Supplementary Information

Optoelectronic Stimuli-Driven Switchable Memristor with Multilevel Resistance States for Neuromorphic Vision Sensors

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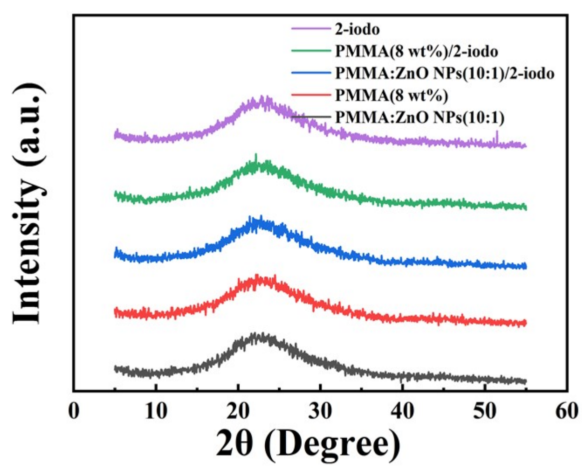

Figure S1. XRD pattern of PMMA/ZnO/2IF and pristine (PMMA/ZnO) film with 2θ vs. intensity plot. As shown in Figure S1, XRD examinations were conducted, and as shown in Figure, the prepared PMMA/ZnO/2IF memristors exhibit an amorphous nature.

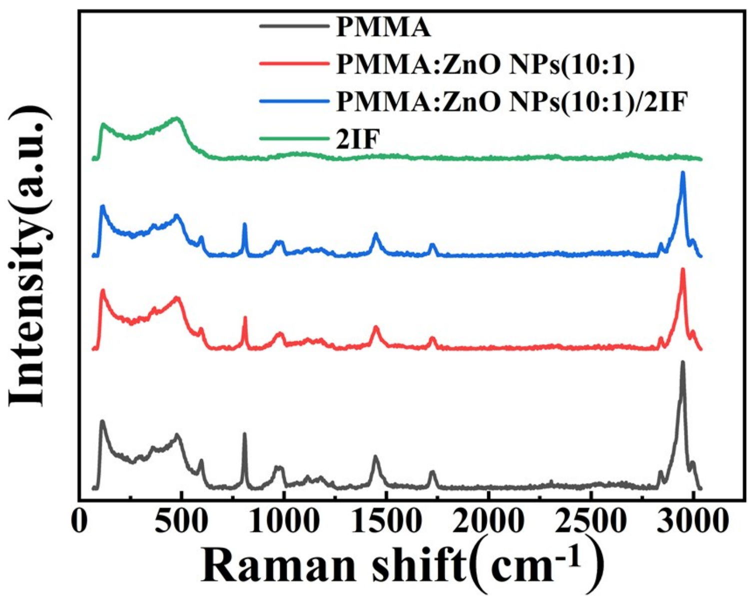

Figure S2. Raman spectra for PMMA, PMMA:ZNO, PMMA/ZnO/2IF and 2IF on a glass substrate.

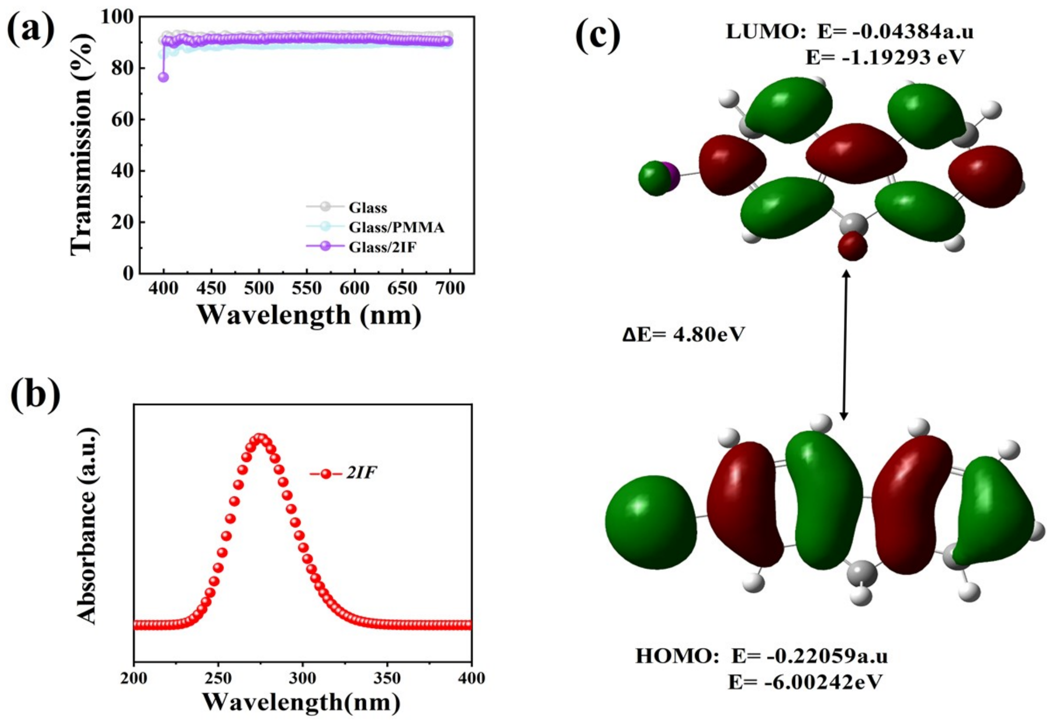

Figure S3. a) Transmittance spectra. b) UV absorbance spectrum for the 2IF material measured by density functional theory (DFT). C) HOMO-LUMO energy gap for 2IF materials. As shown in Figure S3a, the prepared 2IF material has high transmission at the visible range and strong absorbance at the UV region, as shown in Figure S3b. The 2IF martial shows strong UV absorbance and a wider bandgap. As a consequence, the HOMO-LUMO energy gap has been calculated.

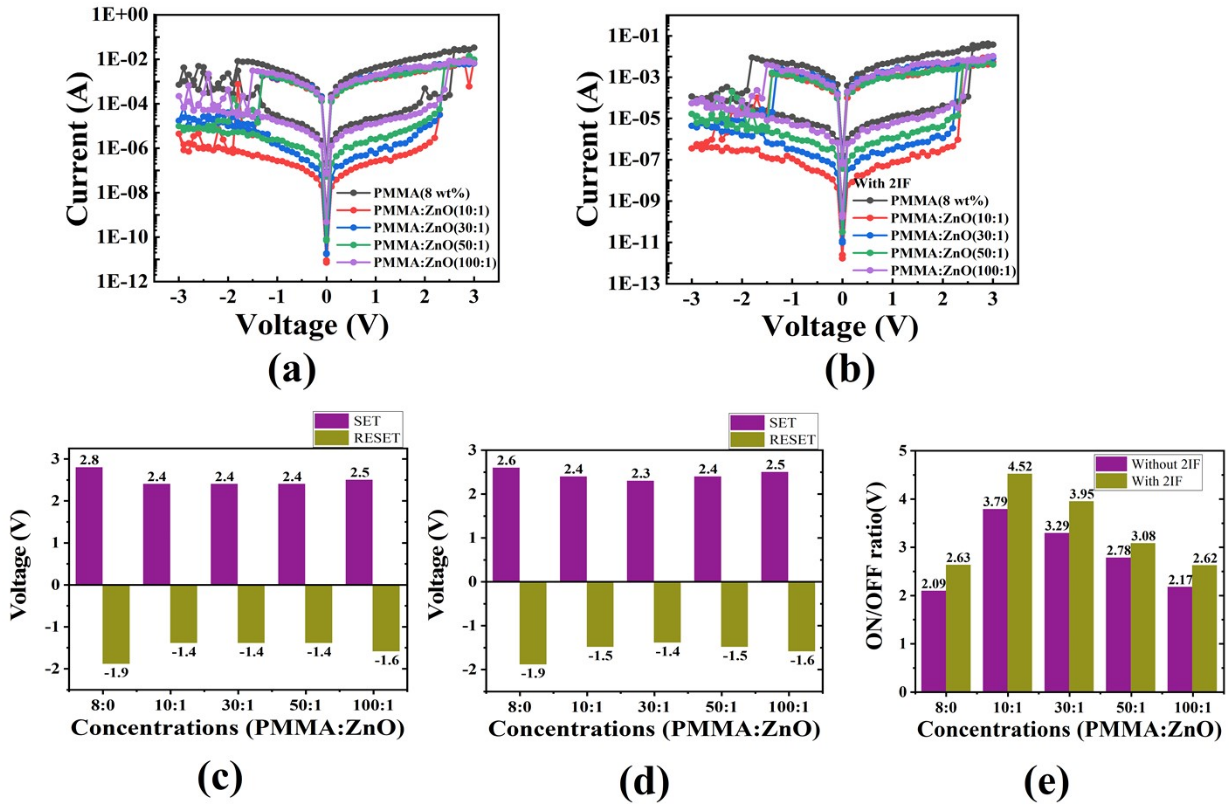

Figure S4. a) Electrical characteristics: the I–V characteristics of various PMMA/ZnO device concentrations, b) Electrical characteristics: the I–V characteristics of various PMMA/ZnO/2IF device concentrations, c). Distributions of set/reset voltages during electrical switching cycles for PMMA/ZnO devices. d) Distributions of set/reset voltages during electrical switching cycles for PMMA/ZnO/2IF devices. e) Corresponding ON/OFF ratio. The device exhibits bipolar switching behavior with sudden SET and RESET. The PMMA/ZnO concentrations have been adjusted to 10:1, 30:1, 50:1, and 100:1, respectively. The on/off ratio, set values, and reset values are all improved at this 10:1 concentration. However, the device's irregular and abrupt switching nature limits the scope of potential neuromorphic computing applications.

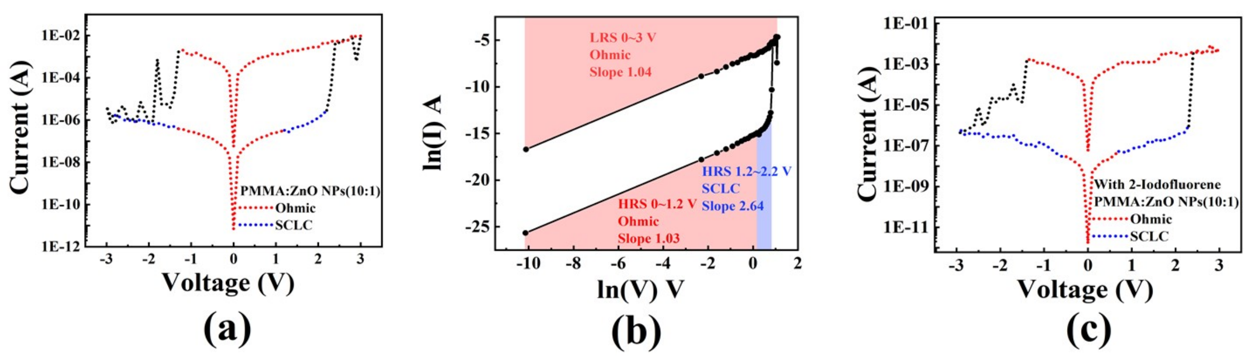

Figure S5. a) The electrical resistive switching characteristic of the memristor for PMMA/ZnO device. b) The fitted I-V curve of the set process of the memristor. C) The electrical resistive switching characteristic of the memristor for PMMA/ZnO/2IF device.

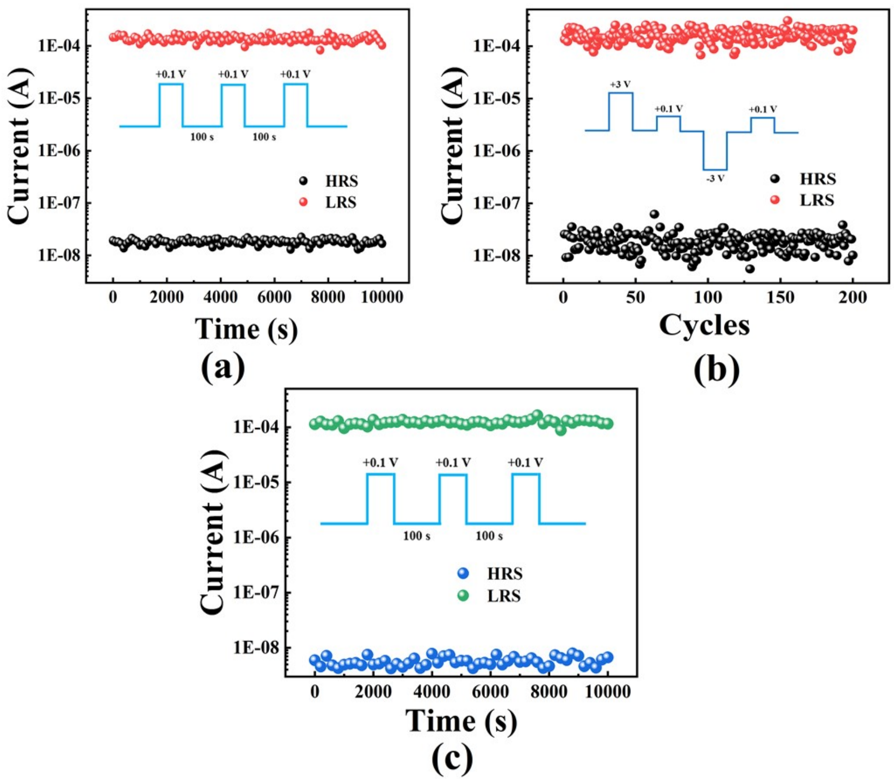

Figure S6. Figure S6. a) Retention test of memristor for 10000 cycles PMMA/ZnO. b) Electrical reliability test of memristor for over 10^3s for PMMA/ZnO device. The device's DC endurance degrades after 100 cycles. The random growth and rupture of the CF during continuous switching cycles cause the device's unstable I-V characteristics and endurance breakdown[28]. Although the PMMA/ZnO device advances SET and RESET behavior and enables neuromorphic computing, its limited durability (200 cycles) continues to be a barrier for neuromorphic applications, as demonstrated in Figure SI. The PMMA/ZnO device must be improved further for artificial synaptic applications. To improve the PMMA/ZnO device, we employed 2IF as an interface layer between the ZnO and the Ag layer. By adding the 2IF layer, the device can drastically reduce the set/reset voltage and the huge ON/OFF ratio and enhance the low/high resistance state (LRS/HRS) ratio. (c) Retention test of memristor for 10000 cycles PMMA/ZnO/2IF in dark mode.

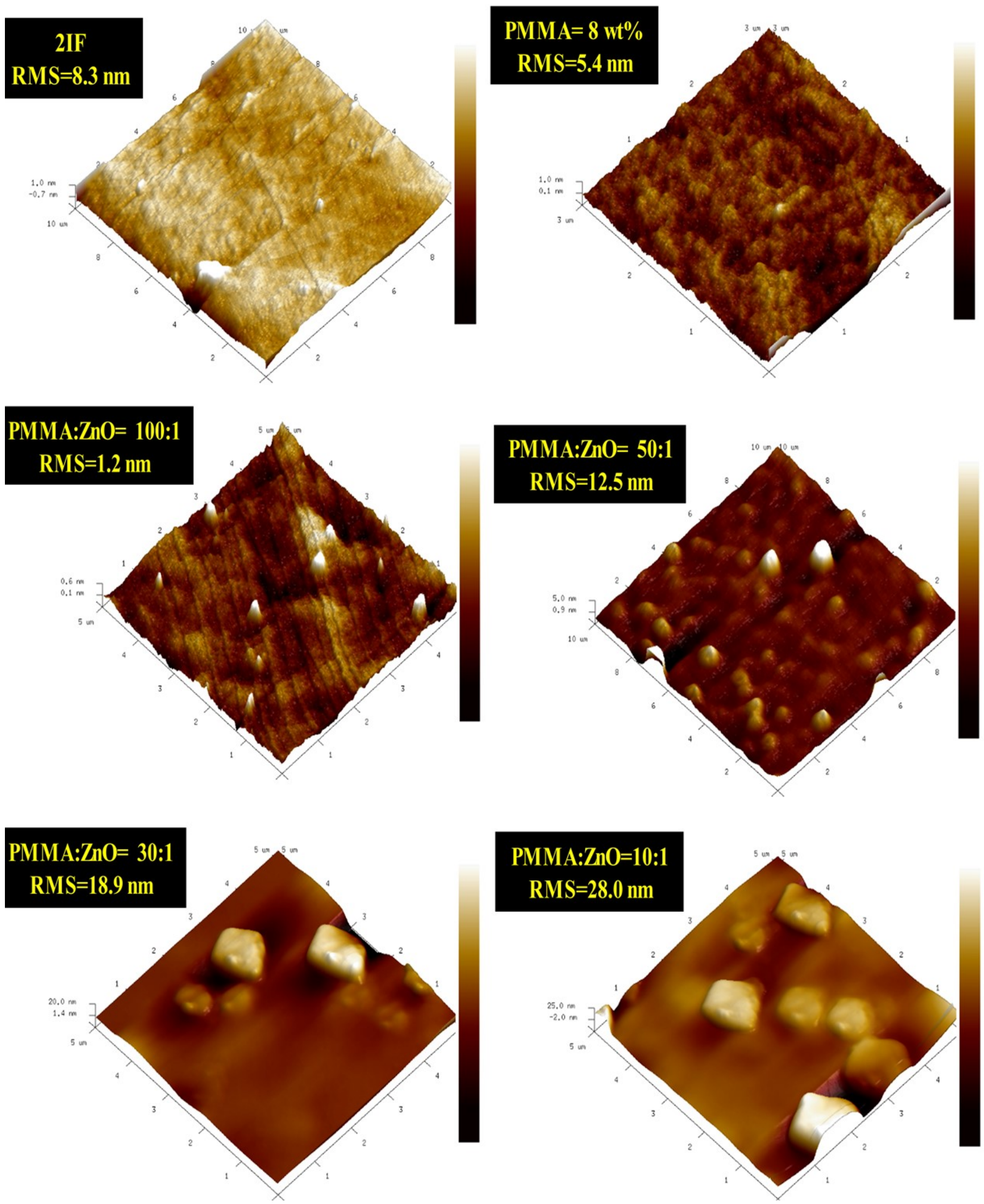

Figure S7: The AFM images of PMMA/ZnO/2IF layer top surface with different conditions within 5 μm × 5 μm.

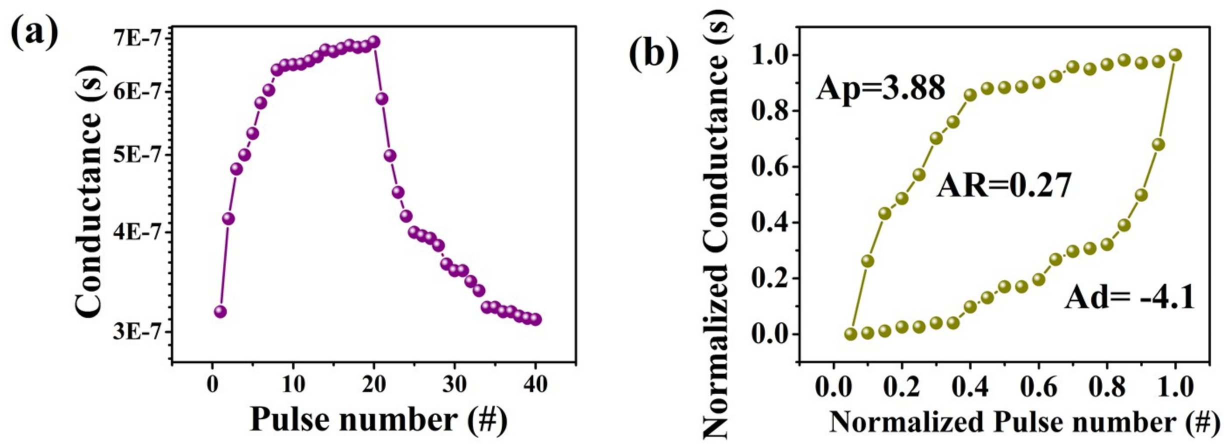

Figure S8. a) Long-term potentiation and long-term depression of the device with pulse numbers.

b) Nonlinear values of potentiation and depression of the device.

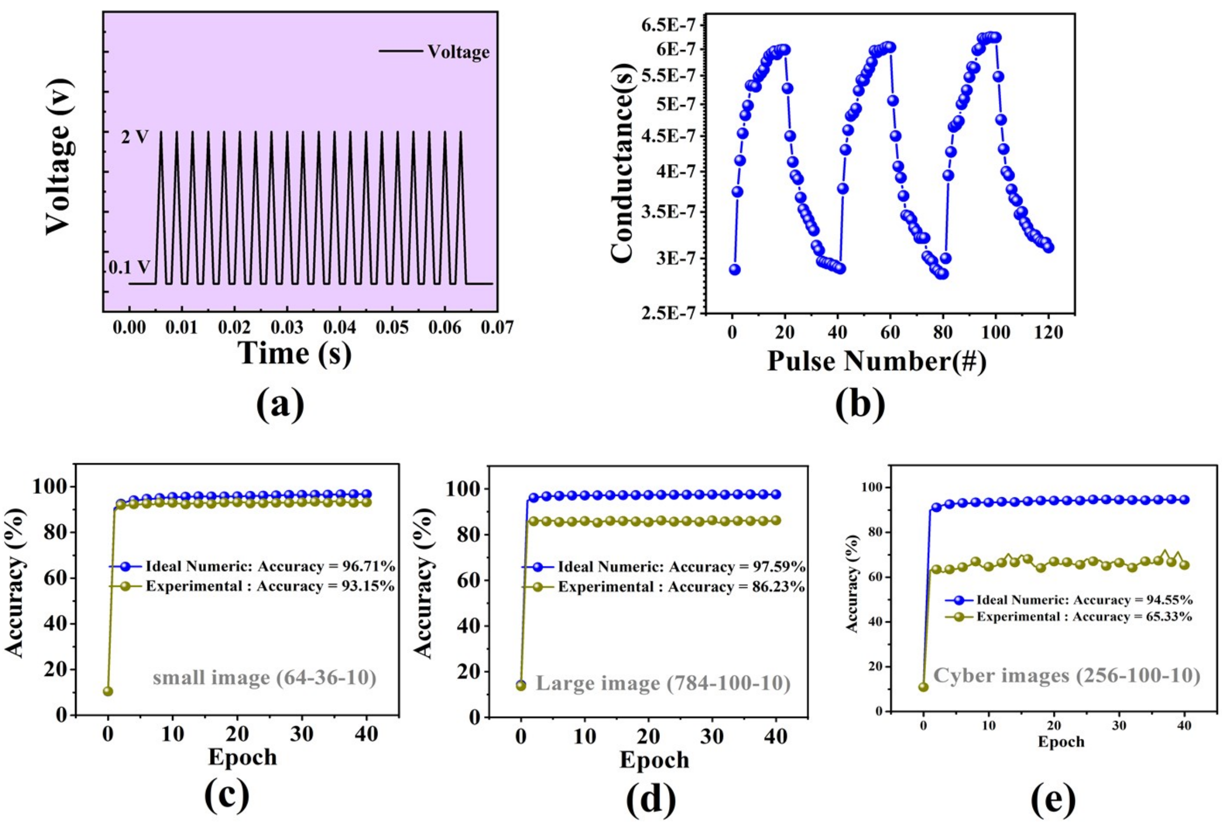

Figure S9: a) Electrical pulses applied on the optoelectronic synaptic device for LTP/LTD emulation. b) Electrical pulses were applied on the optoelectronic synaptic device for LTP/LTD emulation by optoelectronic artificial synapse. c-e) Recognition accuracy evolution with training epochs for small and large handwritten digits and cyber images for PMMA/ZnO2IF.

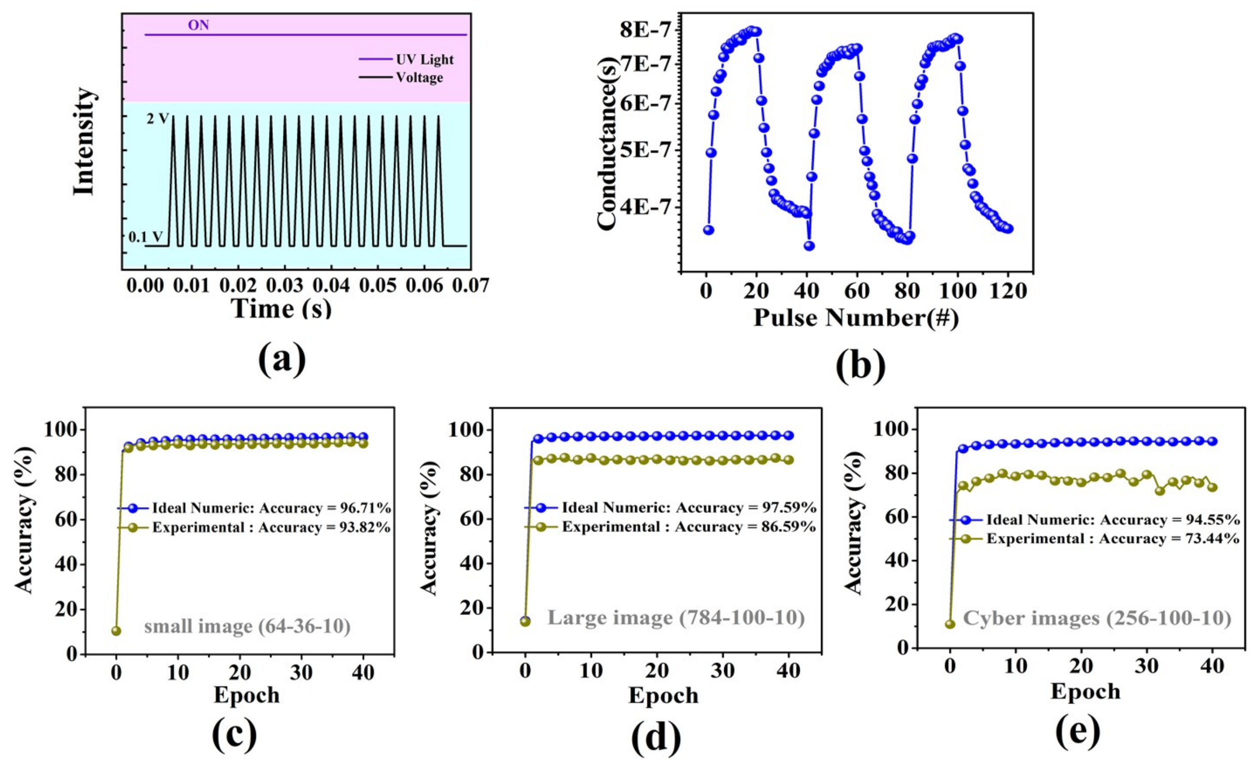

Figure S10: a) Optical and electrical pulses applied on the optoelectronic synaptic device for LTP/LTD emulation. b) Optical-electrical pulses were applied on the optoelectronic synaptic device for LTP/LTD emulation by optoelectronic artificial synapse. c-e) Recognition accuracy evolution with training epochs for small and large handwritten digits and cyber images for PMMA/ZnO2IF.

Table S1: Optoelectronic resistive switching properties of memristors with and without UV light set, rest, and ON/OFF ratio.

sample	**Without light**	**UV light**
Set voltage (V)	2.4	1.2
Reset voltage (V)	-1.5	-0.8
ON/OFF ratio (Read=0.2V)	4.52	5.40

References

1. T.-L. Tsai, H.-Y. Chang, J. J.-C. Lou and T.-Y. Tseng, *Appl. Phys. Lett.*, 2016, **108**, 153505.