Memristors Based on Multilayer Graphene Electrodes for Implementing a Low-power Neuromorphic Electronic Synapse

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1.The current-voltage(I-V) sweeps of 100 cycles.

In this work, we test the I-V with the $Ta/Ta_2O_5/AlN$/Graphene device and logarithmically to the ordinate current, black arrow indicates the direction of voltage scanning. The data is shown in Fig. S1(a-b).

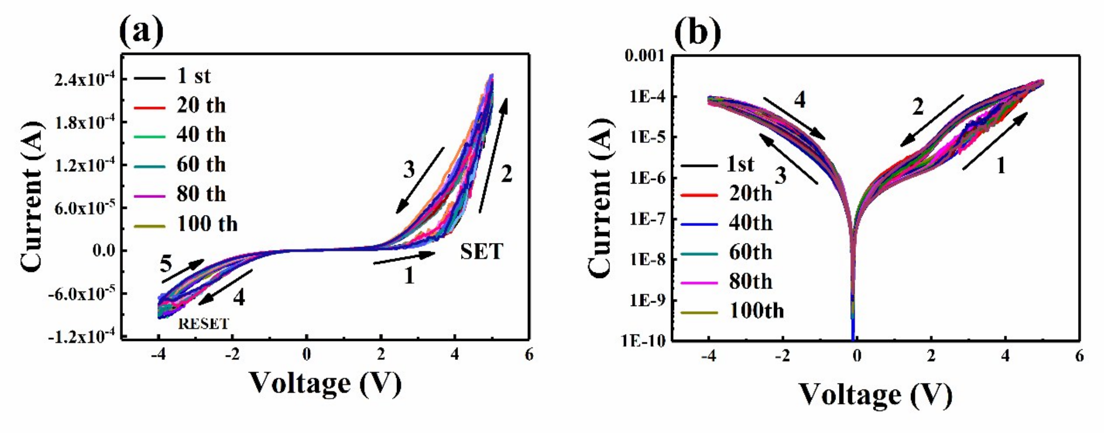

Fig. S1 (a)100 cycles typical I-V sweep for $Ta/Ta_2O_5/AlN$/Graphene device structures. (b)I-V curve after the logarithm of the current.

2. Spiking-time-dependent plasticity (STDP) and Pulse regulation

In neuroscience, synaptic weight is called synaptic plasticity.[1] The top electrode (TE) of the memristor is considered to be the presynaptic membrane, while the bottom electrode (BE) mimics the postsynaptic membrane, which changes synaptic weight by changing the interval between presynaptic and postsynaptic spikes. The phenomenon is called STDP. STDP is a very important factor in neurological learning and memory. If the presynaptic triggers before the postsynaptic, the synaptic weight increases, and the strength of the connection between the two neurons increases. Conversely, if the postsynaptic is triggered before the presynaptic, the synaptic weight is reduced, and the strength of the connection between the two neurons is decreased. Here, we apply a pair of pulses with a voltage of 1 V/-1 V to simulate the application of presynaptic and postsynaptic to the TE and BE, as shown in Fig. S4(b-c). The relative timing (Δt) is defined as the time interval from the end of the presynaptic (postsynaptic) spike to the beginning of the postsynaptic (presynaptic) spike. Fig. S2(a) shows the relative change in synaptic weight (ΔW) versus Δt, where ΔW is defined as $((G_2-G_1)/G_1) \times 100\%$, G_1

and G_2 represents the conductance values before and after the voltage pulse, respectively. The red curve in Fig. S2(a) is fitted by the following equation:[2]

$$\Delta W = \begin{cases} A_+ e^{-\Delta t/\tau_+}, & \Delta t > 0 \\ A_- e^{-\Delta t/\tau_-}, & \Delta t < 0 \end{cases} \tag{1}$$

Where A_+ and A_- are fitting parameters, and τ_+ and τ_- are time constants. STDP is the most important Hebbian learning rule for learning and memory, [3,4] and experimental data shows that the memristor follows asymmetric Hebbian learning rules. The increase and decrease of conductivity represent the excitability and inhibition of synapses, respectively. We obtained the application of 30 consecutive pulses of positive voltage of 2 V and 30 pulses of continuous negative voltage of -2 V to the device. As shown in Fig. S2(b), it can be obtained that the potentiation or depression of conductance changes less and less as the number of pulses increases.[5,6]

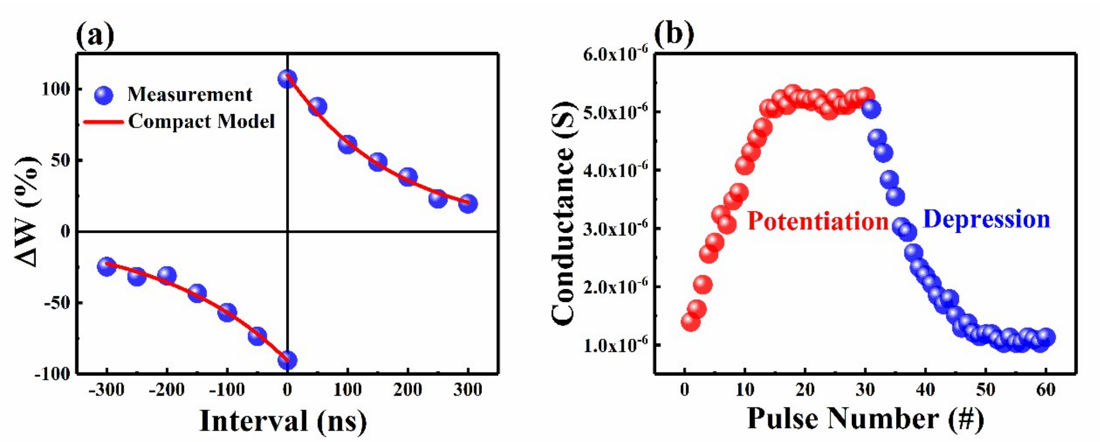

Fig. S2 (a) Simulation of Asymmetric Hebbian Learning Rules STDP. (Where the red line is the result of fitting by equation (1)). (b) Potentiation (1st pulse to 30th pulse) and depression (31st pulse to 60th pulse) relationship between pulse number and conductance.

3. Paired-pulse facilitation (PPF)

The pulse with amplitude of 1 V and pulse width of 100 ns is applied to the device, and the waveform is as shown in the upper part of Fig. S3(a), and the response current is obtained, as shown in the lower part of Fig. S3(a). The PPF is defined as the percentage increase of the current after the second pulse (I_2) and the current after the first pulse (I_1), expressed as $((I_2-I_1)/I_1) \times 100\%$. [7] Decreasing the Δt between the two potentiating pulses results in an increase in synaptic weight. As shown in Fig. S3(b) showing the relationship between ΔW and Δt. Among them, the red curve is fitted by the following equation: [8,9]

$$\mathrm{PPF}=C_1\exp\left(-\frac{\Delta t}{\tau_1}\right)+C_2\exp\left(-\frac{\Delta t}{\tau_2}\right) \tag{2}$$

Where C_1 and C_2 are the initial fitting parameters and τ_1 and τ_2 represent the relaxation time. For positive voltage pulses, τ_1 and τ_2 are 147 ns and 5.3 μs, respectively. If the double pulse interval is reduced, the memory effect of the preset pulse on the continuous pulse will be enhanced, which is very consistent with the biological synapse.

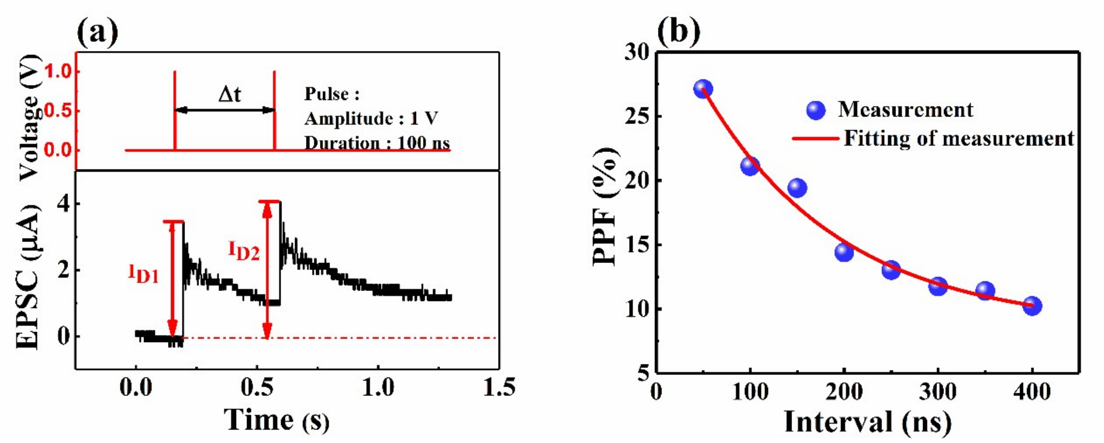

Fig. S3 (a) The EPSC is triggered by a pair of presynaptic pulses (where the pulse amplitude is 1 V, the pulse width is 50 ns and pulse interval Δt is a variable), and I_{D1} and I_{D2} are the read currents of the first and second pulses, respectively. (b) Paired-pulse facilitation (PPF). the memristor ΔW versus the relative peak time interval. (Where the red line is the result of fitting by equation (2)).

4.Schematic diagram of waveform

Fig. S4 is a schematic illustration of a series of applied pulses. Fig. S4(a) shows the continuous application of 30 forward pulses and the continuous application of 30 negative pulses (amplitude 2 V, pulse width 700 ns, interval 300 ns). As shown in Fig. S4(b-c), which represents the pre-synaptic spike and the post-synaptic spike, the Δt is defined as the time interval between the pre-synaptic spike to the post-synaptic spike. Where t>0 represents the initial of the presynaptic peak to the post-synaptic peak, and t<0 represents the initial of the post-synaptic peak to the presynaptic spike.

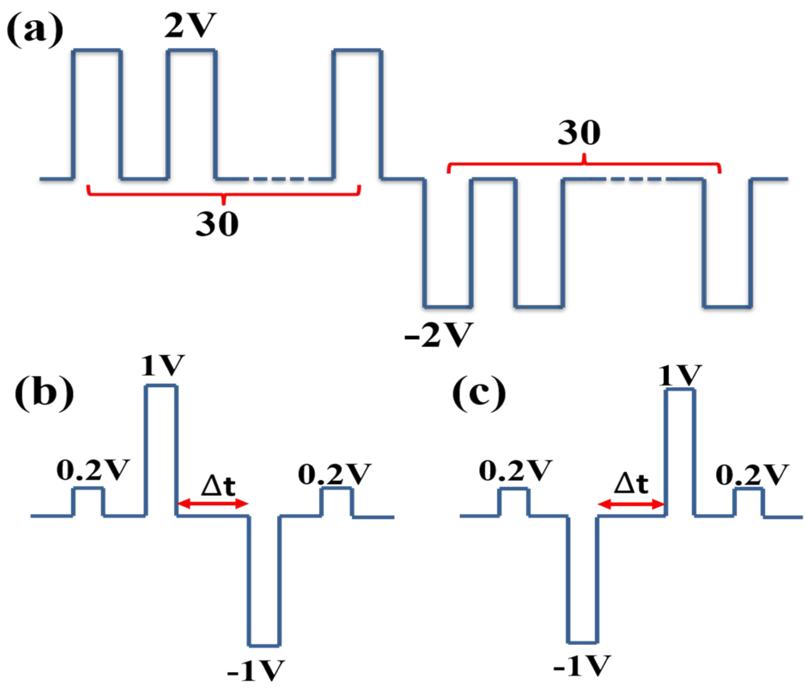

Fig. S4 (a) 30 consecutive forward pulses (2 V amplitude) and 30 negative pulses (2 V amplitude). (b-c) a pair of presynaptic and postsynaptic spikes.

5.The cycle diagram of 300 pulse tests.

Using the pulse waveform of Fig. S4(a), 30 consecutive positive pulses (2 V/ 700 ns/300 ns) and 30 consecutive negative pulses (-2 V/700 ns/300 ns) are applied to the device and then cycled five times in succession. It is as shown in Fig. S5.

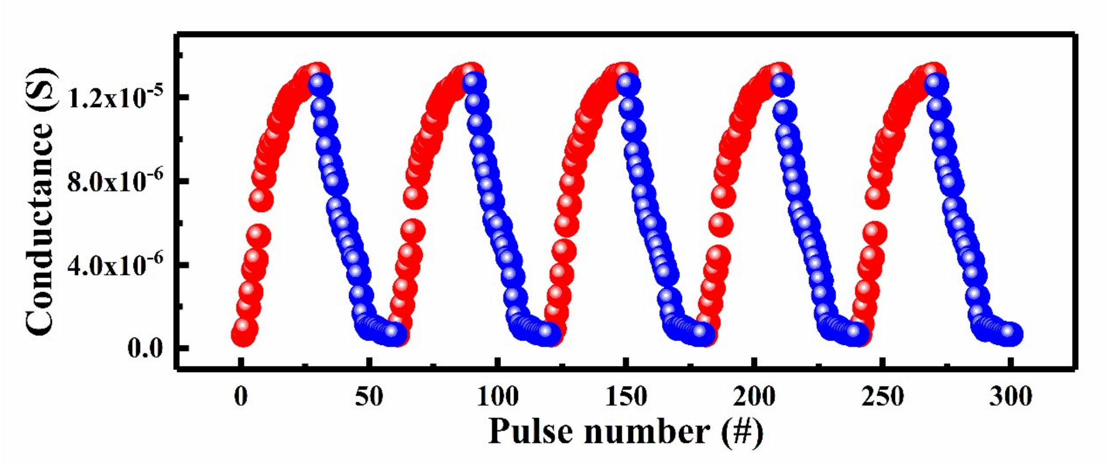

Fig. S5 Conductance change diagram obtained by applying 300 consecutive pulses to the device.

6. Pulse regulation.

Upload accuracy file by MATLAB format which describes the relationship of training times and accuracy, 77.2% is at 50th training, as shown in Fig. S6(a-c). And upload four weights maps with respect to 1st, 2nd and 50th training whose accuracy are shown in their subtitle.

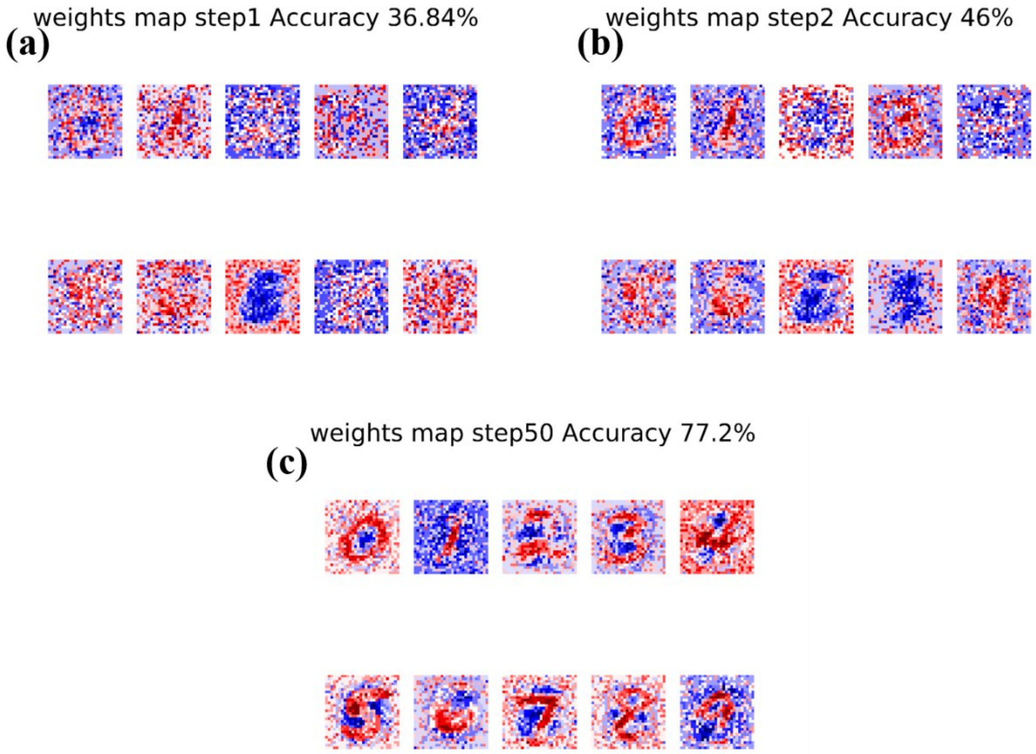

Fig. S6 Pulse regulation. (a-c) The training times are 1st,2nd,50th and the corresponding accuracy rate.

7.Energy of a write event

Fig. S7(a) shows the real-time voltage and current data plots for the device in a write event, with arrows indicating the time required to calculate the energy. It can be observed that as the number of pulses increases, the device current is gradually increasing. We calculated power consumption by the formula $P = U \times I$, and then got the relation between power and time, and finally calculated the integral of the curve by $W = \int P\, dt = \int (U \times I)\, dt$, [10] so the required modulation energy of each pulse is obtained as shown in Table S1.

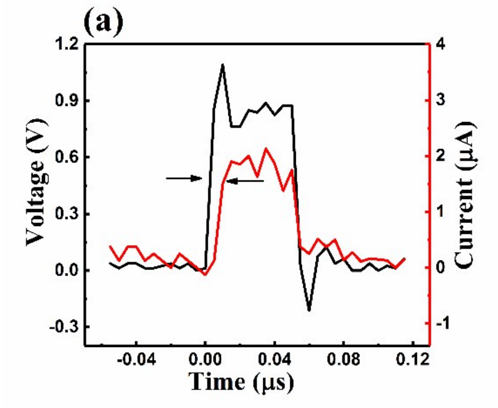

Fig. S7 (a) Power and time at one writing event.

Table S1. The energy value at one writing event

Ordinal Number of Pulse	**1st**	**2nd**	**3rd**	**4th**	**5th**	**6th**	**7th**	**8th**	**9th**	**10th**
Energy (J)	**1.99×** 10^{-14}	**2.53×** 10^{-14}	**3.11×** 10^{-14}	**3.51×** 10^{-14}	**3.71×** 10^{-14}	**3.70×** 10^{-14}	**3.72×** 10^{-14}	**3.80×** 10^{-14}	**3.79×** 10^{-14}	**3.82×** 10^{-14}

8. Short-term plasticity (STP) and Long-term plasticity (LTP).

Here, we set a pulse with an amplitude of 1 V, a pulse width of 50 ns, and an interval of 50 ns. Different numbers of stimulation pulses (10, 20, 30, 40, 50) are applied to the device, and it can be found that the synaptic weight drops. The trend is shown in Fig. S8(a-e). The relaxation time constant τ is an important factor in fitting the retention curve. As the number of pulses increases, the relaxation time τ also increases, as shown in Fig. S8(f).

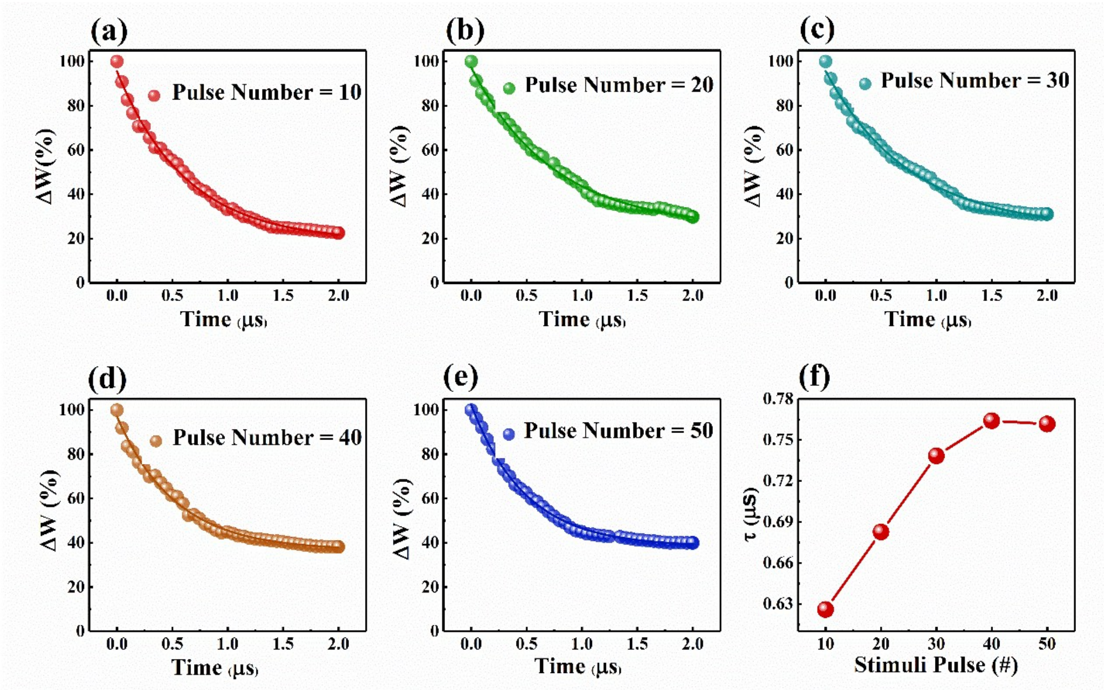

Fig. S8 Short-term plasticity and long-term plasticity. (a-e) Conversion of STP to LTP under the same pulses (The pulses amplitude, duration, and interval were set as 1 V/50 ns/50 ns) with different number. Here, the number of pulses is 10,20,30,40 and 50, respectively. The fitting result is obtained by Equation (3). (f) The relationship between the relaxation time (τ) obtained by the fitting curve of (a-e) and the number of stimulation pulses.

9. EPSC rate of change and energy consumption

The energy consumption for a single different pulse duration is defined as $I_{PEAK} \times U \times t$, I_{PEAK}, U and t represent the peak current of the EPSC, the voltage at which the device is turned on, and the duration of the pulse, respectively. We can see that as the pulse width decreases, the energy consumption decreases rapidly, as shown in Fig. S9(a). We define the rate of change of EPSC $(I-I_0)/I_0\times100\%$, where I represents the current after 50 ns pulse stimulation and the current after 1.5 μs pulse decay, and I_0 represents the initial current. As shown in Fig. S9(b), it can be seen that as the number of pulses increases, the rate of change of EPSC is significantly improved after 1.5 μs, and the conversion from STP to LTP is realized.

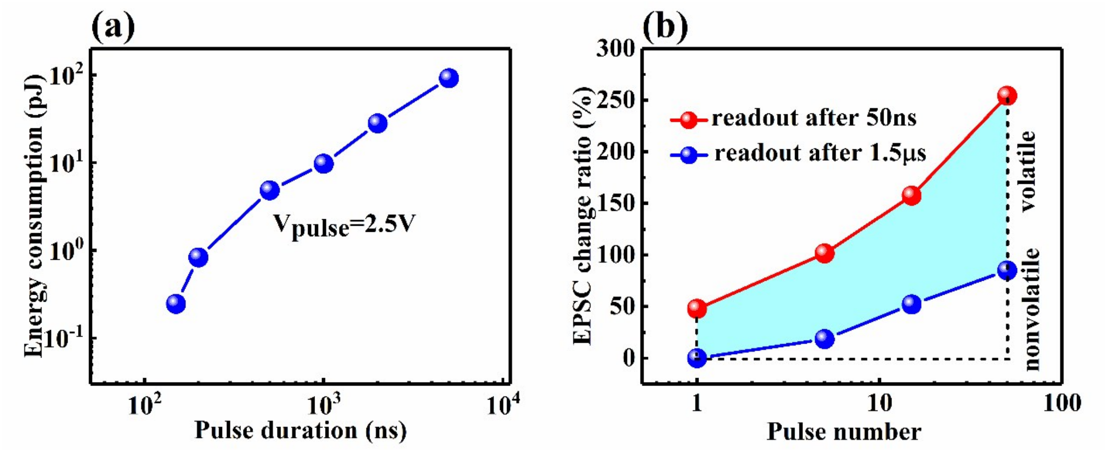

Fig. S9 (a) Energy consumption for a single pulse duration. (b) EPSC change rate under different pulse numbers.

10. "learning and forgetting experience".

Fig. S10(a-d) shows the cycle of performing two learning/forgetting.

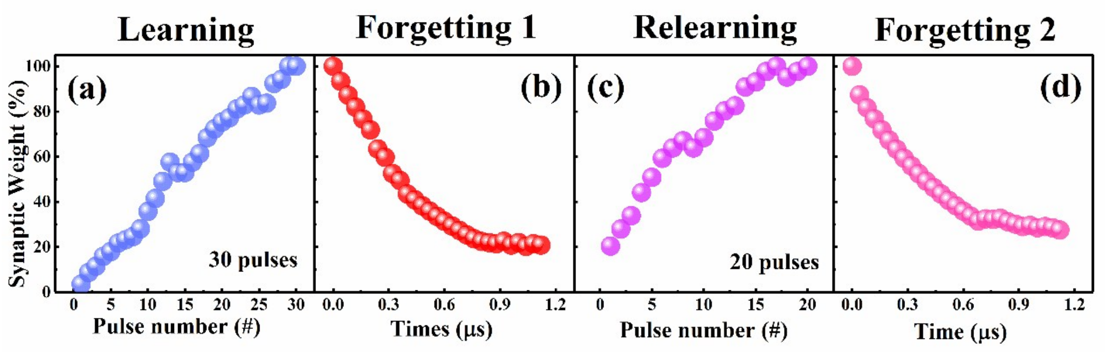

Fig. S10 Simulation of the "learning experience" behavior. (a) Continuous positive pulse stimulation for 30 consecutive spikes. (b) Spontaneous attenuation of synaptic weight. (c) Increased synaptic weight with 20 positive pulse stimulations again (d) Second synaptic weight Spontaneous decay over time.

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