

Supporting Information

Physical Reservoirs based on MoS₂-HZO Integrated Ferroelectric Field-effect Transistors for Reservoir Computing Systems

Lingqi Li,^a Heng Xiang,^a Haofei Zheng,^a Yu-Chieh Chien,^a Ngoc Thanh Duong,^a Jing Gao,^a and Kah-Wee Ang^{*a}

Department of Electrical and Computer Engineering, National University of Singapore, 4 Engineering Drive 3, 117583, Singapore

E-mail: eleakw@nus.edu.sg

S1. Demonstration of 1D-FeFET array

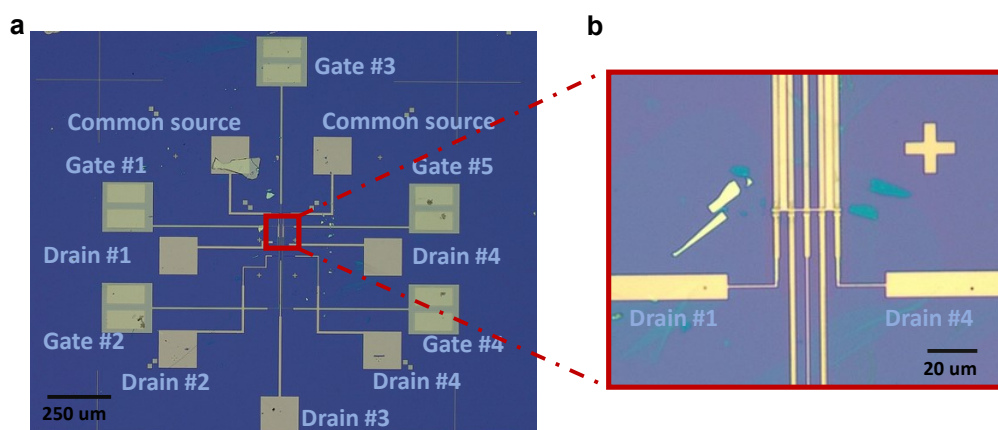


Fig. S1 (a) Microscopic image of the device array utilized in this study, with a scale bar of 250 μm. (b) Zoomed-in view highlighting the flake isolation.

S2. EDX results of MoS₂-HZO FeFETs

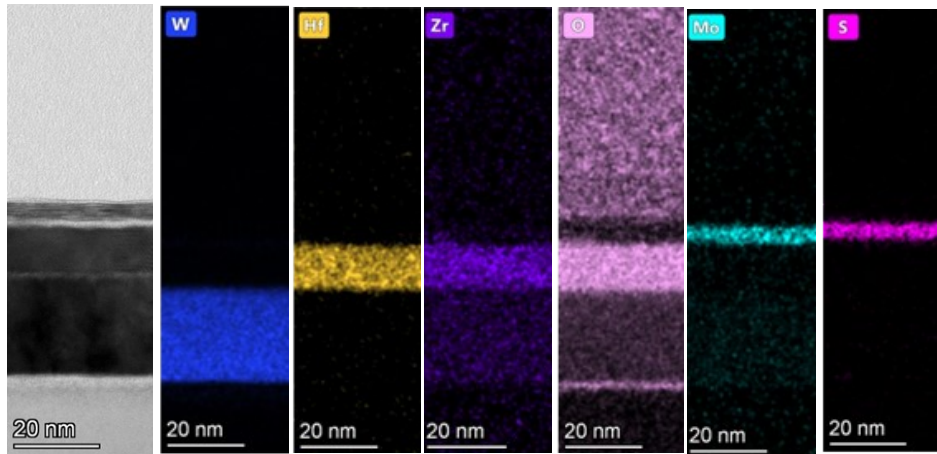


Fig. S2 EDX map comprises W, Hf, Zr, O, Mo, and S elements.

S3. FORC measurement schedule

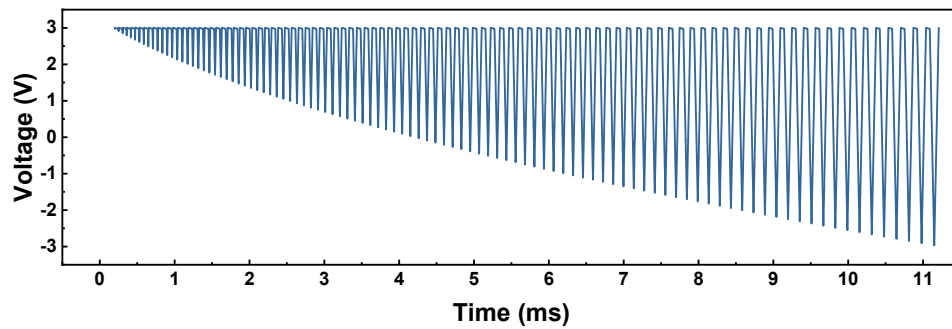


Fig. S3 Electric field for the FORC measurement using MFM capacitor. The reversal electric field (E_r) changes from 3.0 to -3.0 V with a step of -0.01 V, and the external field descends from 3.0 V to E_r , and then ascends to 3.0 V.

S4. Switching current for the NLS model fitting

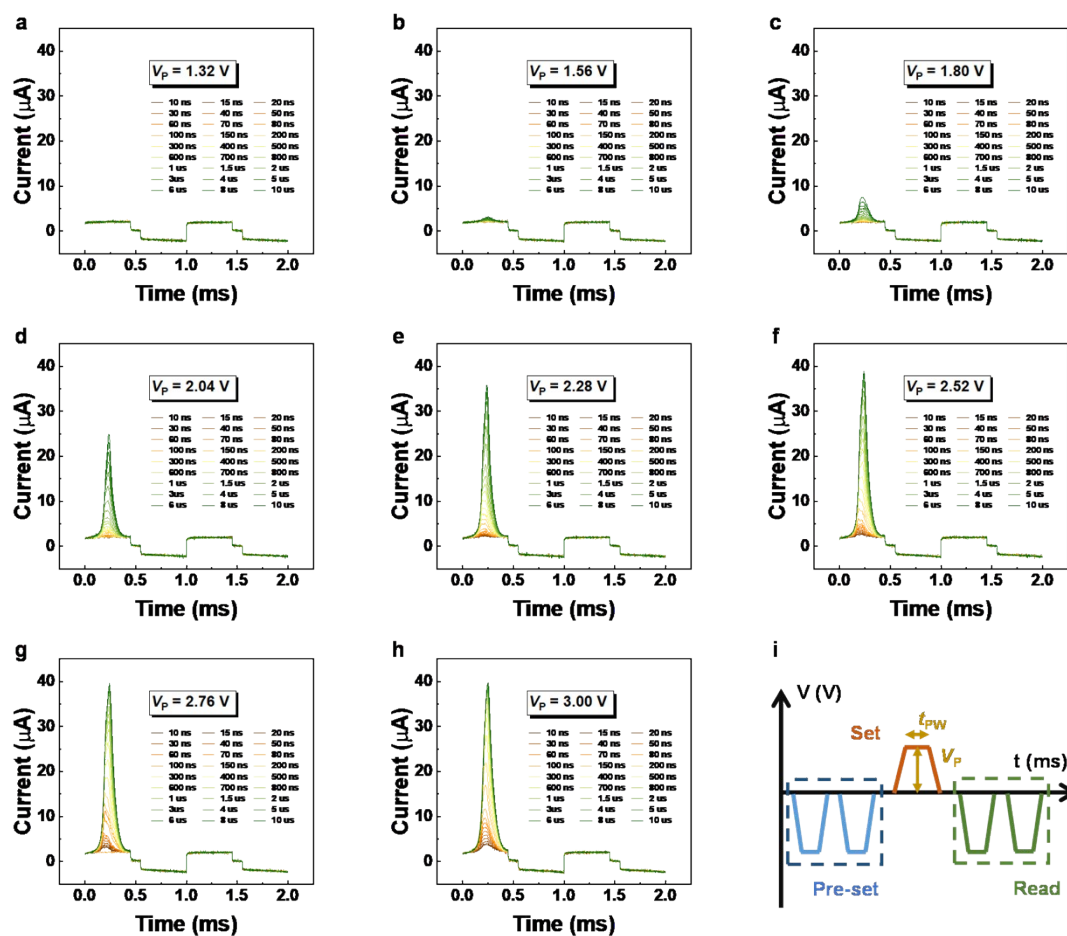


Fig. S4. Switching current at different program voltage (V_p) with varying programming duration (t_p). (a) - (h) The V_p changes from 1.32 to 3.00 V, with a step of 0.24 V, and the t_p varies from 10 ns to 10 μs for each V_p . (i) Illustration of the switching current measurement setup.

S5. Deconvolution of varied results with different ozone pulse time

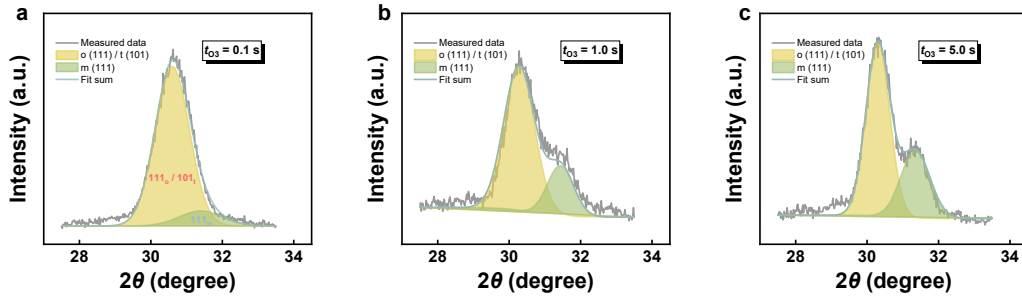


Fig. S5 Deconvoluted XRD results in the 2θ range of 27.5° - 33.5° varied from t_{O_3} of a) 0.1 s, b) 1.0 s, and c) 5.0 s.

S6. Long accumulation of response currents

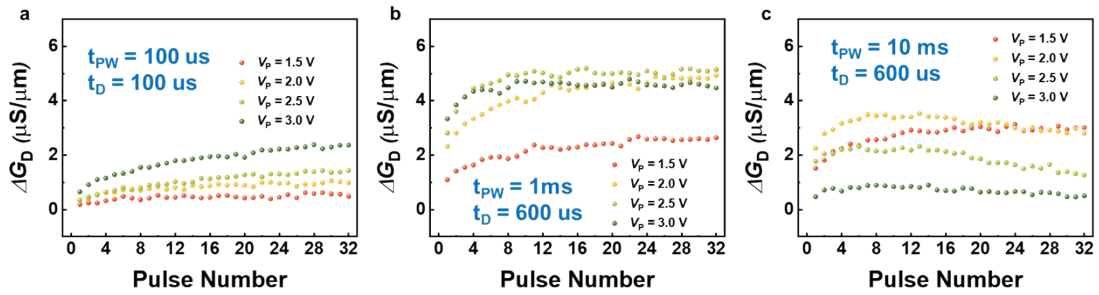


Fig. S6 Synaptic behavior by modulation of the conductance states with the pulse trains. Identical programming bias (V_p) is introduced from 1.5 V to 3 V for all the input pulse scheme. The read voltage (V_R) of 0.5 V with $V_D = 0.5$ V is fixed to all the input pulse trains. (a) $t_{PW} = 100 \mu s$, $t_D = 100 \mu s$ (b) $t_{PW} = 1 ms$, $t_D = 600 \mu s$ and (c) $t_{PW} = 10 ms$, $t_D = 600 \mu s$.

One can observe that when we apply a short program pulse width (t_{PW}), the conductance difference (ΔG_D) significantly increase with higher programming gate bias (V_p). These observations indicating that more dipolar switching in the HZO

ferroelectric layer under larger voltage. On the other hand, when we further increase the t_{PW} , the ΔG_D will not be directly proportional to the magnitude of V_p , which can be attributed to the serious trapping effect reduce the current. When we further increase the t_{PW} , the electron trapping become seriously which led the ΔG_D smaller. This experimental result is consistent with previous reports¹⁻⁴ that the ferroelectric response is faster than the trapping response.

S7. Critical Role of Interfacial Layer Al_2O_3

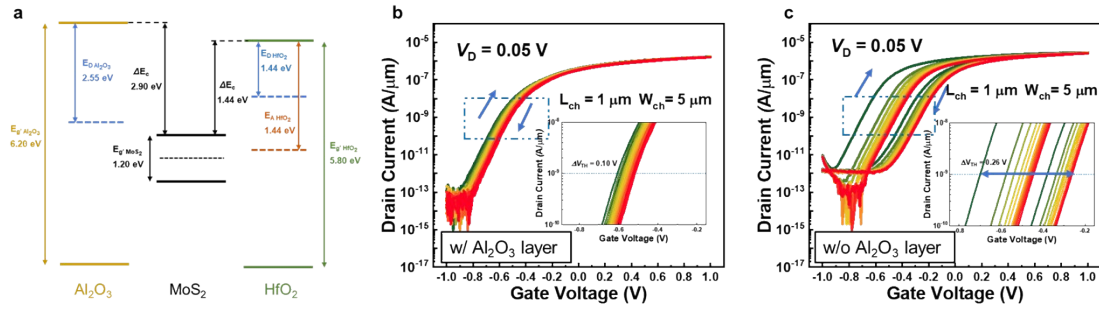


Fig. S7. (a) Energy band diagrams showing the band alignment of few-layer MoS_2 to the high-k dielectric layer of Al_2O_3 and HfO_2 .⁵⁻⁷ The accurate value of bandgaps, defect levels and band offsets were reported by several previous reports which extracted from the experiments. Transfer characteristics with $f = 0.5$ Hz at $V_D = 50$ mV and V_G from -1.0 V to 1.0 V for MoS_2 FETs (b) with and (c) without 2 nm Al_2O_3 interface layer on 10 nm HZO layer.

The transistor performance will be affected by the interfacial interaction between MoS_2 and the high-k dielectric layer. In this work, we use a thin Al_2O_3 layer (~ 2 nm) works as the interfacial layer between MoS_2 and the ferroelectric HZO layer to minimize the detrimental charge trapping effect. As shown in the depicted energy

band diagrams (**Fig. S7a**), the acceptor-like traps (dark red) of HfO_2 are adjacent to the MoS_2 intrinsic Fermi level which energetically favorable to charge trapping effect. Compared with HfO_2 , the oxide defect states in the energy band of Al_2O_3 shows the energy difference with the conduction band of MoS_2 which lead the detrimental charge trapping difficult and will lower the oxide trap density. Therefore, the depicted energy band diagrams confirm that the Al_2O_3 align well with the MoS_2 so that suitable for the transistors.

Subsequently, the MoS_2 FETs with or without Al_2O_3 interface layer are fabricated to verify the aforementioned deduce. Note that all the process except the Al_2O_3 deposition was conducted together for these two samples. Then the same measurement scheme at V_D of 50 mV and the sweeping frequency (f) of 0.5 Hz was used for the devices. One can observe that the extracted clockwise hysteresis ΔV_{TH} from the device with Al_2O_3 interface layer is much smaller ($\sim 0.10\text{V}$), while the ΔV_{TH} is 0.26 V of the device without Al_2O_3 over the same voltage range. Therefore, we have validated that address the interfacial interaction between MoS_2 and the high-k dielectric layer by Al_2O_3 interface layer is suitability.

S8. MoS₂ FETs with pure Al₂O₃ gate stack

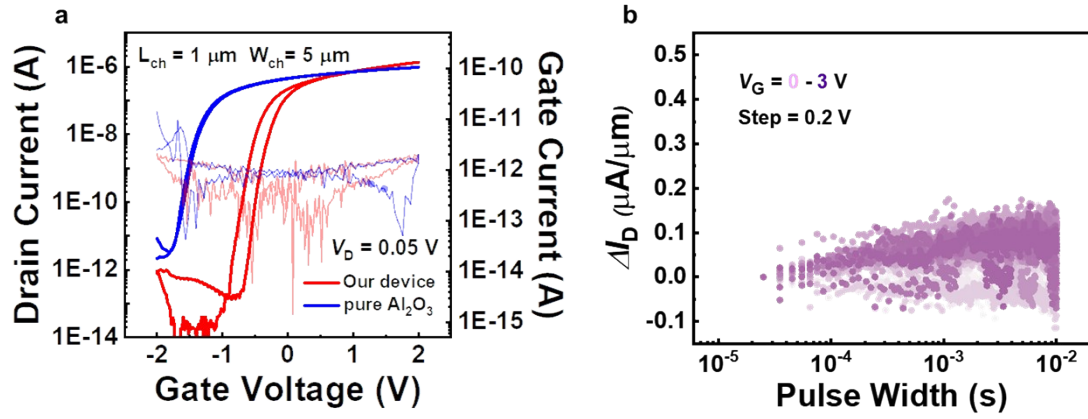


Fig. S8. (a) Transfer characteristics and gate leakage current at $V_D = 50$ mV and V_G from -2.0 V to 2.0 V for MoS₂ FETs using 12 nm pure Al₂O₃ or 2 nm Al₂O₃ stacks with 10nm HZO layer with f of 0.50 Hz. (b) The dynamic voltage response under a 3.0 V input pulse trains with $t_{PW} = 100 \mu s$ and $t_D = 100 \mu s$.

It is evident that the device based on pure Al₂O₃ shows a considerably smaller transient drain current compared to the HZO-based device, as shown in Fig. 2e. It confirms that the dynamic behavior observed is attributed to the ferroelectric HZO rather than the interfacial layer Al₂O₃. Furthermore, the smaller V_{TH} in our device can be attributed to enhanced electrical control.

S9. CV characteristics of the HZO/Al₂O₃ capacitor

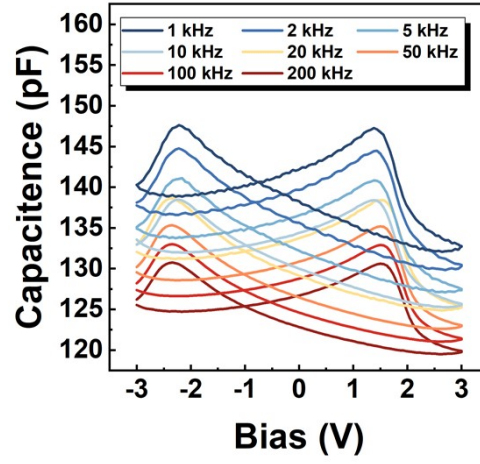


Fig. S9. The frequency-dependent capacitance-voltage (CV) characteristics of HZO/Al₂O₃ capacitor in similar MFM structure as in Fig. 1f. The thicknesses of HZO and Al₂O₃ are 10 nm and 2 nm, respectively.

References

- 1 K. Lee, H.-J. Lee, T. Y. Lee, H. H. Lim, M. S. Song, H. K. Yoo, D. I. Suh, J. G. Lee, Z. Zhu, A. Yoon, M. R. MacDonald, X. Lei, K. Park, J. Park, J. H. Lee and S. C. Chae, *ACS Appl. Mater. Interfaces*, 2019, **11**, 38929–38936.
- 2 M. Si, X. Lyu, P. R. Shrestha, X. Sun, H. Wang, K. P. Cheung and P. D. Ye, *Appl. Phys. Lett.*, 2019, **115**, 072107.
- 3 N. Gong, X. Sun, H. Jiang, K. S. Chang-Liao, Q. Xia and T. P. Ma, *Appl. Phys. Lett.*, 2018, **112**, 262903.
- 4 X. Lyu, M. Si, P. R. Shrestha, K. P. Cheung and P. D. Ye, in *2019 IEEE International Electron Devices Meeting (IEDM)*, 2019, p. 15.2.1-15.2.4.
- 5 Y. Y. Illarionov, T. Knobloch, M. Wlatl, G. Rzepa, A. Pospischil, D. K. Polyushkin, M. M. Furchi, T. Mueller and T. Grasser, *2D Mater.*, 2017, **4**, 025108.
- 6 I. Shlyakhov, J. Chai, M. Yang, S. Wang, V. V. Afanas'ev, M. Houssa and A. Stesmans, *physica status solidi (a)*, 2019, **216**, 1800616.
- 7 Y. Y. Illarionov, T. Knobloch, M. Jech, M. Lanza, D. Akinwande, M. I. Vexler, T. Mueller, M. C. Lemme, G. Fiori, F. Schwierz and T. Grasser, *Nat Commun*, 2020, **11**, 3385.