Supporting information

Revealing the Potential of 2D WS_2 Memristors as an Artificial Synapse with Resilient Gradual Behavior at High Temperatures for Neuromorphic Applications

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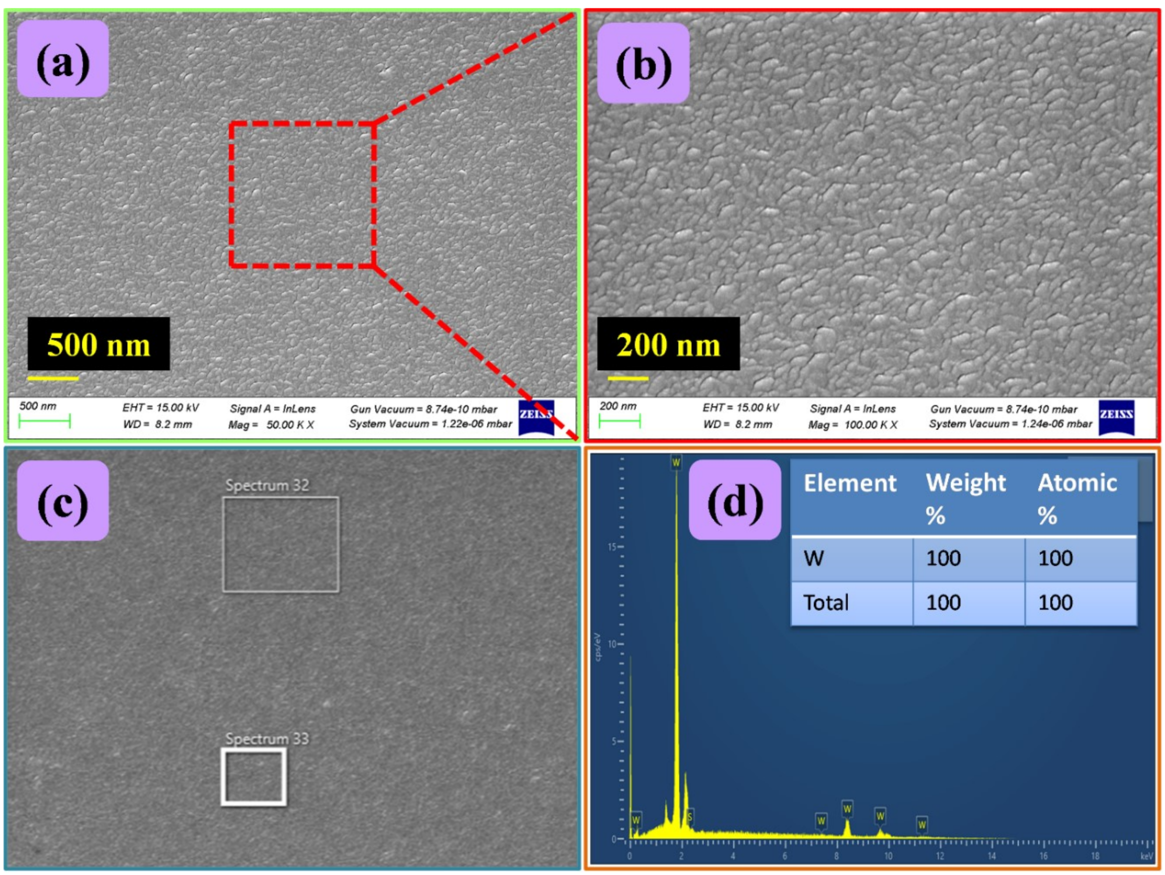

Fig. S1. Top-surface FESEM images of W at magnifications of (a) 50kx, (b) 100kx. (c) Elemental mapping of W. (d) EDS spectrum of W (inset shows elemental concentration).

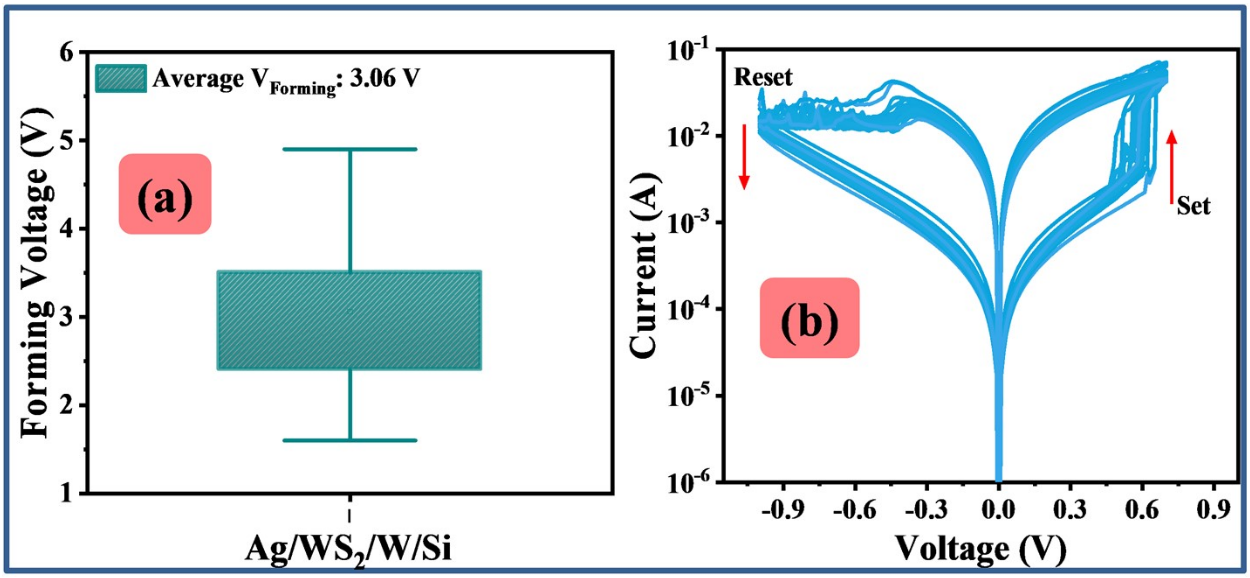

Fig. S2. (a) Statistical distribution of forming voltage of the Ag/WS_2/W memristive device. (b) The 150-cycle I-V curve of the memristive device.

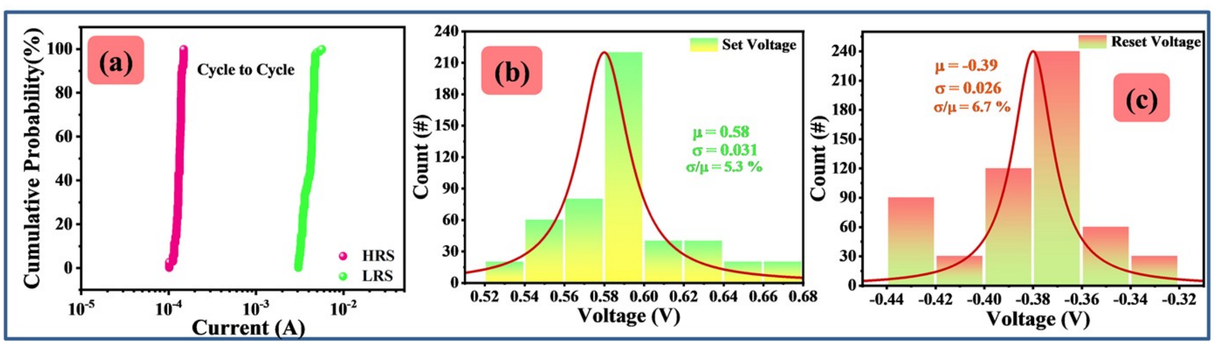

Fig. S3. (a) Cumulative distributions of the HRS and LRS. (b, c) Statistical distribution of set and reset voltages.

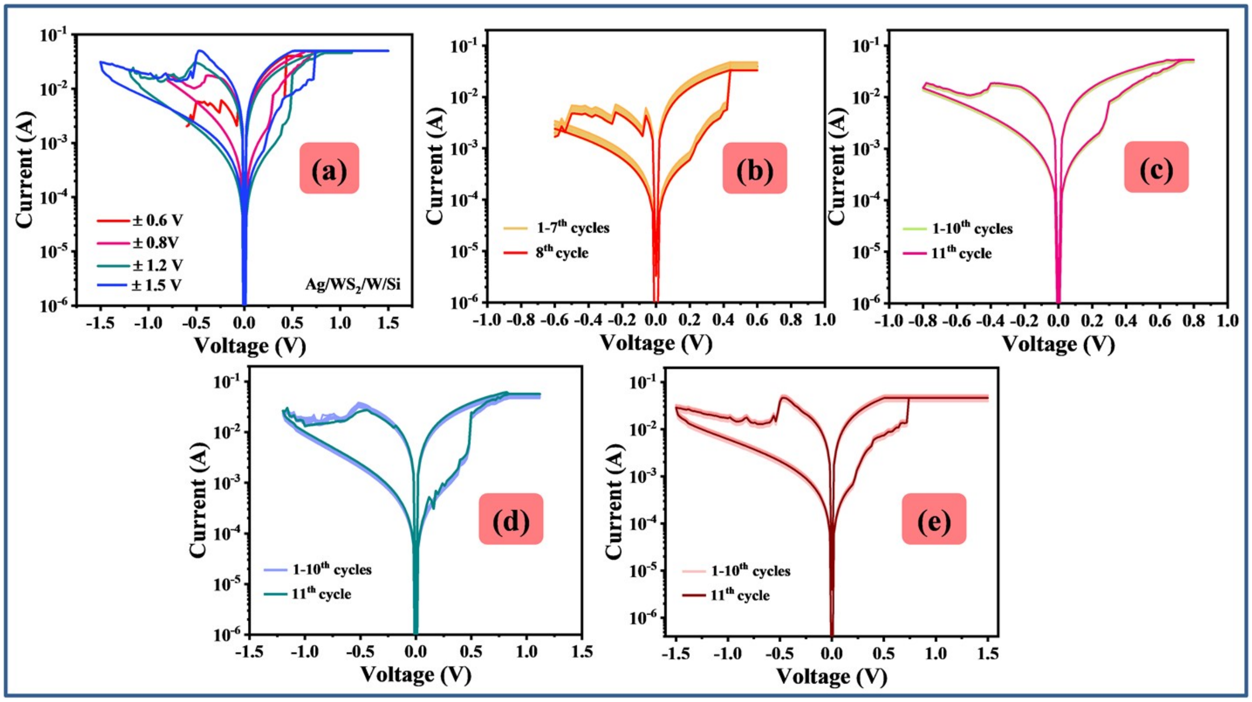

Fig. S4. (a) I-V curve of the memristor device, (b, e) Corresponding multiple IV cycles at different voltage ranges.

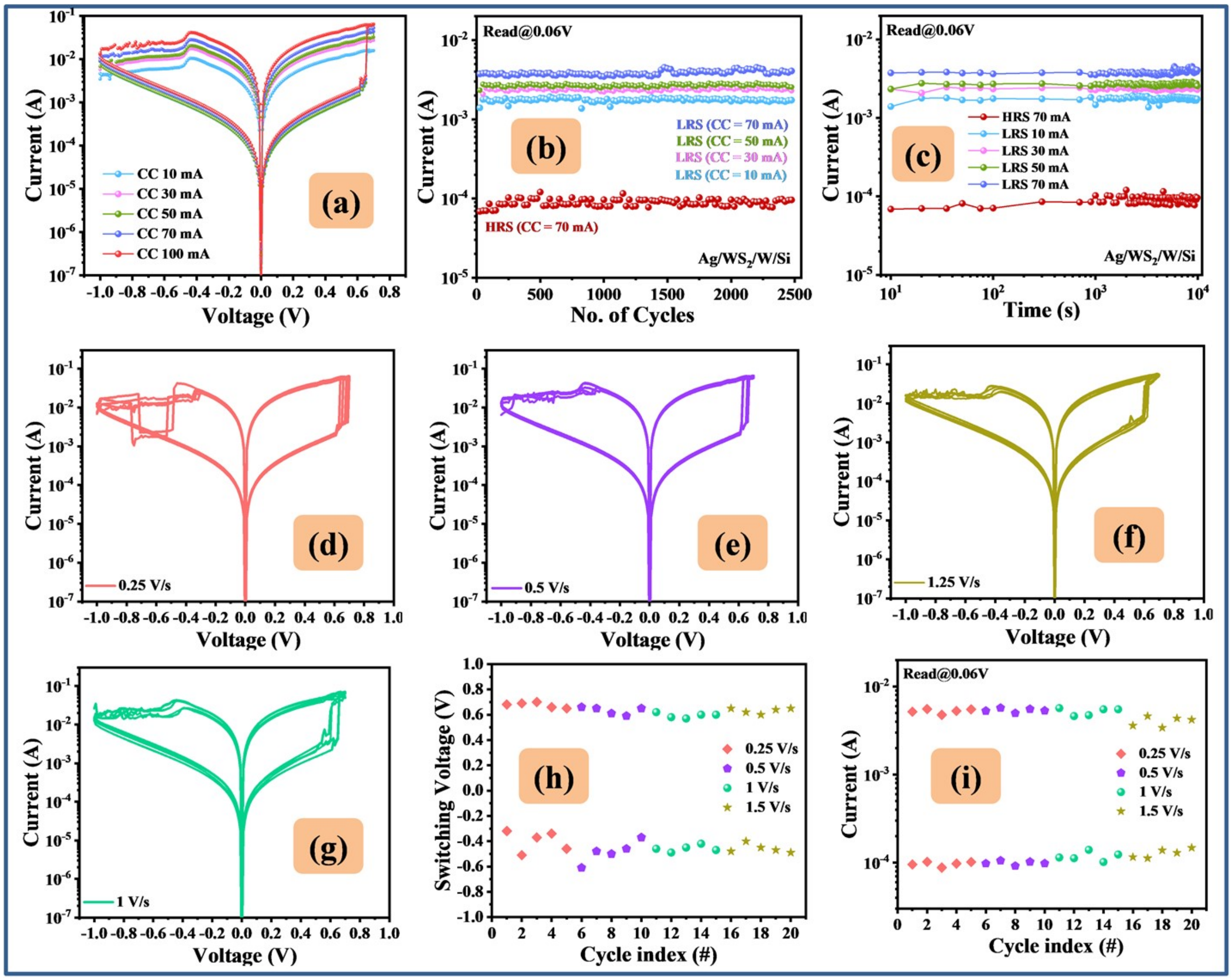

Fig. S5. Effect of compliance current (CC) and voltage scan speed. (a) I-V curve of the memristor device at different CC. (b, c) Endurance, retention at CC (70 mA, 50 mA, 30 mA, 10 mA), respectively. (d-f) I-V characteristics at different scan speeds. (h) The variation of switching voltages with scanning speed. (i) The change in HRS and LRS states.

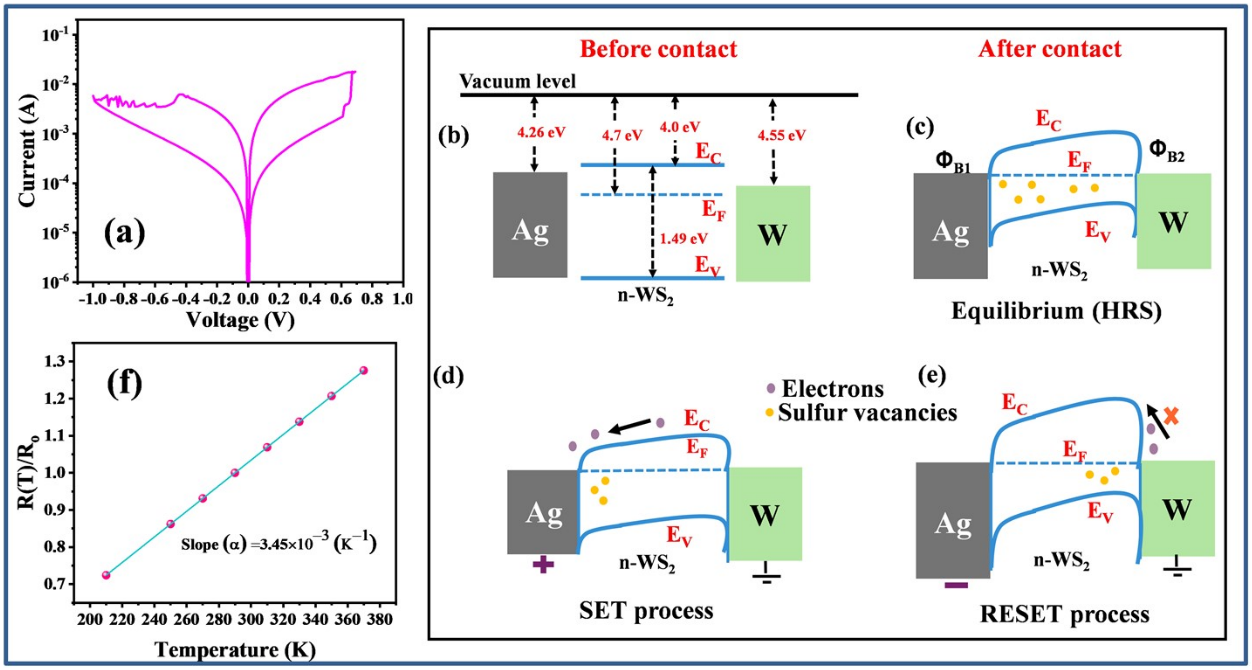

Fig. S6. (a) IV curve with tungsten as top electrode. Energy level band diagram (b) Before contact, (b) after contact (no biasing), (c) SET process, (d) RESET process. (f) Temperature-dependent coefficient of Ag.

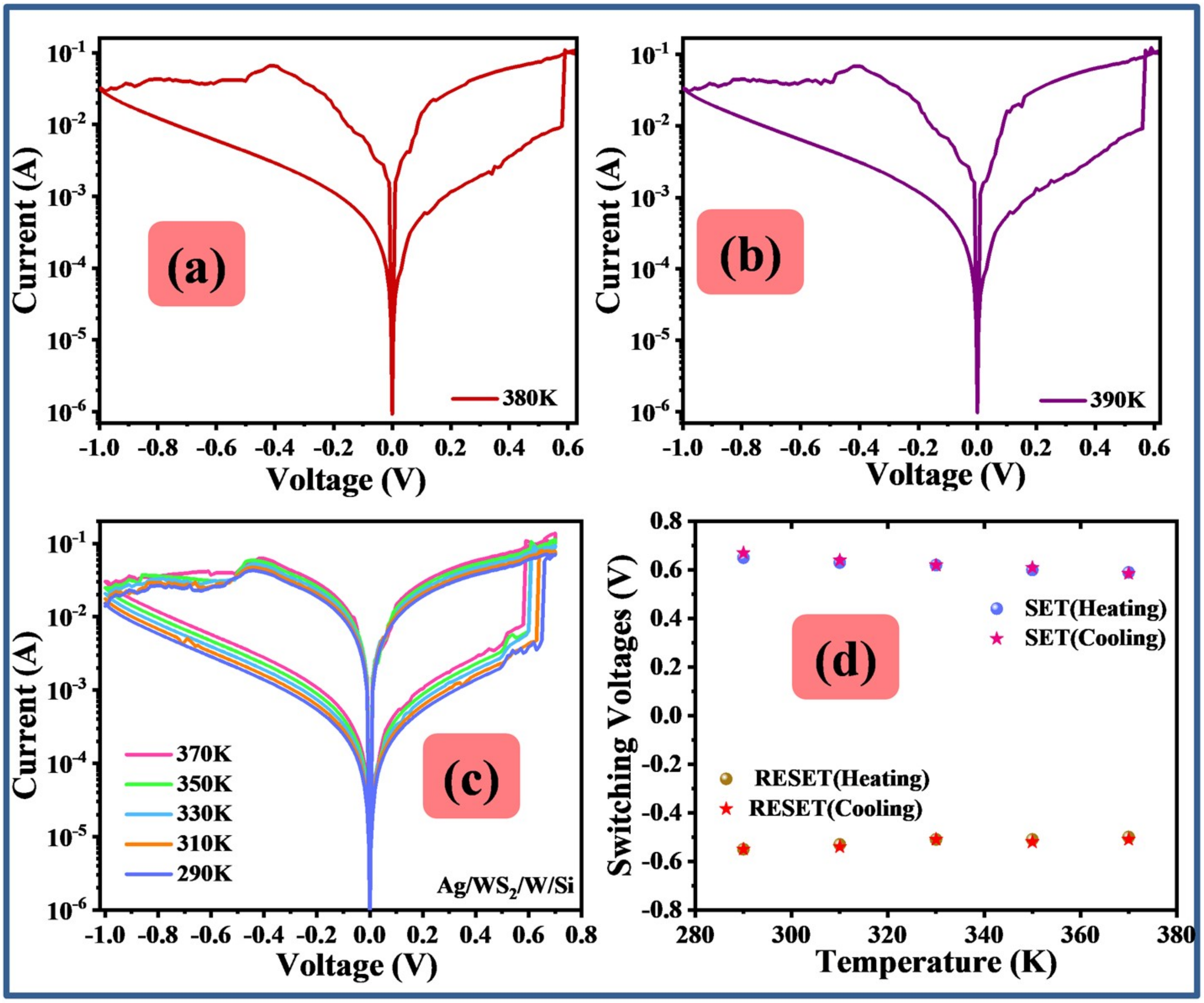

Fig. S7. Current-voltage curve at (a) 380K, (b) 390K. **(**c) Temperature-dependent IV curve while decreasing from 370K to 290K. (d) SET/RESET variation while increasing (heating) and decreasing (cooling) temperature.

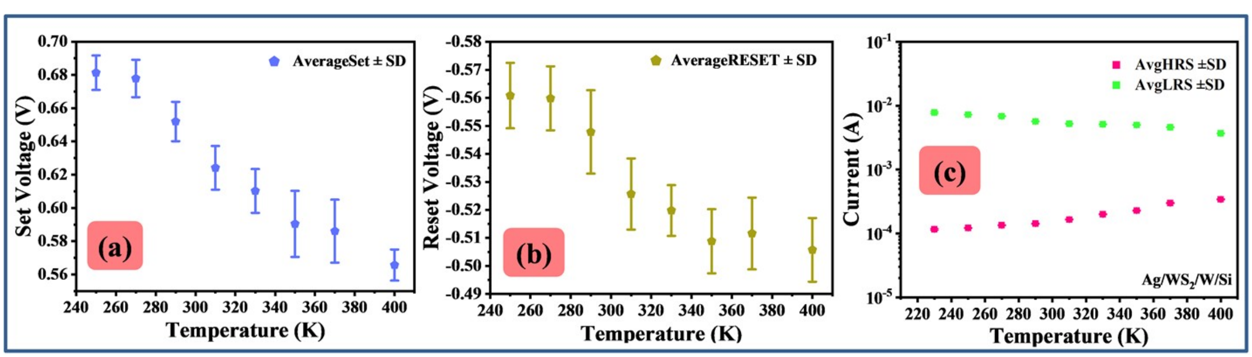

Fig. S8. Temperature-dependent (a, b) SET and RESET voltages, respectively, (c) HRS/LRS states, using error bar plots.

Section S1. **Calculation of barrier height**

The Schottky barrier height (Φ_B) at the Ag/WS_2 interface can be determined using the Richardson Schottky-thermionic emission equation model, which describes the current flow under the influence of an electric field. The current-voltage (I-V) relationship is expressed as:

$$I = A^*T^2 exp[\frac{-q(\Phi_B - \sqrt{qV}/4\pi\varepsilon_r\varepsilon_o d)}{k_B T} \quad (1)$$

where A= π (50 μm)^2 is the contact cell area, and Richardson constant, $A^* = \frac{4\pi\epsilon qMnk^2}{h^3}$, q is electric charge, ϵ is the dielectric constant of the material, $\emptyset_B$ is Schottky barrier height, T is temperature, d is the distance between the top and bottom electrodes, and k is the Boltzmann constant.

Taking the natural logarithm of Eq. (1):

$$\ln(I) = \{\ln\left(AA^*T^2\right) - \frac{q}{kT}\Phi_B\} + \frac{q}{kT}\sqrt{\frac{E}{4\pi\varepsilon}}\sqrt{V}$$

Thus, a plot of ln(I) against $\sqrt{V}$ yield a straight line, where the intercept provides the information about the barrier height ($\emptyset_B$). The intercept at $\sqrt{V}$=0 is given by:

$$Intercept = \{\ln\left(AA^*T^2\right) - \frac{q}{kT}\Phi_B\}$$

The term $\ln\left(AA^*T^2\right)$ is much smaller compared to $\frac{q}{kT}\emptyset_B$, allowing it to be neglected for barrier height estimation. Thus, $\emptyset_B$ can be approximated from the intercept as:

$$\emptyset_B = -\left(\frac{kT}{q}\right) \times Intercept$$

Using this, the Schottky barrier at the Ag/WS_2 interface is determined as 0.38 eV.

Table S1. Comparison of prior reported WS_2-based memristor devices.

Device structure	Modulation method	Synaptic behavior	Non-linearity (LTP/D)	No. of pulses /cycle	Pulse information (Amplitude /width)	Endurance (LTP/LTD)	Time constants (τ_1, τ_2)	Refs.
Pd/WS_2/Pt	Spin Coating	PPF, STDP, STP to LTP	No	No	PPF: 2V (500 ns) STDP: ± 2V (500 ns)	No	N/A	[1]
TiN/BTO/WS_2/TiO_x/Pt	Bilayer	LTP/D, PPF	N/A	200	LTP/D: 0.7 V to -0.72 V (50 μs)	50 cycles	(0.065 ms, 0.75 ms)	[2]
Ag/$Zr_{70}W_{30}$/Pt	Hybrid nanocomposite	LTP/D, PPF/D	(2.5, -0.12)	100	LTP/D: 0.36 to 0.44 V (-0.34 to -0.42 V)	20 cycles	(0.06 μs, 0.186 μs)	[3]
Pt/TiN/HfO_2/WS_2/Pt	Bilayer	LTP/D, PPF/D, STDP, SRDP	(1.5, 0.45)	100	LTP/D: ±1.4 V (1 μs), PPF/D: ±1.2 V (2 μs)	6 Cycles	N/A	[4]
Ag/WS_2/W	**Sputtering**	**LTP/D, PPF, D**	**(1.24, 1.16)**	**120**	**LTP/D: ±0.8 V (20**	**>100 cycles**	**7.17 ms,**	**This Work**

					ms) PPF/D: ±0.8 V (10 ms)		**130.37 ms**	

References

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