

## RESEARCH ARTICLE

**HfAlO-based ferroelectric memristors for artificial synaptic plasticity**

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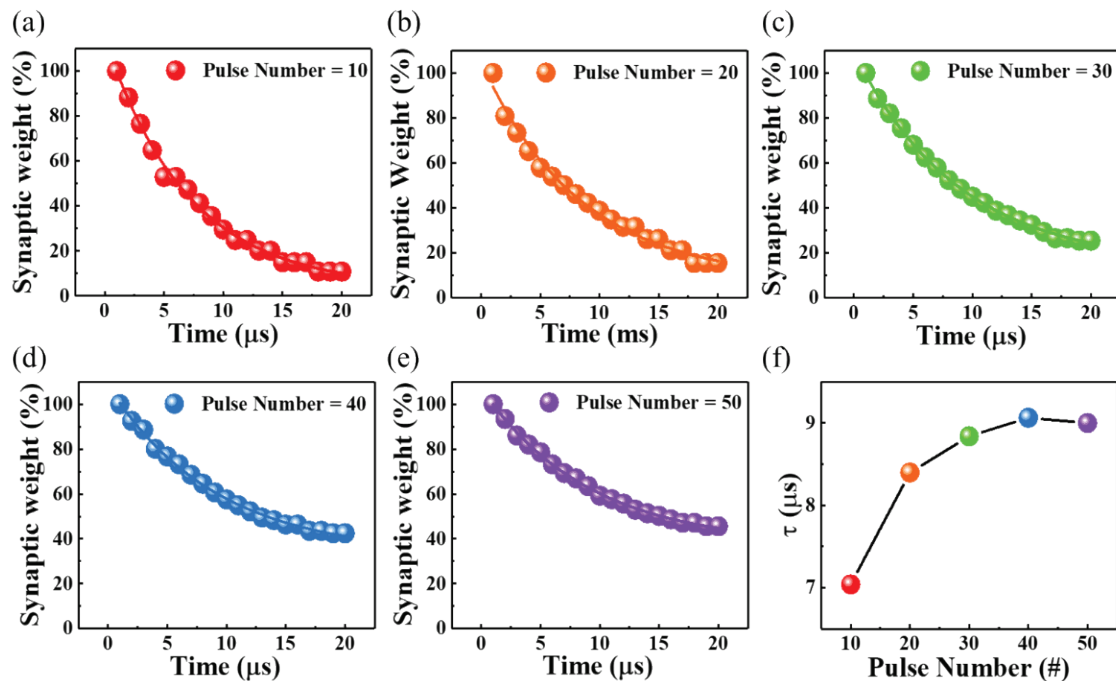
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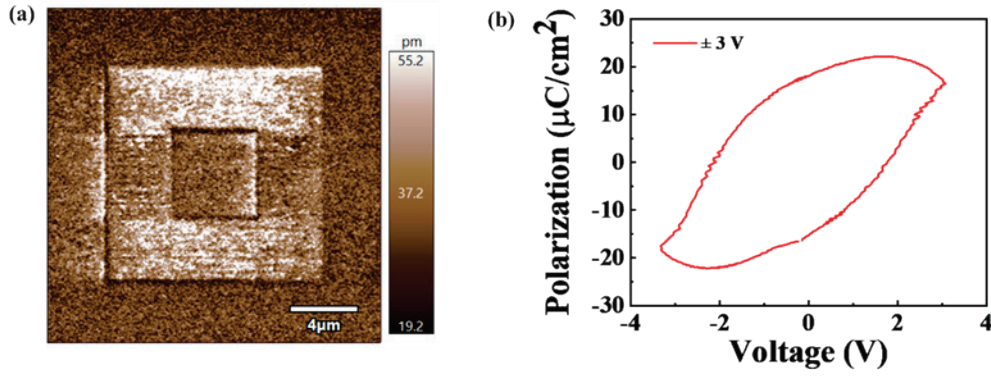
Received March 16, 2023; accepted May 4, 2023

## Supporting Information



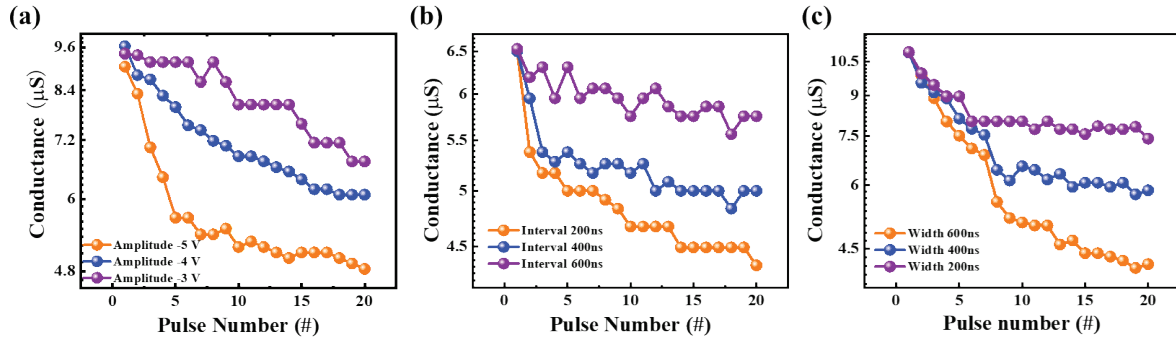
**Fig. S1** Short-term to long-term transition. (a–e) Synaptic weight change and fitted curves versus different pulse numbers (10, 20, 30, 40, and 50). (f) Measured relaxation time change versus pulse number.

As shown in Figs. S1(a)–(e), the normalized synaptic weight decreases rapidly at the beginning, and then slowly decreases within a few weeks, until it finally reaches a moderate level. In order to explain the current decay, a modified Kohlrausch equation is used to fit the normalized current, which is often used as a forgetting function in psychology:  $I_t = I_0 + A \exp(-t/\tau)$ , where  $I_0$  is the steady-state current,  $A$  is the pre-factor, and  $\tau$  is the relaxation constant of the forgetting rate. As the number of stimulation pulses increases, the relaxation time increases, as shown in Fig. S1(f). The results verified the feasibility of transitioning from STP to LTP in the prepared memristor Pd/HfAlO/LSMO/STO/Si.



**Fig. S2** (a) PFM amplitude mapping. (b)  $P-V$  loop.

PFM amplitude mapping is shown in Fig. S2(a). To further verify the ferroelectric polarization effect of the HfAlO ferroelectric memristor, the  $P-V$  loop of the memristor was tested as shown in Fig. S2(b). Figure S2(b) shows the  $P-V$  hysteresis of HfAlO films before and after  $\gamma$ -ray irradiation at a swept voltage of  $\pm 3$  V. This indicates that the HfAlO ferroelectric memristor has good polarization properties.



**Fig. S3** Measured device conductance with different pulse amplitudes, pulse intervals, and pulse widths.

Negative voltage pulses can also both increase and decrease the current, as shown in Fig. S3. The negative pulse amplitude is increased from  $-3$  V to  $-5$  V, as shown in Fig. S3(a). It can be found that the conductance of the memristor decreases as the negative pulse amplitude increases when the pulse width and pulse interval are  $500$  ns and the pulse number is  $20$  cycles. Next, Fig. S3(b) illustrates the effect of different pulse intervals on the conductance. The amplitude of the programmed pulse is  $-5$  V and the width is  $500$  ns. It is observed that the trend of the conductance changing with the pulse interval is opposite to the trend of the pulse amplitude. Finally, the effect of pulse width on conduction variation was investigated. Figure S3(c) shows the effect of pulse width on conduction at  $-5$  V pulse amplitude and  $500$  ns pulse interval. The results show that the conductivity decreases with increasing pulse width. Figure R4 proves that device conductance is adjusted by different negative pulse parameters.

The LRS partial  $I-V$  curve can be fitted to the  $I=Asinh(BV)$  function, where the fitting constants are  $A = 2.32$  and  $B = 1.79$ , respectively. The expressions of  $A$  and  $B$  are respectively as follows [1]:

$$A = 2Ne\nu\exp\left(-\frac{U}{kT}\right),$$

$$B = \frac{ae}{\delta kT},$$

where  $N$  is the number of electrons per unit volume,  $e$  is electronic charge,  $\nu$  is the probability of transition without barriers (per second),  $U$  is the height of the barrier,  $k$  is Boltzmann constant,  $T$  is the

temperature,  $a$  is the half width of the barrier, and  $\delta$  is the thickness of the film. According to  $I=N \cdot e \cdot s \cdot d$ , the value of  $N$  can be obtained. According to  $R=\rho \cdot d/s$  and  $\rho=1/(N \cdot e \cdot v)$ , the value of  $v$  can be obtained. In the above expressions,  $s$  is the cross-sectional area of the device and  $d$  is the thickness of the device (total thickness of Pd, HfAlO and LSMO). Substituting  $N$  and  $v$  into the expression for  $A$ ,  $U$  is calculated to be 0.29 eV. Substituting the film thickness  $\delta$  into the expression for  $B$ ,  $a$  is calculated to be 0.38 nm. The values of  $a$  and  $U$  are relatively reasonable values of the correct order of magnitude as reported in the previous study [2]. Therefore, the width and height of the barrier are 0.76 nm and 0.29 eV, respectively.

## References

- [1] J. Zhang, T. Yang, J. Wang, J. Zhao, and X. Yan, *Science China Mater.* 64, 179 (2021)
- [2] P. Dayan and L. Abbott, *Theoretical Neuroscience: Computational and Mathematical Modelling of Neural Systems*, Cambridge: MIT Press, 2001