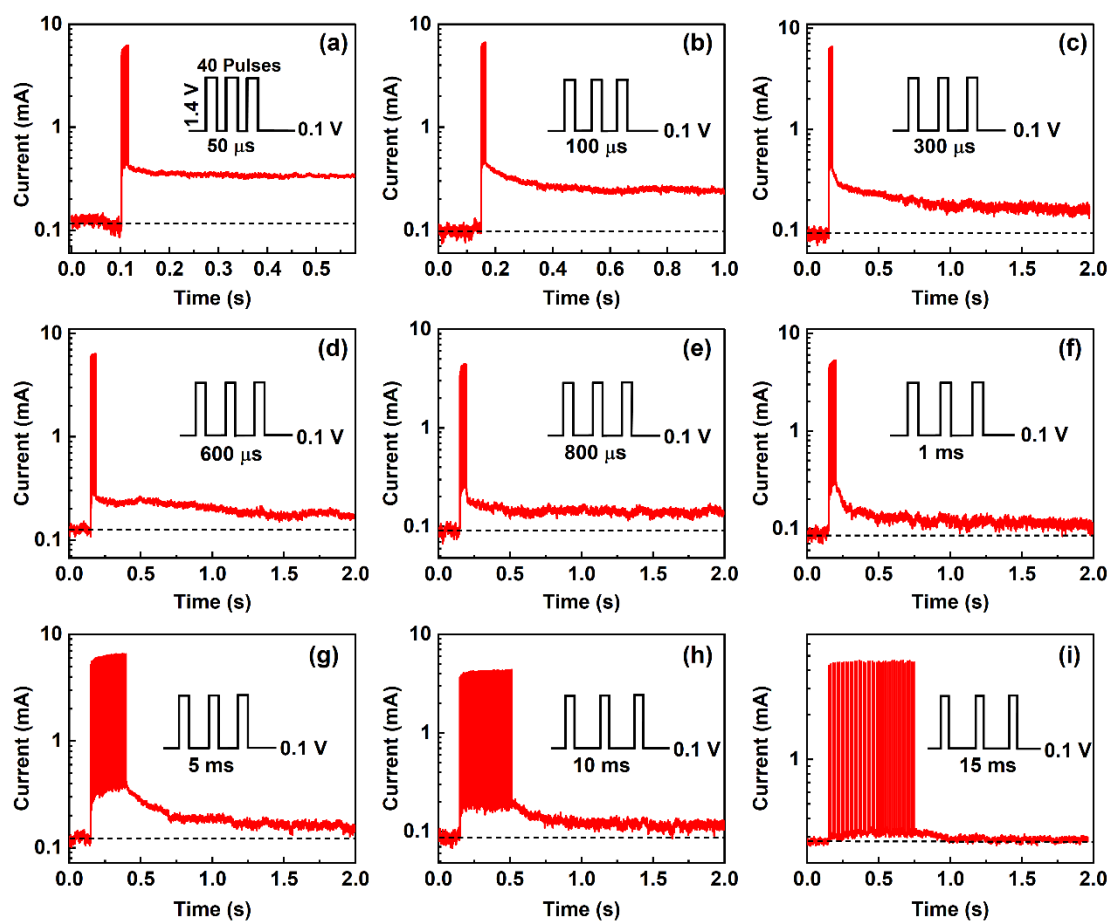
**Fig. S6.** Post-synaptic current response in FTO/ZFO/Ag device for 150 consecutive (a) positive (+1.5 V) and (b) negative (-2 V) pulse amplitudes. Post-synaptic current response in FTO/TiO<sub>2</sub>/Ag device for 150 consecutive (c) positive (+1.5 V) and (d) negative (-2 V) pulses.



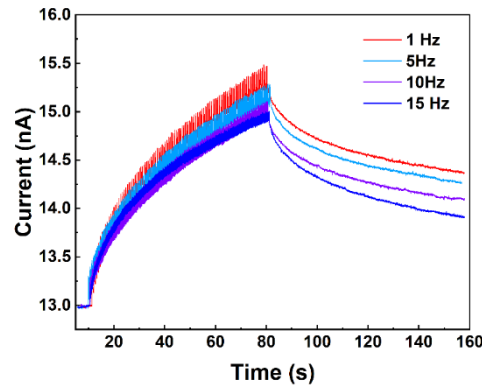
**Fig. S7.** Synaptic potentiation response of the FTO/TiO<sub>2</sub>/ZFO/Ag device corresponding to varying pulse amplitudes ranging from 0.8 V to 2 V, with a pulse width of 500  $\mu$ s and a 200  $\mu$ s pulse interval.



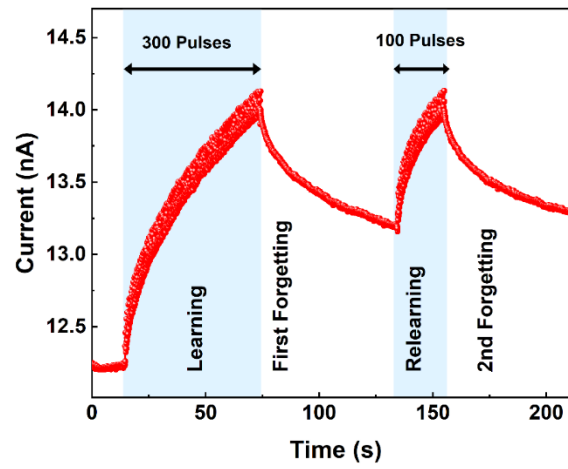
**Fig. S8.** Current modulation under 20 positive (1.4 V) and 20 negative (-1.8 V) pulses for 15 consecutive cycles.



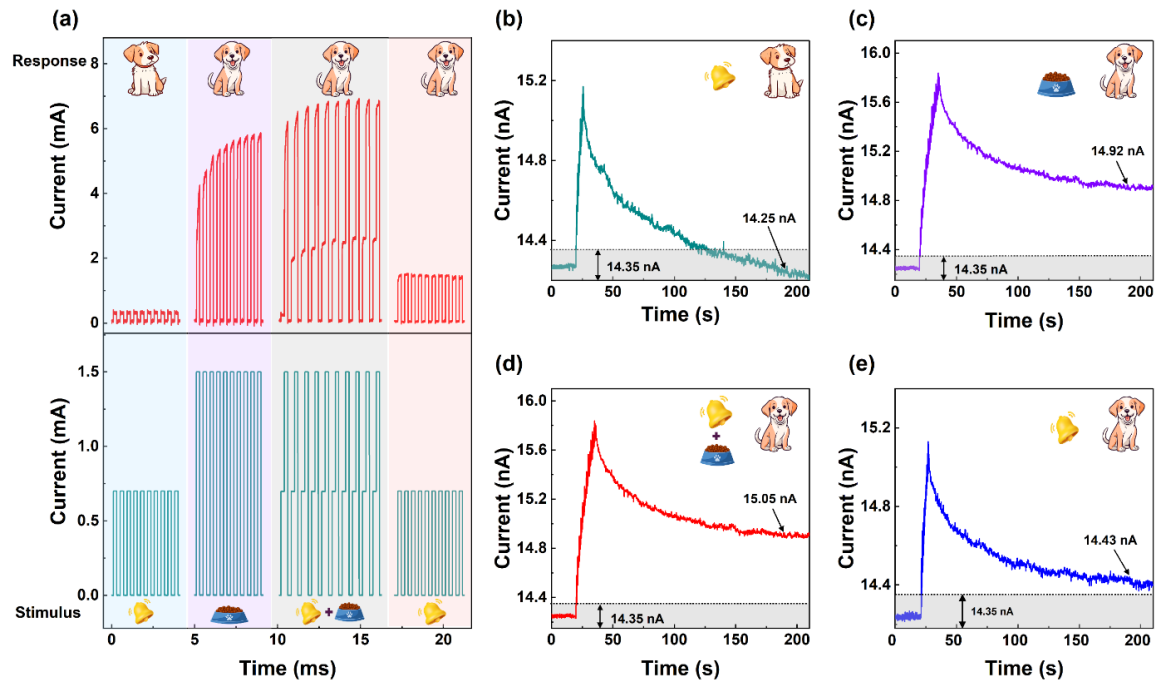
**Fig. S9.** SRDP behavior with varying pulse interval/rate shows a decrease in EPSC for longer pulse durations due to enough charge relaxation time between consecutive pulses. The inset shows the pulse parameters for device testing.



**Fig. S10.** EPSC behavior of the device decreases as the frequency of light pulses increases.

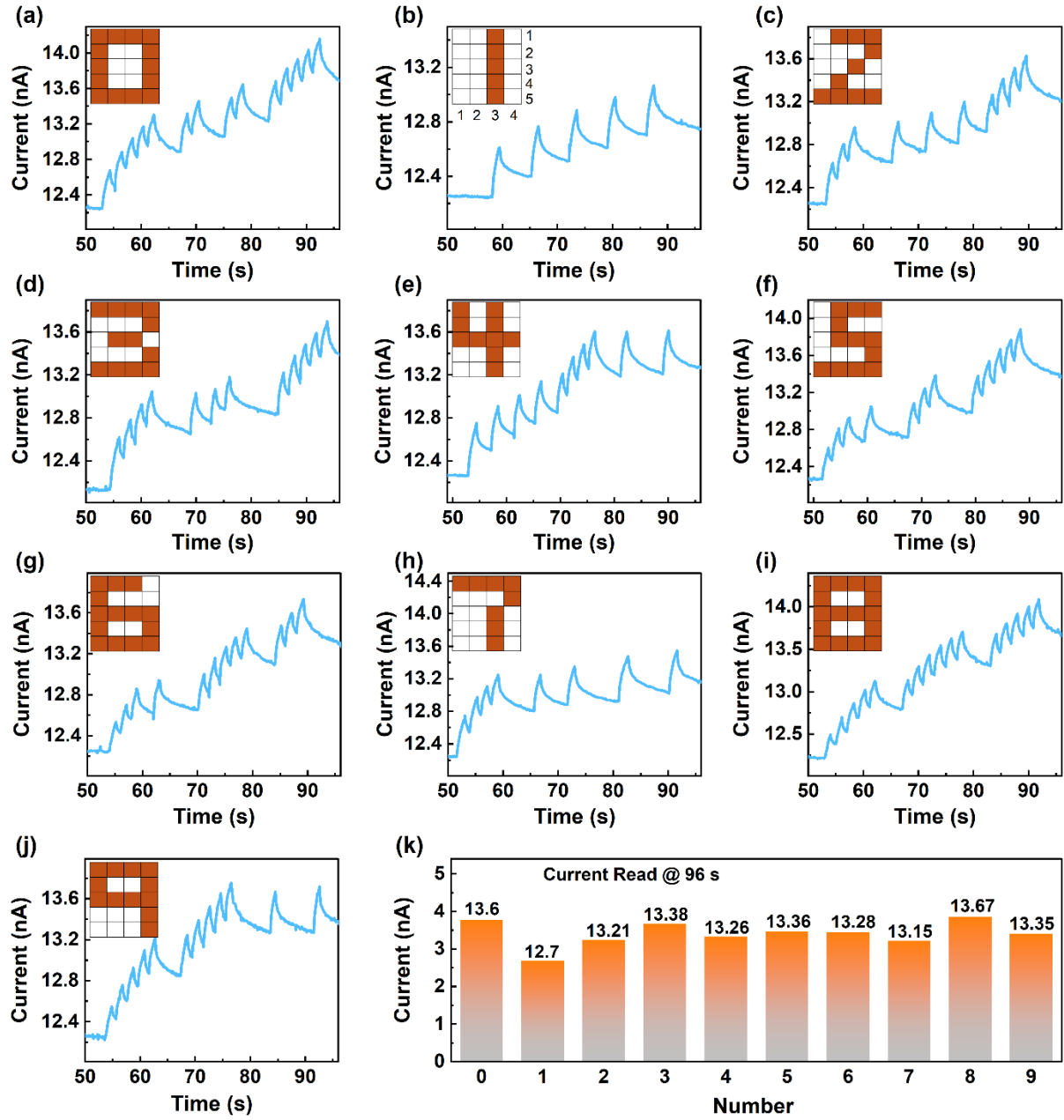


**Fig. S11.** Change of EPSC confirms optical pulse-induced simulation of learning, forgetting, and relearning behavior under 445 nm/10 mW cm<sup>-2</sup> illumination.



**Fig. S12.** Pavlov's classical conditioning under (a) electrical and (b-e) optical stimuli.

## Reservoir Computing



**Fig. S13.** EPSC current measurement for digit pattern from 0 to 9.

**Note:** Here, a reservoir computing system is described where the input layer is formed by processed images of digits 0-9, each represented as a  $4 \times 5$  pixel matrix and then converted into a  $1 \times 20$  pixel sequence, as illustrated in Figure S13. Each digit image is transformed into a linear sequence of temporal optoelectronic pulses, for example, "1110100011111001" for digit "6". Here, "1" denotes an optical ON/OFF pulse with a duration of 1s/1 s, while "0" corresponds to a 2 s OFF period, all applied at a read voltage of 0.1 V.