

## **Ion-gating Synaptic Memristor Based On Tri-layer $\text{HfO}_x$ Composition Regulation**

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### **AFFILIATIONS**

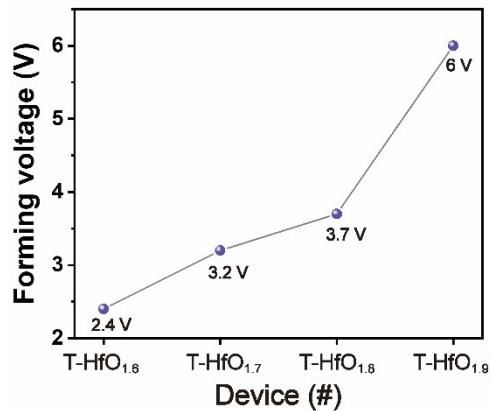
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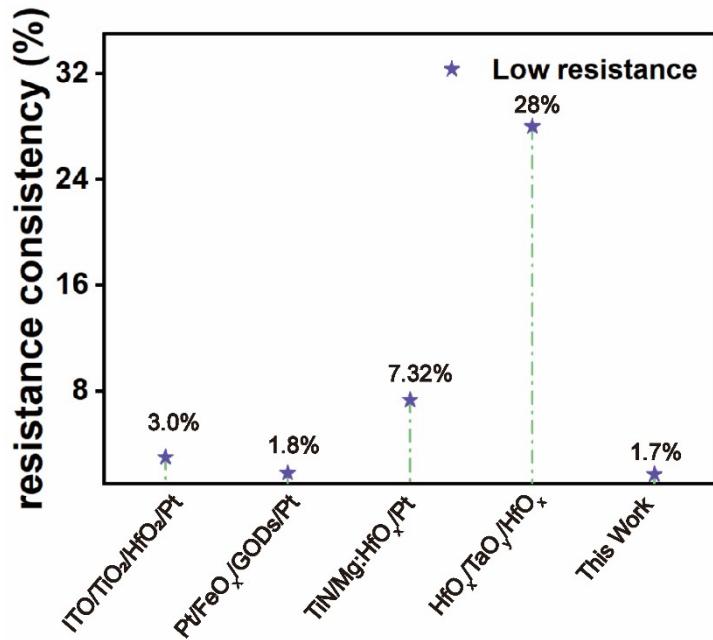
## S1 Device Forming voltage comparison diagram



**Fig.S1.** Comparison diagram of Forming voltage values of four devices

As shown in [Fig.S1](#), the Forming voltage of T-HfO<sub>1.6</sub> is 2.4 V, that of T-HfO<sub>1.7</sub> is 3.2 V, that of T-HfO<sub>1.8</sub> is 3.7 V, and that of T-HfO<sub>1.9</sub> is 6 V. It can be seen that the Forming voltage increases with the increase of x value.

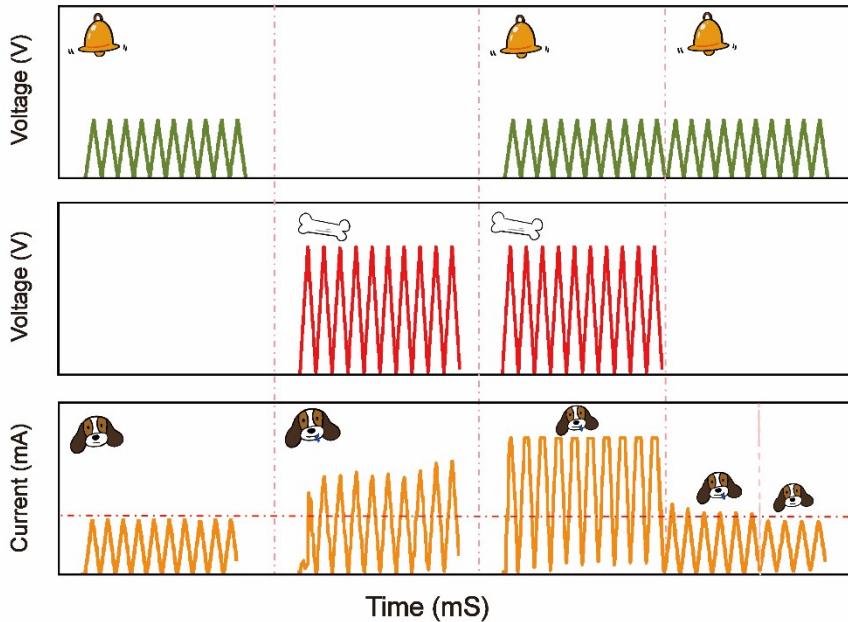
**S2 Comparison diagram of low resistance consistency of T-HfO<sub>1.7</sub> with other memristor devices**



**Fig.S2.** Low resistance consistency comparison diagram

As shown in [Fig. S2](#), comparing T-HfO<sub>1.7</sub> with other memristor devices<sup>1-4</sup> with improved consistency, T-HfO<sub>1.7</sub> has good low-resistance consistency because we control the CFs breakage in the HfO<sub>2</sub> middle layer.

### S3 Pavlov's dog experiment of T-HfO<sub>1.7</sub> device



**Fig.S3. Device implementation of Pavlov's dog process diagram**

As shown in [Fig. S3](#), first, a single bell ringing does not cause the dog to salivate, the T-HfO<sub>1.7</sub> is placed in a low conduction state, and the pulse stimulation with an amplitude of 1V is used to simulate the bell ringing, and the T-HfO<sub>1.7</sub> is still in a low conduction state. The food alone causes the dog to salivate, and a pulse stimulus with a amplitude of 3V is used to simulate the food, which can be seen to put the T-HfO<sub>1.7</sub> in a high conduction state; Further, in the training process, the dog was fed when the bell rang, establishing a connection between the food and the bell, after this training, when the food was removed, the bell sounded, the dog would also secrete saliva, and voltage was applied to the T-HfO<sub>1.7</sub> at the same time. It can be observed that when the 3V pulse stimulation was removed, the device was still in a high conductivity state, but with the increase of time, after the food was removed, the T-HfO<sub>1.7</sub> was still in a high conductivity state. The association between food and ringing gradually weakens and disappears.

### S3 Estimation of non-ideal factors

The mathematical fitting of T-HfO<sub>1.7</sub>'s simulated weight update behavior is as follows<sup>5</sup> :

$$G_{LTP} = B \left( 1 - e^{-\frac{N}{A}} \right) + G_{min} \#(1.1)$$

$$G_{LTD} = G_{max} - B \left( 1 - e^{-\frac{N - N_{max}}{A}} \right) \#(1.2)$$

$$B = (G_{max} - G_{min}) / \left( 1 - e^{-\frac{N_{max}}{A}} \right) \#(1.3)$$

Where G<sub>LTP</sub> and G<sub>LTD</sub> are functions of the LTP/LTD curve varying with the number of pulses N, G<sub>max</sub> and N<sub>max</sub> is the saturation conductance value and the maximum number of pulses allowed to be applied, respectively, and A and B are the parameters to be fitted.

Further, the nonlinearity ( $\alpha$ )can be fitted:

$$\alpha = \frac{1.726}{A + 0.162} \#(1.4)$$

**Table S1. CV of LRS, HRS and Nonlinear parameter of LTD/LTP between synaptic memristor devices**

Device	CV of LRS/HRS	Nonlinear parameter of LTD/LTP	Ref.
Au/HfO <sub>x</sub> /TaO <sub>y</sub> /HfO <sub>x</sub> /Ti/TiN	0.28/0.57	5.3/11.9	4
Pt/HfO <sub>2</sub> NRs/TiN	0.219/0.312		6
Pt/TiO <sub>x</sub> /ZnO/SiO <sub>2</sub> /TaN	0.155/0.326	8/0.52	7
Al/A-C-B-D TiO <sub>x</sub> /Al		2.4/4.6	8
n-Si/HfO <sub>2</sub> /WO <sub>3</sub> /Ag		2.49/5.54	9
T-HfO <sub>1.7</sub>	0.017/0.126	1.55/4.55	This work

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