SUPPLEMENTARY INFORMATION

1. Reliability evaluations in HfO₂-based synaptic devices.

The write endurance of HfO₂-based synaptic devices with Ag TE and TaN TE were shown in Figure S1 a and c, respectively. The reset/set switching characteristics were measured using the DC sweep mode and a reading voltage of 0.05/0.1 V at room temperature, respectively. The stability and repeatability of resistance states during write endurance measurement were achieved in both Ag TE and TaN TE. It is worth to underline that HfO₂-based devices with TaN TE shows a more stable cycling switching ability with an ON-OFF ratio over 10². Figure S1 b and d shows the retention characteristics of the HfO₂-based devices with Ag TE and TaN TE at room temperature/85°C with a read voltage of 0.05/0.1V, respectively. The ON-OFF ratio of both Ag TE and TaN TE are stable and can be sustained at a high value over 10 years. Thus HfO₂-based material system exhibits satisfactory electrical characteristics for NVM application.

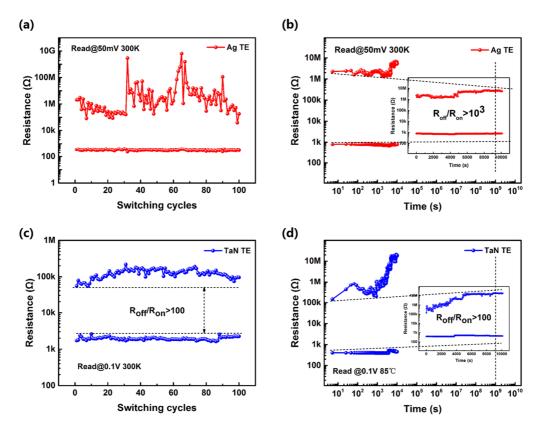


Figure S 1 Device reliability evaluations in HfO2-based synaptic devices. The

write endurance results by DC sweeping mode in Ag TE(a) and TaN TE (c). retention measurements of the HfO2-based devices with Ag TE (b) and TaN TE (d) at room temperature/85°C, respectively. A read voltage of 0.05V/0.1V were applied on the Ag/TaN TE during the electrical measurements, respectively.

2. AC pulse response in TaN/HfO₂/Pt devices.

The AC pulse response was implemented in TaN/HfO₂/Pt devices. Pulses with 2 V/50 ns and -2 V/100 ns were applied for SET and RESET programming, respectively (Figure S2). Stable and repeatable switching characteristic with a steady on-off ratio over 10^2 was obtained in TaN/HfO₂/Pt devices. These TaN/HfO₂/Pt devices exhibit superior electrical characteristics for ultra-fast NVM application. The pulse duration for RESET programming is longer than the time for the SET programming. This is because the SET process is a positive feedback between the formation rate of V₀-based filament, the temperature and local field strength¹, while RESET process is a negative feedback.

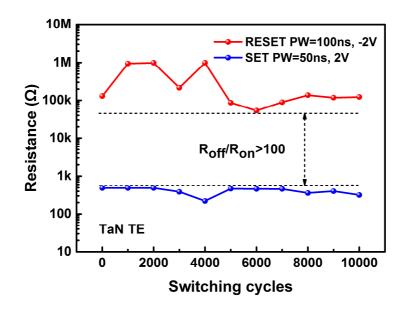


Figure S 2 AC pulse response in TaN/HfO₂/Pt devices. Pulses with 2 V/50 ns and -2 V/100 ns were applied for SET and RESET programming, respectively.

3. Current transport analysis in Ag/HfO₂/Pt devices.

The conduction mechanism of I-V characteristics in LRS and HRS under different

compliance currents were analyzed in Ag/HfO₂/Pt devices. As is shown in Figure S3, the current transportation in Both LRS and HRS exhibit ohmic conduction mechanism at positive electrical field in Ag/HfO₂/Pt devices. The diameter of Ag filament can be modulated by vary the value of compliance current, thus multi-level resistance states can be realized in Ag/HfO₂/Pt synaptic devices. These multi-level conductance states are essential to mimic the synapse abilities in neuromorphic computing system.

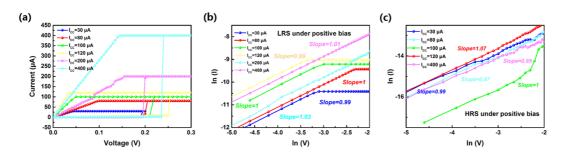


Figure S 3 Current transport analysis in Ag/HfO₂/Pt devices. (a) I-V curves under different compliance currents were measured at positive electrical field. The current transportation in Both LRS (b) and HRS (c) exhibit ohmic conduction mechanism at positive electrical field in Ag/HfO₂/Pt devices. **REFERENCE**

 Guan, X.; Yu, S.; Philip Wong, H.-S. On the switching parameter variation of metal-oxide RRAM - Part I: Physical modeling and simulation methodology. *IEEE Trans. Electron Devices* 2012, 59, 1172–1182.