Supplementary Information (SI) for Journal of Materials Chemistry C.
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Supplementary information

6-Bit Multilevel and Highly Linear Synaptic Plasticity in Sputtered AlScN Ferroelectric Capacitors on (100) Si

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Section 1: PUND Method and Polarization-Bound Leakage Current

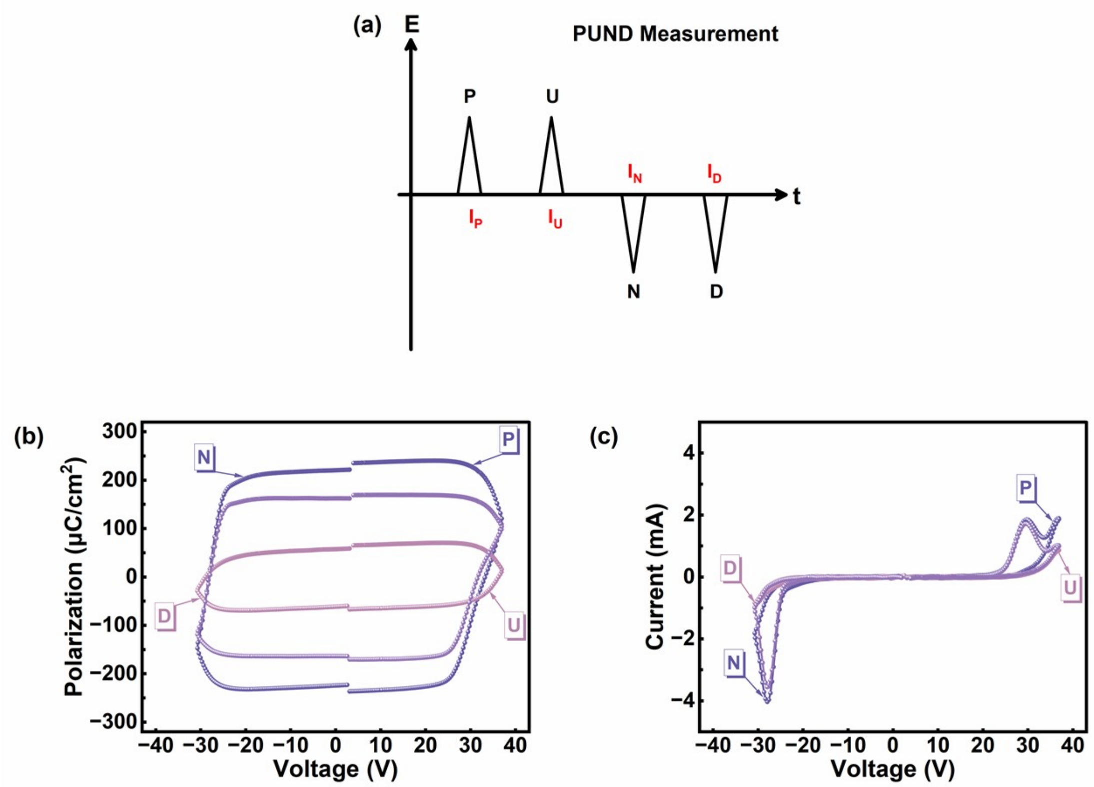

Fig. S1 (a) Measurement principle of the PUND method. (b) P-V and (c) I-V characteristics of the AlScN capacitor obtained using the PUND method, showing the polarization response, non-polarization response, and residual polarization curves.

The voltage waveform applied in the PUND method is shown in Fig. S1(a), and its subtraction principle is described by the following equations:

$$\begin{aligned} I_P &= I_{Switching_+} + I_{Non-Switching_+} \\ I_N &= I_{Switching_-} + I_{Non-Switching_-} \end{aligned} \quad \#(S1)$$

where:

$I_{Switching+}$ and $I_{Switching-}$ represent the polarization switching (ferroelectric) current components during the positive and negative pulses, respectively.

$I_{Non\text{-}Switching+}$ and $I_{Non\text{-}Switching-}$ represent the non-ferroelectric components, including leakage current and dielectric displacement current.

Generally, the non-switching pulses current I_U and I_D , which are not expected to involve ferroelectric switching, can be approximated as:

$$I_U \approx I_{Non-Switching_+}$$
$$I_D \approx I_{Non-Switching_-} \#(S2)$$

This allows the ferroelectric components to be isolated via subtraction:

$$I_{Switching_+} = I_P - I_U$$
$$I_{Switching_-} = I_N + I_D \#(S3)$$

Subsequently, the polarization response is obtained by integrating the switching current over time and dividing by the electrode area:

$$P = \frac{1}{S}\int I(t)dt \#(S4)$$

where *S* is the electrode area, *I(t)* is the time-dependent current during the switching process. The complete P-V and I-V curves obtained using this method are shown in Figs. S1(b) and (c).

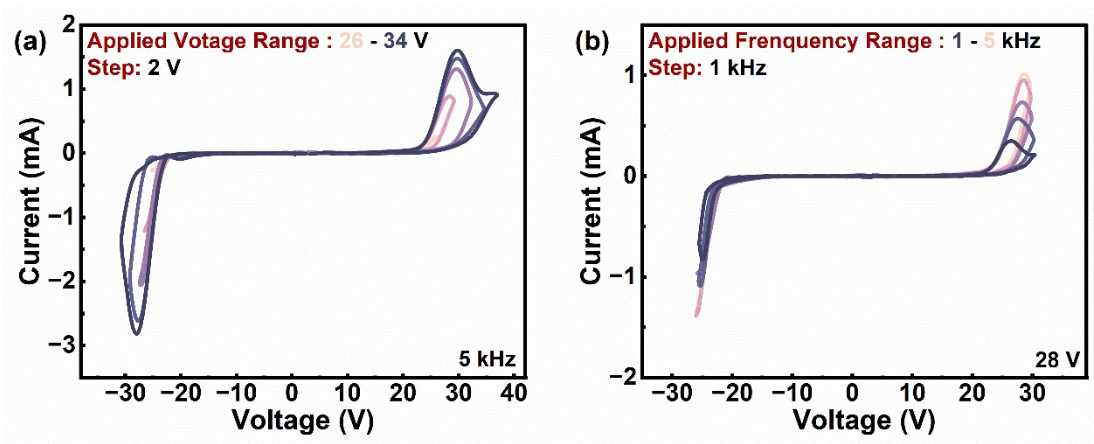

Fig. S2 (a) I-V characteristic of the AlScN ferroelectric capacitor under different voltages and (b) frequencies.

Figs. S2(a) and (b) present the I–V characteristics of AlScN ferroelectric capacitors measured by the PUND method under different voltages and frequencies, exhibiting clear ferroelectric responses. Noticeable secondary current peaks are observed near the maximum voltage, which cannot be eliminated by the PUND method. This phenomenon is associated with the electric-field- and polarization-switching-assisted migration of O_N defects. ***(Small 2025, 21; doi: 10.1002/smll.202509729)***

Section 2: I–V Characteristics of the AlScN Ferroelectric Capacitor

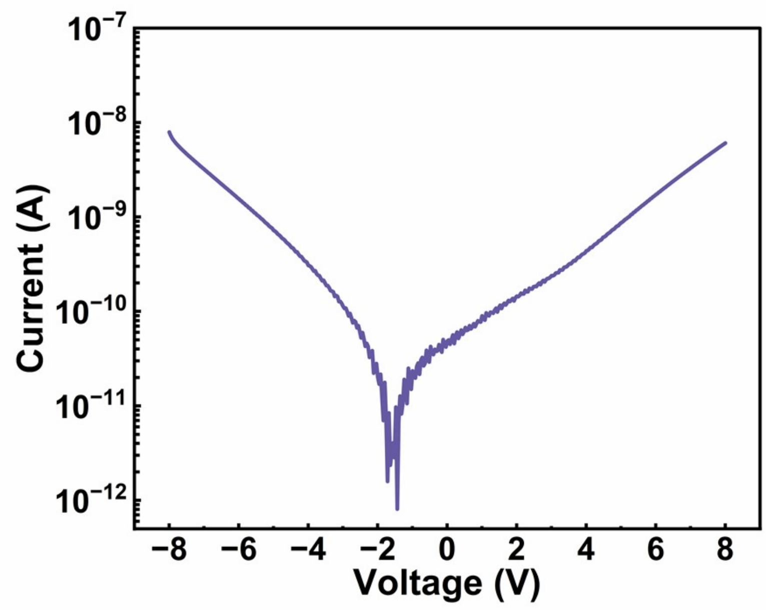

Figure. S3 The I–V curve of the AlScN ferroelectric capacitor measured from -8 V to 8 V.

Section 3: Device to Device Uniformity Results

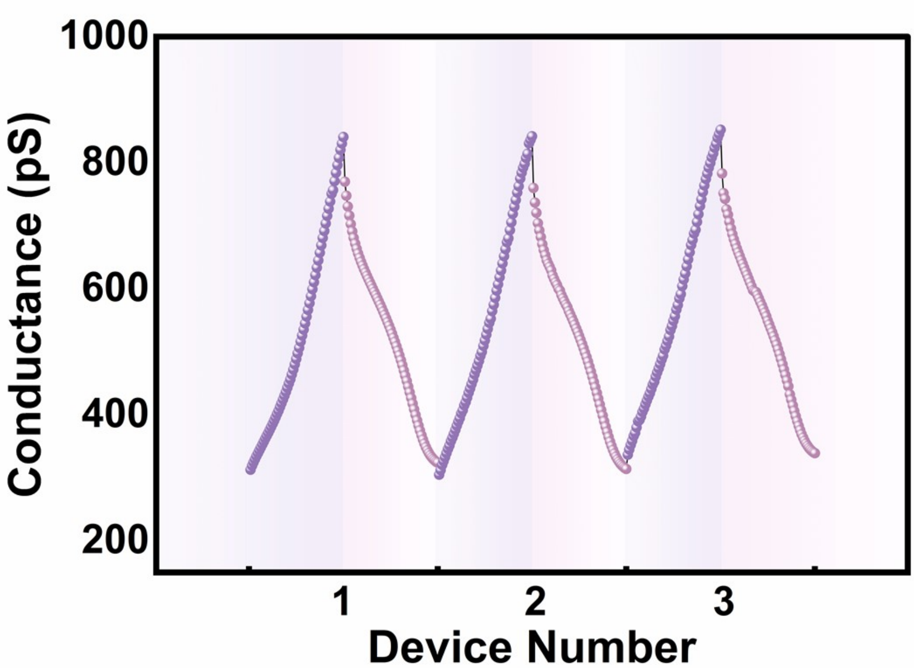

Fig. S4 LTP and LTD weight update processes over three devices.

Section 4: Fatigue performance analysis

The improvement in fatigue endurance of the device can be understood from two perspectives: material defects and the operating voltage of the device.

(1) AlScN deposition is known to be highly sensitive to oxygen, and unintentionally incorporated oxygen is commonly present in sputter-deposited AlScN ferroelectric films. It is generally accepted that the dominant defect types in AlScN ferroelectric films are nitrogen vacancies (V_N) and oxygen substituting nitrogen sites (O_N). ***(Appl. Phys. Lett. 2024, 125; doi: 10.1063/5.0211892)*** In our previous work, we found that the fatigue endurance of AlScN ferroelectric films exhibits a strong negative correlation with the oxygen content (the concentration of O_N defects). ***(Small 2025, 21; doi: 10.1002/smll.202509729)*** Moreover, oxygen in the ambient environment can gradually penetrate into the device during fatigue cycling, acting as an additional defect source and further participating in the fatigue process. However, a lower oxygen content does not necessarily imply a lower overall defect density in AlScN films. M. R. Islam et al. reported that O-doped AlScN films show reduced leakage current than undoped AlScN films. ***(Advanced Electronic Materials 2025, 11; doi: 10.1002/aelm.202400874)*** In our previous studies, we proposed that this behavior can be attributed to the preferential occupation of nitrogen vacancies by oxygen atoms, and that O_N defects contribute less to leakage conduction than V_N defects. In the present work, the deposition parameters of the AlScN films were further optimized by increasing the N_2/Ar ratio during sputtering, leading to a further reduction of unintentional oxygen incorporation (see Fig. 1) compared with our earlier studies. ***(Small 2025, 21; doi: 10.1002/smll.202509729)*** We speculate that this optimization places the oxygen content within an appropriate window, where oxygen can effectively compensate nitrogen vacancies without introducing excessive O-related defects that would otherwise exacerbate leakage current and fatigue behavior.

(2) The synaptic functionality of our device is realized through the gradual modulation of the ferroelectric polarization state by voltage pulse stimulation. To enable incremental polarization switching, the pulse amplitudes used in this work (±20.5 V to ±23.65 V) are deliberately lower than those typically applied during ferroelectric hysteresis measurements (as shown in Fig. 2). As a result, each individual pulse induces switching of only a fraction of ferroelectric domains rather than complete polarization reversal. Under such operating

conditions, fatigue measurements performed at the device working voltage naturally lead to improved fatigue endurance. Consistent with this interpretation, Hyunmin Cho et al. reported fatigue behaviors under different polarization switching extents, showing that the fatigue endurance can differ by up to four orders of magnitude when the conventional $2P_r$ values are 10 μC/cm^2 and 200 μC/cm^2, respectively. ***(Nat Commun 2026; doi: 10.1038/s41467-025-68221-2)***

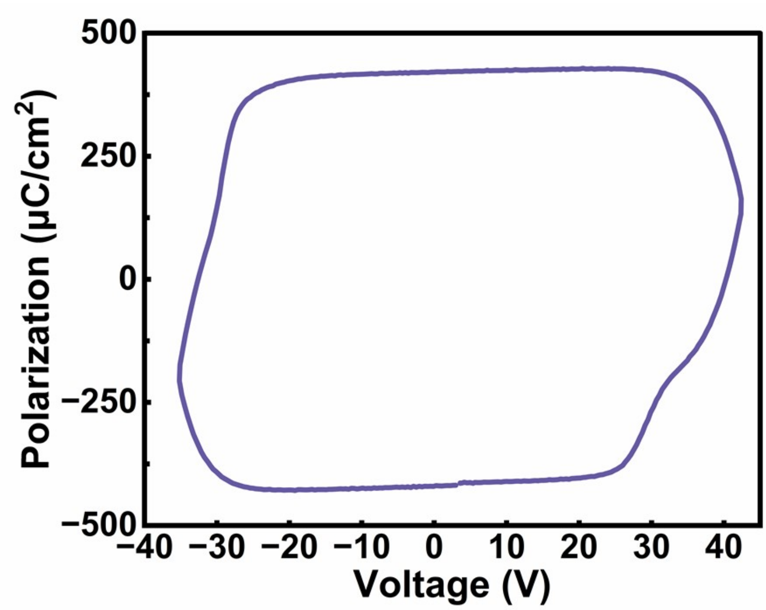

Fig. S5 P–E hysteresis loop measured under a 39 V triangular pulse.