Supplementary Information (SI) for Journal of Materials Chemistry C. This journal is © The Royal Society of Chemistry 2025

## **Supporting Information**

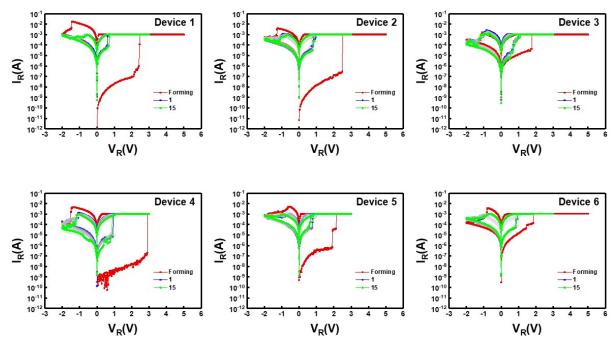
Arbitrary-Ordered Pulsed Programming Achieving 11 Well-Separated Programming Levels via a Multilevel Transistor–Memristor Series Configuration

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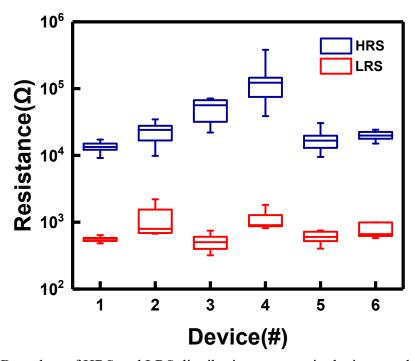
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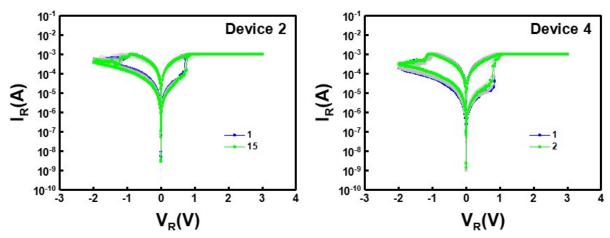
**KEYWORDS:** Memristor, Multilevel resistive switching, Retention, endurance, Transistor—memristor configuration



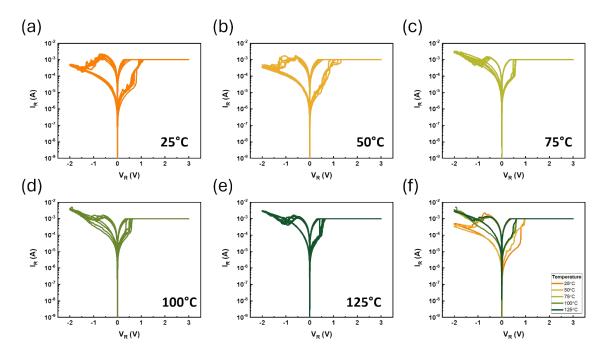
**Figure S1.** Device-to-device (D2D) I–V characteristics of Pt/Ta/ZrO<sub>x</sub>/Pt memristors (Device 1 to Device 6). Each cell underwent a forming process followed by 15 consecutive switching cycles. The consistent bipolar switching behavior and stable current levels confirm good D2D uniformity and operational reliability.



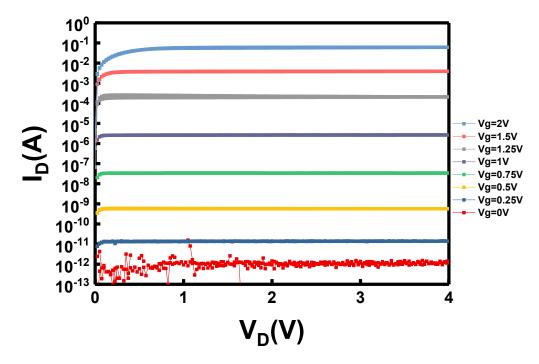
**Figure S2.** Box plots of HRS and LRS distributions across six devices, each measured over 15 switching cycles. The results reflect reasonable device-to-device variation and switching stability, supporting the reproducibility shown in Supplementary Figure S1.



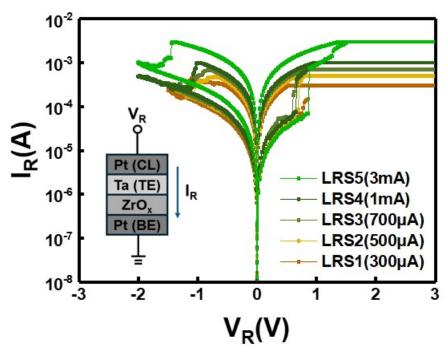
**Figure S3.** I–V characteristics of Device 2 and Device 4 remeasured after 50 days, showing stable resistive switching over 15 cycles. The preserved switching functionality confirms excellent environmental stability and long-term operational reliability of the devices.



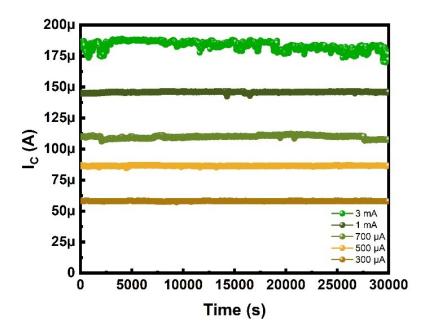
**Figure S4.** (a-e) Temperature-dependent I–V characteristics of the memristor measured at 25 °C, 50 °C, 75 °C, 100 °C and 125 °C under a fixed compliance current of 1 mA. (f) Overlap of representative I–V characteristics measured at 25 °C, 50 °C, 75 °C, 100 °C, and 125 °C.



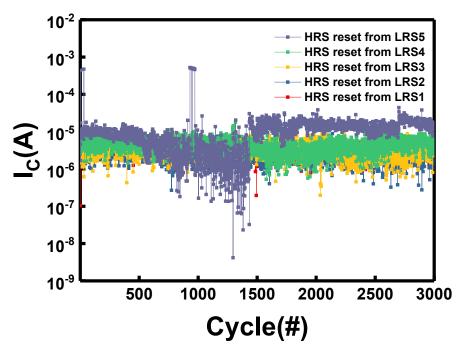
**Figure S5.** Transfer characteristics of the commercial NMOS transistor (2N7000). (a) Output curves under different  $V_g$  levels demonstrate well-controlled saturation current. This  $V_g$ -tunable current-limiting behavior enables the transistor to serve as an effective current compliance (CC) component during the SET process.



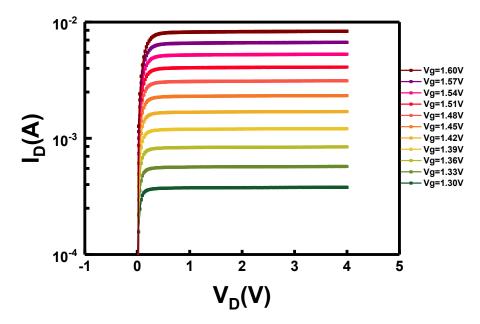
**Figure S6.** DC sweep curves of multilevel resistance states measured from a single memristor device under direct compliance current control. The linearity is highly consistent with that observed in the transistor–memristor series configuration, confirming that the current-limiting function does not distort intrinsic device behavior.



**Figure S7.** Retention performance of LRS1–LRS5 states measured on a single memristor at a read voltage of 0.1 V under 85 °C for 30,000 s, showing stable current without noticeable degradation, confirming excellent thermal stability and data retention capability.



**Figure S8.** Endurance characteristics of HRS obtained from different initial LRS states (LRS1 to LRS5). The results demonstrate that the reset process consistently leads to a stable HRS, regardless of the preceding LRS, confirming the device's immunity to prior state history and its reliable switching repeatability.



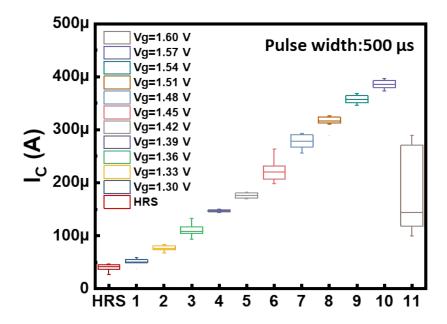
**Figure S9.**  $I_DV_D$  characteristics of the 2N7000 transistor measured under gradually increased gate voltages ( $V_g = 1.30$  to 1.60 V), corresponding to programmed level 1 to 11, showing distinct and well-separated current levels.

## Incremental Vg Device 1 Device 2 Device 3 Device 4 Device 5 Device 8 Device 6 Device 7 Device 9 Device 10 € 450 \_0 300 Program level Program level Decremental Vg Device 1 Device 2 Device 3 Device 4 Device 5 8 7 6 5 4 3 Program level Device 6 Device 7 Device 8 Device 9 Device 10

**Figure S10.** Statistical analysis of device-to-device (D2D) uniformity for 11 programmable levels measured from ten devices fabricated under identical conditions.

<u>300</u>

8 7 6 5 4 3 Program level



**Figure S11.** Box plot of read current distributions for 11 programmable levels obtained under a 500 μs pulse width. The results show ten well-defined low-resistance states and one high-resistance state (HRS).

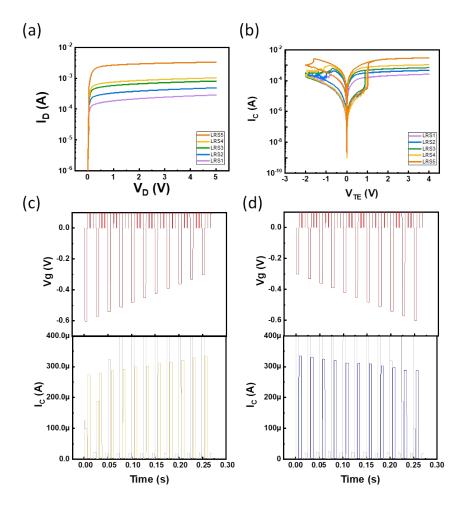


Figure S12. Validation of multilevel operation robustness using a commercial transistor (2SK170). (a)  $I_D$ - $V_D$  characteristics of the 2SK170 transistor measured under gradually increased gate voltages (Vg = -0.3 V, -0.46 V, -0.48 V, -0.53 V and -0.55 V), corresponding to programmed LRS1 to LRS5, showing distinct and well-separated current levels. (b) Multilevel resistance states (LRS) are achieved by varying the Vg of a series-connected 2N7000 transistor. The applied Vg are -0.3 V, -0.46 V, -0.48 V, -0.53 V and -0.55 V. (c, d) Incremental ( $-0.6 \rightarrow -0.3 \text{ V}$ ) and decremental ( $-0.3 \rightarrow -0.6 \text{ V}$ ) Vg programming schemes and their corresponding experimental results, showing the current responses  $I_C$  during programming and corresponding statistical distributions of the read currents for each level.

Table S1 HRS Current read at -0.1 V corresponding to different LRS states

State	HRS current@-0.1 V		
LRS5 (3mA)	7.34E-06 A		
LRS4 (1mA)	2.53E-06 A		
LRS3 (700 μA)	5.28E-06 A		
LRS2 (500 μA)	6.00E-06 A		
LRS1 (300 μA)	3.46E-06 A		

 $\label{eq:comparison} \textbf{Table S2} \ \ \text{Comparison of Transistor-Memristor Architectures for Multistate and } \ V_g$   $\ \ \text{Programming}$ 

Device structure (T+R)	Number of States	Retention	Endurance	$ m V_g$ pulse programing	Bidirectional Multistate	Ref.
2N(7000)+ Pt/Ta/ZrO <sub>x</sub> /Pt	8	> 10000 s (25 °C)	>3000	<b>√</b>	<b>√</b>	This work
NMOS+ Ta/TaO <sub>x</sub> /Pt	6	-	>106	<b>√</b>	×	[1]
ITO/SiN/TaN (Only R)	8	> 10000 s (25 °C)	>1000	-	<b>√</b>	[2]
NMOS+ Ti/HfO <sub>2</sub> /TiN	6	> 10000 s (125 °C)	$>5x10^5$	×	×	[3]
CMOS+ TiN/Ti/HfO <sub>2</sub> /TiN	9	-	10	×	×	[4]
NMOS+ TiN/Ti/HfO <sub>2</sub> /TiN	9	-	-	<b>√</b>	×	[5]

## **Reference:**

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