

Standards for the Characterization of Endurance in Resistive Switching Devices

Mario Lanza,* Rainer Waser, Daniele Ielmini, J. Joshua Yang, Ludovic Goux, Jordi Suñe, Anthony Joseph Kenyon, Adnan Mehonic, Sabina Spiga, Vikas Rana, Stefan Wiefels, Stephan Menzel, Ilia Valov, Marco A. Villena, Enrique Miranda, Xu Jing, Francesca Campabadal, Mireia Gonzalez, Fernando Aguirre, Felix Palumbo, Kaichen Zhu, Juan Baustista Roldan, Francesco Maria Puglisi, Luca Larcher, Tuo-Hung Hou, Themis Prodromakis, Yuchao Yang, Peng Huang, Tianqing Wang, Yang Chai, Kin Leong Pey, Nagarajan Raghavan, Salvador Dueñas, Tao Wang, Qiangfei Xia, and Sebastian Pazos



Cite This: <https://doi.org/10.1021/acsnano.1c06980>



Read Online

ACCESS |

Metrics & More

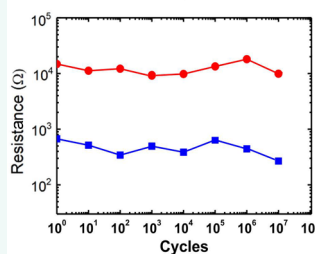
Article Recommendations

ABSTRACT: Resistive switching (RS) devices are emerging electronic components that could have applications in multiple types of integrated circuits, including electronic memories, true random number generators, radiofrequency switches, neuromorphic vision sensors, and artificial neural networks. The main factor hindering the massive employment of RS devices in commercial circuits is related to variability and reliability issues, which are usually evaluated through switching endurance tests. However, we note that most studies that claimed high endurance $>10^6$ cycles were based on resistance *versus* cycle plots that contain very few data points (in many cases even <20), and which are collected in only one device. We recommend not to use such a characterization method because it is highly inaccurate and unreliable (*i.e.*, it cannot reliably demonstrate that the device effectively switches in every cycle and it ignores cycle-to-cycle and device-to-device variability). This has created a blurry vision of the real performance of RS devices and in many cases has exaggerated their potential. This article proposes and describes a method for the correct characterization of switching endurance in RS devices; this method aims to construct endurance plots showing one data point per cycle and resistive state and combine data from multiple devices. Adopting this recommended method should result in more reliable literature in the field of RS technologies, which should accelerate their integration in commercial products.

KEYWORDS: resistive switching, memristor, memory, variability, reliability, characterization, metal-oxide, endurance

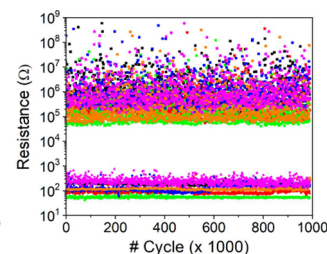
EXTREMELY WEAK ENDURANCE CLAIM

Read the resistance one/few times per decade
Show data for only one device



HIGHLY RELIABLE ENDURANCE CLAIM

Read the resistance in every cycle
Show data for several devices



Resistive switching (RS) devices are materials systems in which two or more metallic electrodes are connected to an insulating or semiconducting material, whose electrical resistance can be adjusted to specific values (*i.e.*, states) by applying electrical stresses.¹ Most RS devices reported to date have exhibited two resistance states, often referred to as high-resistance state (HRS) and a low-resistance state (LRS), although RS devices with up to 100 states (often referenced with numbers from 1 to n) have been also reported.² Depending on the number of resistance states and their stability, RS devices may be used for different applications. For example, RS devices exhibiting one non-volatile state and one volatile state can be used as selectors to minimize sneak path currents in crossbar array circuits³ and are

being considered for the hardware implementation of 44 electronic neurons in deep neural networks (DNNs)⁴ and 45 spiking neural networks (SNNs).^{5,6} RS devices exhibiting two 46 nonvolatile states have been employed to construct radio- 47 frequency switches,^{7,8} logic gates,^{9–11} stochastic computing 48 systems,^{12,13} and nonvolatile memories (NVM).^{14–16} RS 49

Received: August 14, 2021

Accepted: October 7, 2021

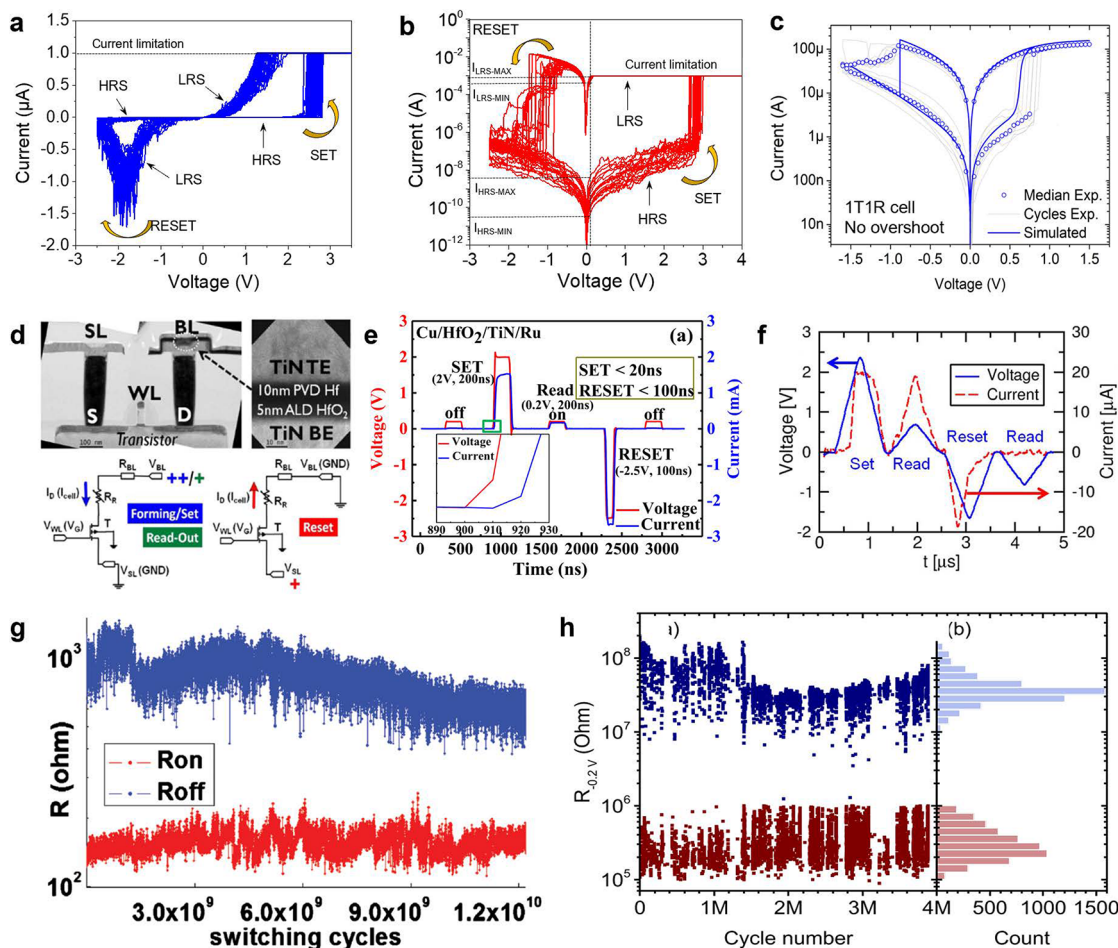


Figure 1. Characterization of the resistive switching phenomenon. (a, b) Current-limited I - V plots demonstrating the presence of nonvolatile bipolar RS in Au/h-BN/Au devices, using linear and logarithmic current scales, respectively. Each plot shows several lines measured in the same device, displaying the cycle-to-cycle variability of the currents. In panel (b) the overshoot can be seen, as the reset current is higher than the current limitation; this indicates that the current limitation tool from the SPA did not act immediately. Reprinted with permission from ref 36. Copyright 2020 Nature. (c) I - V curves collected in a memristor connected to a transistor. The current limitation takes place immediately and there is no overshoot. Reprinted with permission from ref 118. Copyright 2021 MDPI. (d) Cross-sectional TEM image of a 1T1R cell and equivalent electrical circuit used for forming, set and reset polarization. Reprinted with permission from ref 27. Copyright 2012 IEEE. (e) I - t plot demonstrating the presence of nonvolatile bipolar RS in Cu/HfO₂/TiN/Ru devices when applying rectangular PVS. Reprinted with permission from ref 33. Copyright 2017 IEEE. (f) I - t plot demonstrating nonvolatile bipolar RS in Ti/HfO_x/TiN device when applying triangular PVS. Reprinted with permission from ref 34. Copyright 2015 IEEE. (g, h) R_{HRS} and R_{LRS} vs cycle plots displaying the write endurance of RS devices with different compositions. (g) Reprinted with permission from ref 91. Copyright 2010 AIP Publishing. (h) Reprinted with permission from ref 147. Copyright 2014 AIP Publishing.

50 devices exhibiting multiple nonvolatile and stable states are
51 being considered for the hardware implementation of
52 electronic synapses in DNNs, as they allow implementing
53 computing algorithms (e.g., backpropagation) by updating and
54 maintaining multiple conductance states in each training
55 iteration (often referred to as epoch).^{17,18}

56 Despite multiple device structures exhibiting RS have been
57 reported (i.e., planar junctions, memtransistors),^{19,20} so far the
58 only one being considered by the semiconductor industry is
59 the crossbar array of vertical metal/insulator/metal (MIM)
60 nanocells.^{21–23} The use of crossbar arrays of RS devices for the
61 aforementioned applications is interesting because: (i) this
62 circuit architecture is reasonably easy to fabricate (it requires
63 few lithography steps, less than, for example, a transistor),
64 which makes it also relatively cheap;¹ (ii) it offers a very high
65 integration density (~ 100 Gbit/cm²),²⁴ which can be further
66 enhanced by using three-dimensional (3D) configurations;²⁵
67 (iii) their electrical properties can be tuned (by using different

metallic and insulating materials) to fit the technological
requirements of different applications;²¹ and (iv) the yield,
variability, reliability, and stability reported are the best among
all RS device architectures.^{14–16,26} It is worth noting that many
industrial crossbar arrays employ one transistor in series with
each RS device.^{27–32} At the same time, this configuration
(often referred as one-transistor/one-resistor, or 1T1R
structure) provides a superior control when programming
the conductance of each RS device, and it reduces the
integration density and increases the complexity of the
fabrication process.

The two typical figures-of-merit confirming the presence of
RS in bistable devices are (i) current vs voltage (I - V) plots
created during the application of ramped voltage stresses
(RVS) and (ii) current vs time (I - t) plots created during the
application of pulsed voltage stresses (PVS). When applying
RVS, the presence of RS can be confirmed by the detection of 84
a current increase (HRS-to-LRS transition, i.e., set) and a 85

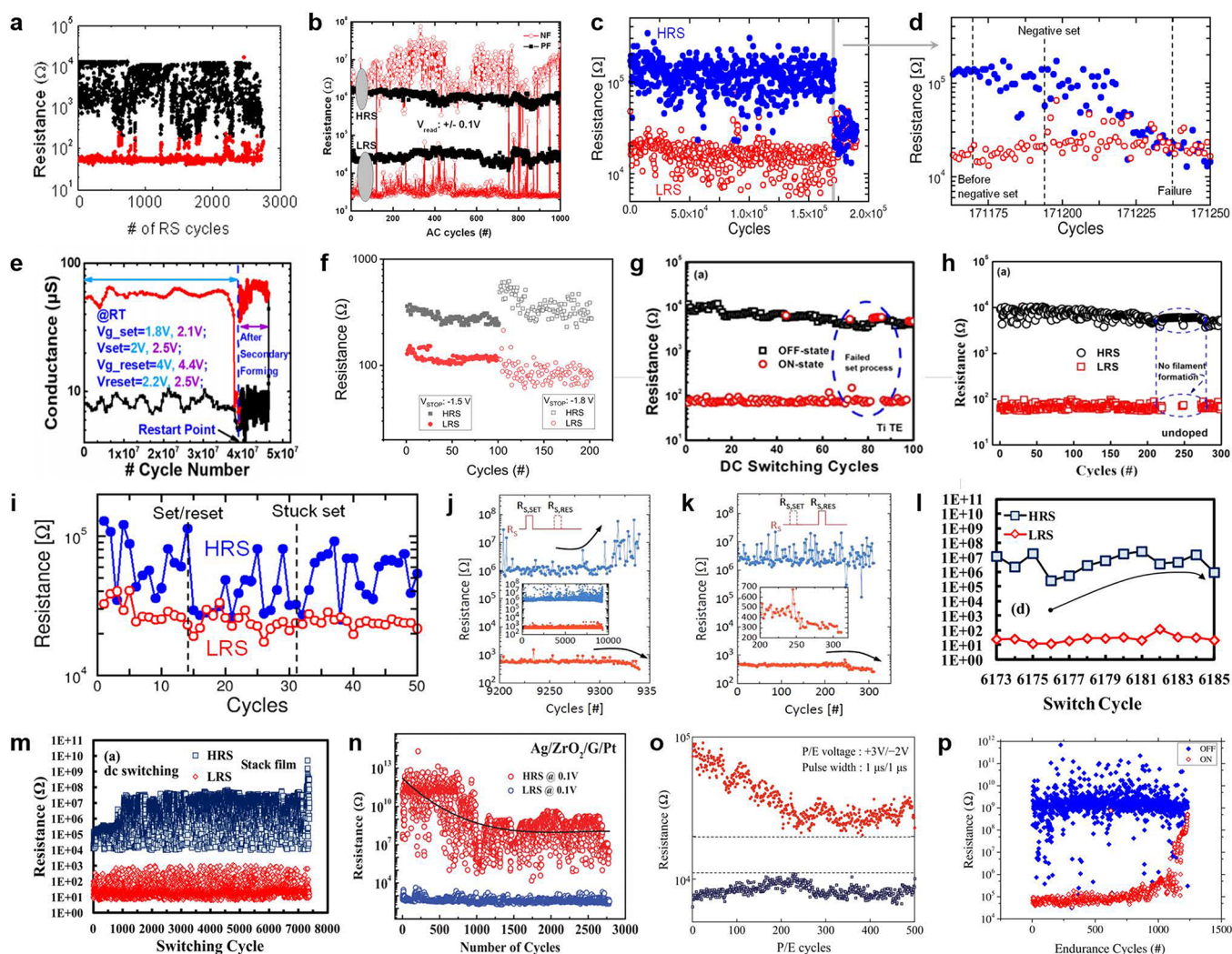


Figure 2. Examples of endurance plots showing undesired and unexpected fluctuations on the values of R_{HRS} and R_{LRS} as the RS device is switched for several cycles. (a) Clear stochastic fluctuations. Reprinted with permission from ref 39. Copyright 2011 AIP Publishing. (b) Similar stochastic fluctuations. Reprinted with permission from ref 40. Copyright 2012 Springer. (c) A progressive decrease of the value of R_{HRS} until the device gets stuck in the LRS. (d) Magnified image of (c) at the gray vertical line. Reprinted with permission from ref 34. Copyright 2015 IEEE. (e) A sudden decrease of the resistance in R_{HRS} , and the device becomes stuck in the LRS. Reprinted with permission from ref 49. Copyright 2019 IEEE. (f) How the values of R_{HRS} and R_{LRS} change with the magnitude of the stress applied, which changes the value of $R_{\text{HRS}}/R_{\text{LRS}}$ and the variability. Reprinted with permission from ref 51. Copyright 2020 MDPI. (g–i) Examples of endurance plots in which the device does not switch for some cycles. (g) Reprinted with permission from ref 52. Copyright 2017 Nature. (h) Reprinted with permission from ref 53. Copyright 2016 ACS. (i) Reprinted with permission from ref 34. Copyright 2015 IEEE. (j–o) Endurance plots of some devices that experience resistance and variability shift as the stress proceeds. (j, k) Reprinted with permission from ref 54. Copyright 2016 Nature. (l, m) Reprinted with permission from ref 55. Copyright 2016 AIP Publishing. (n) Reprinted with permission from ref 56. Copyright 2016 Wiley-VCH. (o) Reprinted with permission from ref 57. Copyright 2015 Springer. (p) Failure of a device because it becomes stuck in R_{HRS} . Reprinted with permission from ref 58. Copyright 2017 AIP Publishing.

86 current decrease (LRS-to-HRS transition, *i.e.*, reset) in the I – V
 87 plot (see Figure 1a); this produces that, for a given read
 88 voltage (typically ~ 0.1 V), the currents read in the forward and
 89 backward sweeps are different (*i.e.*, there is a gap between
 90 them), and the resistances (R_{HRS} and R_{LRS}) can be calculated.
 91 This plot is also often given as the logarithm of the absolute
 92 value of the current (see Figure 1b), which allows visualizing
 93 the $R_{\text{LRS}}/R_{\text{HRS}}$ ratio much more clearly. It is worth noting the
 94 RVS aimed to collect hysteretic I – V curves are normally
 95 current limited, either using the semiconductor parameter
 96 analyzer (Figure 1b) or using a series transistor (Figures 1c,d)
 97 or resistor. Collecting I – V sweeps can be very slow, especially
 98 when registering small currents < 1 nA, and in some cases
 99 measuring one single RS cycle can take tens of seconds and up

to 1 min. When applying PVS, pulses with relatively high
 100 voltages (typically $> \pm 2$ V) are used to induce the state
 101 transitions, and read pulses with small voltages (typically ~ 0.1
 102 V) are intercalated to read the current; then, the values of R_{HRS}
 103 and R_{LRS} are calculated dividing the read voltage by the average
 104 current detected during the read pulses. Most researchers
 105 employ PVS with a rectangular shape (Figure 1e),³³ although
 106 triangular PVS have also been employed (Figure 1f);³⁴ this
 107 could be considered another type of PVS. The I – t plots
 108 obtained during PVS may allow quantifying the set and reset
 109 times (t_{SET} and t_{RESET}), which could be used to calculate the
 110 set and reset energies (E_{SET} and E_{RESET}). Moreover, all RS
 111 technologies work under PVS; for these reasons, and because
 112 (in general) it is much faster, the application of PVS is the
 113

114 most recommended method to study RS devices. Confirming
115 the presence of RS in multistate devices is also carried out
116 through the same figures-of-merit ($I-V$ and $I-t$ plots). Still, in
117 such cases determining the state resistances and the switching
118 voltages, energies and times reliably are often more complex
119 due to the smaller differences between the states (*i.e.*, the value
120 of R_{LRS}/R_{HRS} in a bistable RS device is normally much higher
121 than the value of R_{n+1}/R_n in a multistate device, where $n + 1$ is
122 the state adjacent to n that is more conductive than n).³⁵

123 One of the most important properties of RS devices for any
124 application is the write cycling endurance. In the field of RS,
125 this is defined as the maximum number of programming cycles
126 that a device can undergo before its electrical characteristics
127 start to deviate outside the allowed ranges (often called the
128 operation window),²⁶ even if that happens only temporarily
129 during one cycle. In bistable RS devices, one operating cycle is
130 defined as one set transition plus one reset transition;¹ in
131 multistate RS-based devices, the definition of cycling
132 endurance is not so well established, although some authors
133 defined it as the number of transitions from the most to the
134 least resistive states stopping at all intermediate states (*i.e.*,
135 potentiation) and back to the most resistive state (*i.e.*,
136 depression).² The required electrical characteristics of RS
137 devices and their allowed ranges for multiple electronic
138 technologies are described in detail in ref 21. For example,
139 the typical operation windows for RS devices used as NVM are
140 $R_{HRS}/R_{LRS} > 10$, V_{SET} and $V_{RESET} < 1$ V, E_{SET} and $E_{RESET} < 10$
141 pJ, t_{SET} and $t_{RESET} < 10$ ns, t_{HRS} and $t_{LRS} > 10$ years, where R , V ,
142 E , and t denote resistance, voltage, energy, and time,
143 respectively, and t_{HRS} and t_{LRS} are the duration of the
144 resistances states if no electrical stress is applied, often referred
145 as retention times. In RS devices for other applications,
146 additional figures-of-merit may also be important, such as LRS
147 resistance and HRS capacitance in radiofrequency switches
148 (which should be $< 50 \Omega$ and < 2 fF, respectively),⁷ switching
149 slope in selectors and electronic neurons (which should be < 1
150 mV/decade),³⁶ and linearity in multistate electronic synapses
151 for ANNs (which is evaluated through the linearity factor, c , in
152 the equation $G = G_{MIN} + G_0[1 - e^{-cN}]$, which ideally should be
153 0). In this expression G is the conductance of a given state,
154 G_{MIN} is the minimum conductance, G_0 is a constant value
155 describing the conductance window, and N is the number of
156 pulses of equal voltage applied.^{37,38}

157 After a number of operating cycles, the electrical character-
158 istics of RS devices may start to deviate until failure, either
159 because the devices become stuck at one specific resistance
160 state or because their electrical characteristics stop matching
161 the technological requirements or their allowed tolerances.
162 Therefore, characterizing and quantifying the endurance of RS
163 devices is critical to assess their reliability and potential for
164 integration in commercial electronic systems. As measuring all
165 the parameters of RS devices (*e.g.*, R_{HRS} , R_{LRS} , V_{SET} , V_{RESET} ,
166 E_{SET} , E_{RESET} , t_{SET} , t_{RESET} , t_{HRS} , t_{LRS} , linearity, capacitance, *etc.*)
167 for a large number of cycles is not feasible, the indicator
168 selected in most cases to quantify the cycling endurance is the
169 resistance, and plots showing the values of R_{HRS} and R_{LRS} vs
170 cycle number (often referred as endurance plot) have been
171 presented (see Figure 1g,h). Unfortunately, the endurance
172 plots reported in many research articles have been constructed
173 using methods that are highly questionable; this has created a
174 blurry vision of the real endurance (and reliability) of RS
175 devices. In this review, we aim at clarifying which are the most
176 convenient methods to quantify the endurance of RS devices

and to summarize which endurance claims have been 177
supported by enough data and which require further 178
demonstrations. 179

180 FAILURE MECHANISMS

In RS devices, the set and reset transitions are induced by 181
applying electrical stresses to the metallic electrodes, which 182
produce the modification of the atomic structure of the 183
insulator or semiconductor between them.¹ While the 184
magnitude of the stress-induced structural changes can be 185
roughly adjusted by tuning the voltage, duration, and 186
separation of the PVS, accurate control of the number of 187
atoms moved in each cycle and their position in the device has 188
never been achieved and it is considered to be impossible.^{12,13} 189
Therefore, the conductance in LRS (after set) and in HRS 190
(after reset) can be very different after each RS cycle (Figure 191
2a,b),^{39,40} and the switching voltages, times, and energies may 192
also be slightly different in each cycle,³³ which is why it is often 193
said that the RS is a stochastic phenomenon.^{12,13} In fact, the 194
cycle-to-cycle variations of R_{HRS} , R_{LRS} , V_{SET} , V_{RESET} , E_{SET} , 195
 E_{RESET} , t_{SET} , and t_{RESET} are so unpredictable that they have 196
been employed as an entropy source in true random number 197
generators (TRNG) and physical unclonable functions (PUF) 198
for data encryption.⁴¹⁻⁴³ 199

Most MIM-like RS devices fail during cycling because of the 200
degradation of their microstructure, often associated with the 201
irreversible penetration of metallic atoms from the electrodes 202
and/or the formation of atomic vacancies in the insulator (*e.g.*, 203
O vacancies in HfO_2);^{44,45} this results in a permanent LRS (see 204
Figure 2c,d).³⁴ Thermal effects related to the high currents 205
flowing in LRS can also contribute to accelerate the 206
degradation of the insulator, as they promote atomic 207
diffusion.⁴⁶⁻⁴⁸ In this article, we are not examining all the 208
physical, chemical, and thermal phenomena producing 209
incorrect/undesired ionic motion in RS devices resulting in 210
endurance failure, for two reasons: (i) RS devices have been 211
fabricated using many different materials and discussing all of 212
them in detail would be very space consuming; and (ii) 213
multiple articles have claimed switching mechanisms and 214
movement of ions without any solid evidence, just drawing 215
speculative schematics based on imagination, and that are not 216
supported by any nanochemical measurement or atomistic 217
simulation, which are the recommended characterization 218
methods. There is a huge amount of misleading and erroneous 219
literature in this field, and we do not want to promote it. Even 220
in the case of oxygen vacancies in transition-metal oxides, 221
which has been studied in hundreds of articles, the community 222
still did not agree on where the oxygen atoms go (some articles 223
claim that they form a reservoir at the interface, and some 224
others that it interacts with the electrode forming a metal 225
oxide). For more information related to the endurance failure 226
mechanisms of each specific RS device, please explore the 227
literature about a specific materials combination. 228

The degradation of the microstructure of an insulator is a 229
process that might take place suddenly or progressively 230
depending on the materials employed and their thicknesses. 231
For example, ref 34 shows that in $\text{Ti}/\text{HfO}_x/\text{TiN}$ devices, the 232
values of R_{HRS} , R_{LRS} , and R_{HRS}/R_{LRS} experienced a progressive 233
drift until the two states became indistinguishable (see Figure 234
2c,d). On the contrary, ref 49 shows that $\text{TiN}/\text{HfO}_x/\text{TEL}/$ 235
 TiN devices reached the LRS abruptly (see Figure 2e); TEL 236
stands for thermal enhanced layer, but the authors of this 237
article did not share its composition. In addition, the same 238

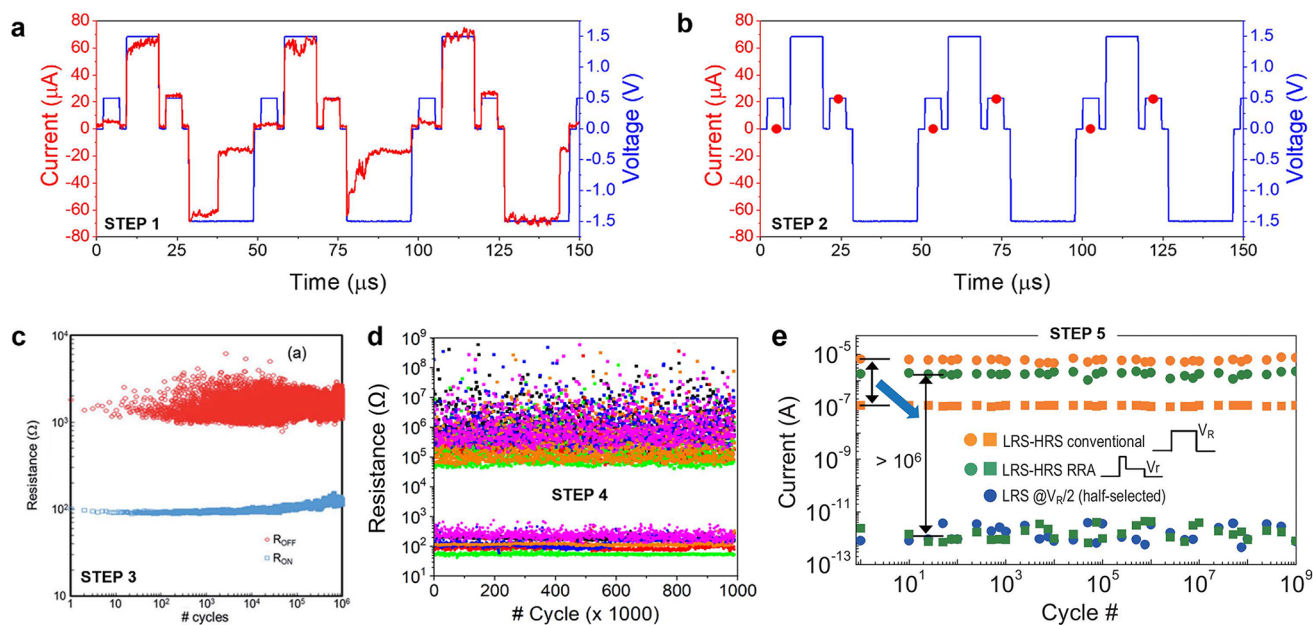


Figure 3. Recommended process to characterize the endurance of RS devices. (a) PVS displaying the currents through a MIM-like RS device when a sequence of read, write, read, and erase pulses is applied. (b) PVS with the same sequence as (a) but measuring current only during the read pulses. The read time is schematically indicated with a red ball. (c) Endurance of a Pt/Ta₂O₅/Ta device, showing the resistance in each cycle. Reprinted with permission from ref 176. Copyright 2017 Wiley-VCH. (d) Illustration of endurance plot for 10⁶ cycles for five devices. (e) Endurance plot showing a few data points per decade, aiming to characterize high endurance >10⁶ cycles. This test would be acceptable when only applied after steps 1–4 have been conducted. It is highly recommended to provide >50 points per decade for 10 devices. Reprinted with permission from ref 90. Copyright 2015 IEEE.

239 device may exhibit progressive endurance degradation when
 240 applying some specific electrical stresses and sudden endurance
 241 degradation for others (e.g., larger voltage, current, duration).⁵⁰
 242 The values of R_{HRS} , R_{LRS} , and $R_{\text{HRS}}/R_{\text{LRS}}$ and their dispersions
 243 also depend on the stresses applied (see Figure 2f).⁵¹
 244 Moreover, sometimes the resistance of a RS device can get
 245 unpredictably stuck at one state for some time (i.e., it stops
 246 switching even if read, write, and erase pulses are applied) and
 247 recover the normal functioning suddenly.⁵² For example, if in
 248 one set event too many ions in the MIM cell have been
 249 displaced, the voltage applied to induce the reset might not be
 250 high enough. In such cases, triggering the reset may require
 251 several stresses in order to get those impurities to drift back,
 252 which means that the device is stuck in the LRS for some
 253 cycles (see Figure 2g,h),^{52,53} and conversely, if in one cycle the
 254 reset event displaces too many ions, in the following cycle the
 255 voltage applied to induce the set may not be high enough;
 256 increasing again the conductance of the device may require the
 257 application of multiple stresses, leading to the device being
 258 stuck in the HRS for some cycles (see Figure 2g-i).^{34,52,53} In
 259 addition, R_{HRS} , R_{LRS} , and their dispersions may suddenly or
 260 progressively change (see Figure 2j-o),^{54–57} resulting in an
 261 alteration of the energy consumption per state transition. In
 262 the worst case, these unexpected changes can result in $R_{\text{HRS}}/$
 263 R_{LRS} being too low (i.e., <10) for some cycles, or even
 264 permanently (see Figure 2o).⁵⁷
 265 It is also worth noting that RS devices may stick in an
 266 irreversible HRS (Figure 2p) because the metallic wires of the
 267 crossbar array melt due to the high currents driven in the
 268 LRS,⁵⁸ which promotes electromigration.⁵⁹ This has been
 269 often observed in RS studies coming from academia (where
 270 the stability of the materials is poorer and the contamination is
 271 higher than in the industry), especially when studying devices
 272 with wires narrower than 100 nm.

In multistate RS-based electronic synapses, endurance failure
 can be understood as a change in the number of resistance
 states per potentiation and depression cycle beyond an
 acceptable range. This can also be understood as a prohibitive
 change in the number of electrical stresses that one needs to
 apply to increase/decrease the resistance of the devices
 between the required value.

ENDURANCE CHARACTERIZATION METHOD

For all the above reasons, characterizing the cycling endurance
 of RS devices requires measuring their electrical properties in
 every cycle; otherwise, one has a very high probability that the
 switching is ineffective for some cycles, which leads to an
 overestimation of the endurance lifetime. Measuring R_{HRS} and
 R_{LRS} in every cycle is especially important when studying RS
 devices made of advanced materials (e.g., 2D materials,
 MXenes, perovskites) and nanostructures (e.g., nanowires,
 nanotubes, memtransistors), as their switching mechanisms
 and their reliability have not been demonstrated by different
 groups, and therefore, they are still controversial and not
 widely accepted. Unfortunately, many studies in the field of RS
 do not show robust enough evidence of high endurance, as
 they did not measure the resistance in every cycle; several
 studies even claimed endurance >10⁶ cycles showing a plot
 with even <20 data points per state (in total),^{60–86} which are
 insufficient to confirm the device reliability. Registering the
 current in few cycles also raises a doubt whether the applied
 electrical stress is the optimal biasing condition for practical
 applications. Moreover, most studies only display the
 endurance plot of one single device, ignoring how the
 dispersion of R_{HRS} and R_{LRS} varies from one device to another.
 These insufficient characterization results make it hard to
 evaluate the potential of the RS devices.

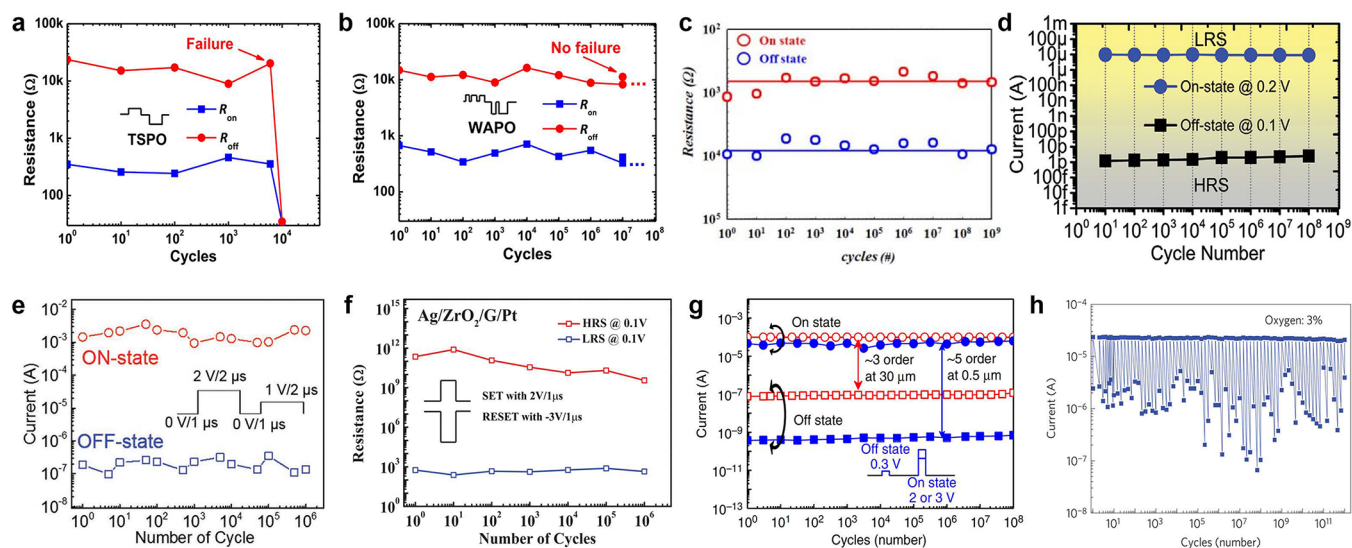


Figure 4. Examples of plots that do not provide enough data to reliably support their endurance claims. In these studies, R_{HRS} and R_{LRS} have not been measured in every cycle, so it cannot be confirmed that the devices actually switched so many times, and hence the cycle-to-cycle variability by be underestimated. (a, b) Reprinted with permission from ref 177. Copyright 2015 API Publishing. (c) Reprinted with permission from ref 68. Copyright 2014 IEEE. (d) Reprinted with permission from ref 100. Copyright 2021 Wiley-VCH. (e) Reprinted with permission from ref 101. Copyright 2018 Wiley-VCH. (f) Reprinted with permission from ref 56. Copyright 2016 Wiley-VCH. (g) Reprinted with permission from ref 62. Copyright 2013 Nature. (h) Reprinted with permission from ref 123. Copyright 2011 Nature.

305 The correct method to characterize the endurance of RS
 306 devices requires five steps, which need to be carried out
 307 sequentially. The first step consists of applying a sequence of
 308 read, write, read, and erase PVS during few (*i.e.*, <10) cycles
 309 and measuring the current simultaneously with a high temporal
 310 resolution (*e.g.*, 100 data points per voltage pulse). The typical
 311 resulting plot (Figure 3a) can be used to confirm the presence
 312 of RS by (i) observing the set and reset transitions and (ii)
 313 comparing the average values of R_{HRS} and R_{LRS} during the read
 314 pulses. However, the number of data points that standard
 315 measuring equipment can register in one sequence of PVS (*i.e.*,
 316 one run) is limited. For example, the Keysight B1500A and the
 317 Keithley 4200—two semiconductor parameter analyzers
 318 (SPA) widely used to study RS devices—cannot register
 319 more than 5000 data points per run. Using a temporal
 320 resolution of 100 data points per cycle, that means ~ 50 cycles.
 321 If one wants to measure more data points, then one needs to
 322 first store all the data recorded and then run the PVS sequence
 323 again. While this operation can be automated with the software
 324 of the SPA, it produces an inherent delay of ~ 30 s between
 325 each run that impedes the analysis of long endurance of
 326 millions of cycles. Sophisticated measuring setups may employ
 327 a software (*e.g.*, Labview, Matlab) to control the instrumenta-
 328 tion and to read the data buffer of the SPA while it is
 329 measuring, so that the stress does not need to be interrupted.⁸⁷
 330 In any case, plotting such a huge amount of data points may be
 331 very time-consuming and unfeasible for standard spreadsheet
 332 software (*e.g.*, Excel, Origin). Ref 88 presents a commercial
 333 integrated circuit for the characterization of crossbar arrays of
 334 RS devices; this circuit can overcome the limitations of the
 335 SPA in terms of data collection, and a recent study presented a
 336 high-speed amplifier capable to measure 10^5 I - V sweeps per
 337 second.⁸⁹

338 To avoid these problems, one can apply the PVS sequence
 339 and only register one current data point in each read pulse, as
 340 shown in Figure 3b (step 2). While this produces a loss of
 341 valuable information (*i.e.*, writing/erase currents, t_{SET} , t_{RESET} ,

E_{SET} , E_{RESET}), it allows measurement of the values of R_{HRS} and R_{LRS}
 342 during a few thousands of cycles in each PVS. By
 343 repeating this measurement (*e.g.*, using the loop tool in most
 344 commercial SPAs), it is relatively easy and fast to construct an
 345 endurance plot displaying the values of R_{HRS} and R_{LRS} for a few
 346 millions of cycles (see Figure 3c). Beyond 10^6 – 10^7 cycles, the
 347 methodology described in steps 2–3 to measure endurance
 348 could be very time-consuming. For example, if RS is measured
 349 using read, write, read, and erase PVS with a duration and
 350 interval of 1 μs (*i.e.*, time per RS cycle ~ 8 μs), the total time
 351 needed to measure endurance of 10^{12} cycles would be ~ 92
 352 days. Despite the fact that measuring R_{HRS} and R_{LRS} for $>10^{10}$
 353 cycles is possible and strongly recommended,⁸⁴ an alternative
 354 and acceptable measurement protocol would be as follows.
 355 First, repeat the measurement of endurance up to 10^6 – 10^7
 356 cycles for multiple devices to confirm that the pulse voltages,
 357 duration, and interval produce acceptable switching and that
 358 the values of R_{HRS} , R_{LRS} , and $R_{\text{HRS}}/R_{\text{LRS}}$ fit the technological
 359 requirements for each cycle and for each device (step 4). This
 360 is very important because variability is one of the most
 361 important problems of RS technologies.²⁶ One good way to do
 362 it is by presenting the resistance vs cycle plot for multiple
 363 devices overlapped (see Figure 3d). The more devices and
 364 cycles that one measures, the better, as it will allow a deeper
 365 understanding of not only the materials system employed but
 366 also the effect of the electrical stresses applied. Second, apply
 367 an additional endurance measurement on the same devices,
 368 keeping only few read pulses (randomly selected) per decade
 369 to speed-up the measurement, however extending the number
 370 of cycles far above 10^6 – 10^7 cycles, so that an endurance plot
 371 with enough statistical validity can be constructed (see Figure
 372 3e, *i.e.*, step 5).
 373

This method is similar to quality controls in which only a
 374 certain number of events and/or products of the total
 375 population are analyzed; therefore, again, the more cycles
 376 that one measures, the better. This method has been employed
 377 by companies to characterize the endurance of metal/TMO/
 378

Table 1. Endurance Reported for 2D Materials-Based RS Devices^a

ref	structure	device structure and device sizes	endurance claim and data points	test method
119	G/MoS _{2-x} O _x /G	vertical MIM 25 μm × 25 μm	over 2 × 10 ⁷ 72 data points	pulse voltage stress
100	Pt/hBN/Ag	vertical MIM 250 nm × 250 nm	10 ⁸ 8 data points	pulse voltage stress
158	Au/MoS ₂ /Au	planar FET channel length (2 μm)	15 15 data points	I–V sweeps
20	Au/MoS ₂ /Au	planar FET channel length (1–5 μm)	475 475 data points	I–V sweeps
159	Au/Li _x MoS ₂ /Au	planar FET channel length (~5 μm)	1000 1000 data points	pulse voltage stress
160	Au/MoS ₂ /Au	vertical MIM 2 μm × 2 μm	150 150 data points	I–V sweeps
161	Ag/GaSe/Ag	planar FET channel length (~30 μm)	5000 indistinguishable	I–V sweeps
162	Au/Ti/MoS ₂ /Ti/Au	planar FET channel length (~2 μm)	6 6 data points	pulse voltage stress
163	Au/α-In ₂ Se ₃ /Au	planar FET channel length (~1 μm)	40 40 data points	I–V sweeps
164	Cu/MoS ₂ /Au	vertical MIM 2 μm × 2 μm	21 21 data points	I–V sweeps
165	Au/h-BN/Au	vertical MIM 1 μm × 1 μm	50 50 data points	I–V sweeps
36	Ag/h-BN/Au	vertical MIM 150 nm × 150 nm	80,000 80,000 data points	pulse voltage stress

^aHigh endurance (>1 × 10⁶ cycles) have been only supported using plots that display few data points, and when the values of R_{HRS} and R_{LRS} are measured in each cycle, the maximum endurance recorded has been 80,000 cycles. Researchers in the field of RS should always use the correct characterization method (*i.e.*, report R_{HRS} and R_{LRS} in every cycle) instead of trying to oversell the endurance of their devices using an inaccurate method (*i.e.*, measure one/few data points per decade). The letters “G” and “h-BN” stand for graphene and hexagonal boron nitride, respectively.

379 metal RS devices that showed highly reliable switching
380 behavior when applying steps 1–4, and values >10⁹ cycles
381 have been achieved (see Figure 3e).^{3,90} Engineering the shape
382 of the PVS to minimize reading times and maximize
383 performance has also been reported.³ In any case, measuring
384 the value of R_{HRS} and R_{LRS} for >10¹⁰ cycles is possible (see
385 Figure 1g)⁹¹ and is the preferred way to do it.

386 DISCUSSION AND PROSPECTS

387 We have noticed that a considerable number of publications in
388 the field of RS claimed very high endurance >10⁷ cycles based
389 on a plot that contains few (*i.e.*, < 100) data points for only
390 one device (see Figure 4), that is, ignoring steps 1–4. Such a
391 simplified method is less reliable than the one described in the
392 previous section because it cannot confirm that the device has
393 actually switched in every cycle. As mentioned, the value of
394 V_{SET}, V_{RESET}, t_{SET}, and t_{RESET} can notoriously change in every
395 cycle, and it could be possible that the application of the same
396 pulse voltage or pulse duration could not induce the state
397 transition or could damage the device. Furthermore, the
398 endurance plots in Figure 4 ignore some of the most important
399 challenges of RS technologies, which are cycle-to-cycle and
400 device-to-device variability.²⁶

401 In many cases endurance plots like those in Figure 4 have
402 been created by applying a PVS without reading the current
403 and interrupting the stress to read the resistance by applying
404 either a PVS or an RVS. This process is different than the one
405 described in step 5 of Figure 3 (see previous section), as it
406 requires interrupting the stress and introduces a relaxation time
407 (of the order of seconds).⁹² This relaxation time may affect the
408 state and its properties (*e.g.*, V_{SET}, V_{RESET}),⁹³ which means that

the switching conditions might no longer be appropriate. It is
therefore very important that same values of delays are applied
between write pulses over the whole endurance test. The use of
a RVS to read the current is even worse because the authors
would be claiming high endurance without having confirmed
that the device can switch when applying a PVS. This is
important because an RVS will always reach V_{SET} and V_{RESET}
and the device may switch; however, when applying a fast PVS,
the voltage or duration may not be sufficiently high to always
guarantee the switching, or else may be too high and could
damage the device. Moreover, most RS studies employ RVS
with a current limitation to prevent the degradation of the
device, while this function is not available when applying the
PVS with commercial SPA. Current limitation during PVS
could also be achieved using a resistor or transistor in series
with the RS cell; however, it should not be forgotten that
conventional SPA systems do not provide sufficiently fast
current limitation (during RVS) to prevent device damage
during the set events, plus add additional parasitic capacitance
effects; all of this has an impact in the endurance.^{94–96} The
overshot current caused by the parasitic capacitances leads to
an excessive leakage current across the RS device after the
forming and set process. In most cases it accelerates the device
degradation. Overall, the total integrated stress during a RVS
normally exceeds that of a PVS by orders of magnitude;
therefore, the two types of measurements are not comparable.
Disclosing details about the measuring protocol is very
important.

In addition, MIM-like RS devices with different composi-
tions (*i.e.*, materials, thicknesses) are likely to have different
optimal biasing conditions. As their right balance is crucial for

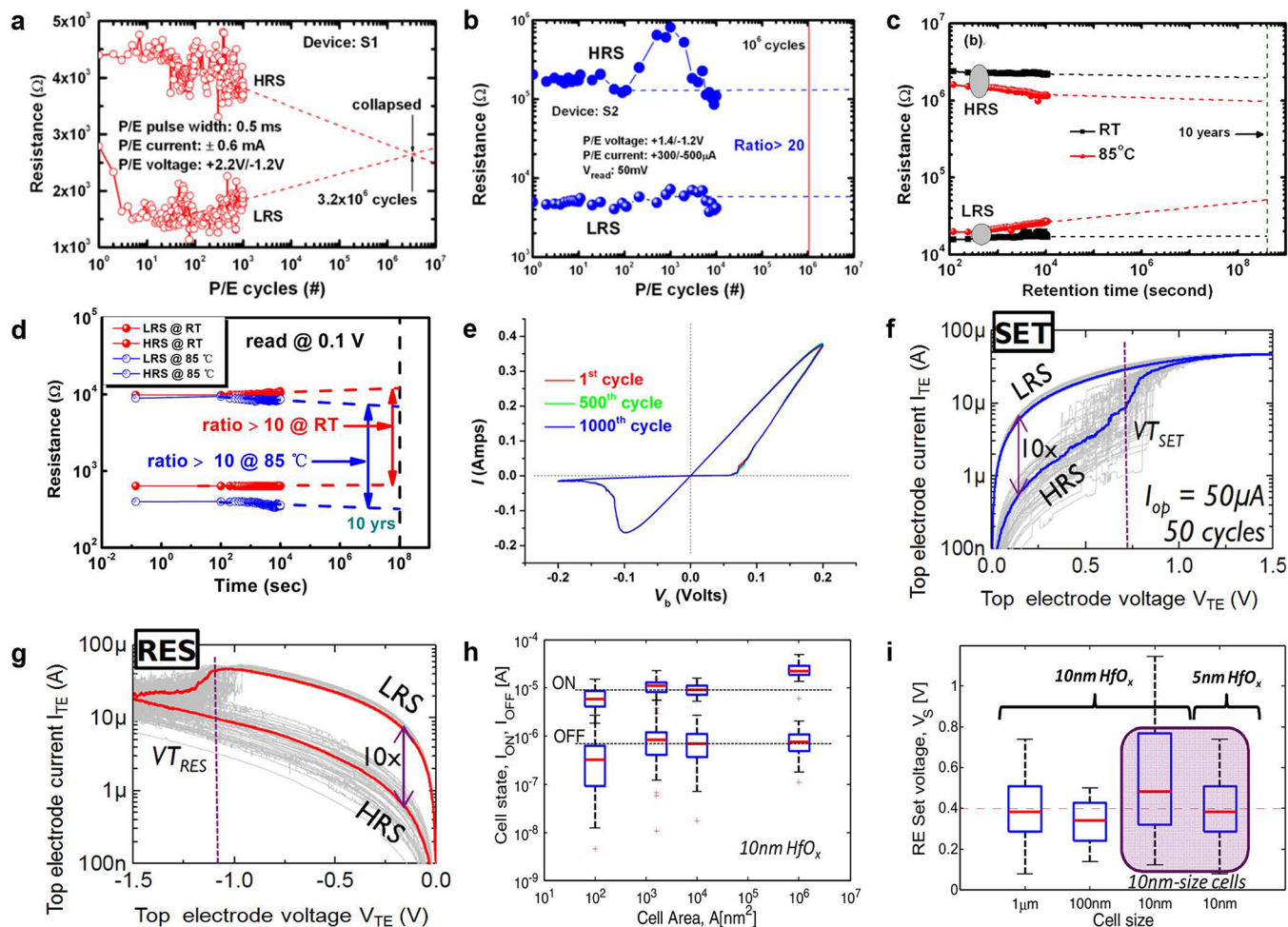


Figure 5. Additional considerations related to the estimation of endurance of RS devices. (a, b) Resistance vs cycle plot presenting a claim of endurance based on the drawing of dashed lines. This method is unreliable. The method used in Figure 3 is recommended. Reprinted with permission from ref 112. Copyright 2012 Springer. (c, d) Current vs time plot measured at a constant voltage to characterize the retention time of the RS device in HRS and LRS. The retention claim of 10 years is unreliable because it is based on extrapolation over several orders of magnitude. The correct method to measure the retention time of RS devices is explained in ref 49. (c) Reprinted with permission from ref 40. Copyright 2012 Springer. (d) Reprinted with permission from ref 113. Copyright 2013 Springer. (e) Current vs voltage plots for the 1st, 500th, and 1000th cycles of a MoS₂-based RS device. This method presenting the data is problematic, as it cannot confirm that the devices switched during all the 1000 cycles and ignores cycle-to-cycle variability. Reprinted with permission from ref 114. Copyright 2016 ACS Publishing. (f, g) Current vs voltage plots measured during 50 cycles in some state-of-the-art MIM-like RS devices fabricated in industrial facilities. These plots indicate the real variability of RS devices, and such a type of characterization is recommended. (f, g) Reprinted with permission from ref 115. Copyright 2014 IEEE. (h, i) Dispersion of the state currents and switching voltages in state-of-the-art MIM-like RS devices fabricated in an industrial facility. Reprinted with permission from ref 22. Copyright 2011 IEEE.

440 a high endurance,⁹⁷ the comparison between different devices
 441 becomes challenging. The popular strategy of using a fixed set
 442 of switching parameters for different stacks might lead to a
 443 false endurance quantification. If the write scheme is optimized
 444 for one specific stack, a potentially better stack could show
 445 inferior endurance due to suboptimal biasing. We think that
 446 this strategy is only valid if the biasing conditions are strictly
 447 specified by the application. In other cases, we recommend
 448 optimizing the switching parameters by adaptive programming
 449 for each tested stack (or even for each device) and comparing
 450 the optimized endurance. A respective algorithm has been
 451 demonstrated.⁹⁸

452 In order to emphasize how important the measuring
 453 protocol is, Table 1 summarizes the highest cycling endurance
 454 ever reported for RS devices made (totally or partially) of 2D
 455 materials and indicates the structure and size of the device, the
 456 method used to characterize the endurance, and the number of

points shown in each plot. As it can be observed, all endurance
 457 claims >10⁵ cycles have been made presenting plots that
 458 contain <100 data points, while when measuring R_{HRS} and R_{LRS}
 459 in each cycle, the maximum endurance achieved is 80,000
 460 cycles. The fact that the endurance registered strongly depends
 461 on the characterization method indicates their important effect
 462 on the device reliability. High endurance have been only
 463 achieved when measuring few cycles, which means that this
 464 method ignores failure mechanisms. Overall, making endurance
 465 claims based on plots showing few (*i.e.*, < 100) data
 466 points is risky and can be unreliable, especially if the authors
 467 are using advanced nanomaterials (*e.g.*, 2D materials, MXene
 468 or perovskites) in which the endurance has never been tested
 469 in each cycle before.

470
 471 As explained in the previous section, in the context of RS
 472 devices measuring R_{HRS} and R_{LRS} for the first 10⁶–10⁷ cycles,
 473 they only take a few minutes or hours, so we do not see any

Table 2. Highest Switching Endurances of RS Devices Ever Reported and the Number of Data Points Presented to Support Such Claims^a

ref	structure	endurance claimed	data points presented	reliability of the claim
136	ITO(or Au)/[Ru(L) ³] ²⁺ /ITO	10 ¹²	indistinguishable	high
90	Pt/TaO _x /Ta	>10 ¹⁰	indistinguishable	high
147	Au/Ni/PR/Pt/Co/BiFeO ₃ /Ca _{0.96} Ce _{0.04} MnO ₃ / YAlO ₃	4 × 10 ⁶	indistinguishable	high
98	Ta/ZrO ₂ /Pt	10 ⁶	indistinguishable	high
97	TiN/TiO _x /HfO _x /TiN	10 ⁶	indistinguishable	high
169	Ag/10%Sb-GeS ₂ /W	>10 ⁵	indistinguishable	high
179	Pt/Ta/TaO _x :Zr/Pt	2.9 × 10 ¹⁰	indistinguishable	medium
131	Pt/TaO _x /Pt	>10 ⁹	indistinguishable	medium
178	Ag/a-Si/poly-Si	>10 ⁸	indistinguishable	medium
166	Pd/Ag/HfO _x /Ag/Pd	10 ⁸	indistinguishable	medium
151	Ta/TaO _x /Pt	10 ⁶	indistinguishable	medium
146	Au/Ti/BFO/BFTO/Pt	>2 × 10 ⁵	indistinguishable	medium
127	Au/BTCO/Pt/Ti/SiO ₂ /Si	10 ⁵	indistinguishable	medium
141	Pt/BST/SRO/STO	10 ⁵	indistinguishable	medium
143	Al/PEDOT:PSS/PMMA/ITO/PET	10 ⁵	indistinguishable	medium
167	Ta/TaO _x /TiO ₂ /Ti	10 ¹⁵	42	low
123	Pt/Ta ₂ O _{5-x} /TaO _{2-x} /Pt	10 ¹²	75	low
120	Pt/TaO _x /Ta ₂ O ₅ /Pt	>10 ¹²	50	low
4	Pt/TaO _x /Ta ₂ O ₅ /Pt	10 ¹²	23	low
155	TE/SLT/BE (1S)	10 ¹¹	110	low
	1S1R	10 ⁵	16	low
168	AlO ₃ /Ta ₂ O _{5-x} /TaO _y	10 ¹¹	33	low
145	Ta/HfO ₂ /Pt	1.2 × 10 ¹¹	34	low
60	Ag/Ag ₃₃ Ge ₂₀ Se ₄₇ /Ag	>10 ¹⁰	~5	low
128	TiN/Hf/HfO ₂ /TiN	10 ¹⁰	31	low
	TiN/Ti/HfO ₂ /TiN	10 ¹⁰	31	low
	TiN/Ta/HfO ₂ /TiN	10 ⁶	25	low
150	TiN/HfO ₂ /TiN	10 ¹⁰	60	low
140	TiN/Hf/HfO ₂ /TiN	10 ¹⁰	31	low
67	Ti/HfO ₂ (1)/O ₂ -HfO ₂ (9)/TiN	10 ¹⁰	11	low
149	Pt/AlO ₂ /Ta ₂ O _{5-x} /TaO _y /Pt	10 ¹⁰	32	low
68	TiN/Gd:SiO ₂ /ITO	10 ⁹	10	low
61	Cu/HfO ₂ /Pt (1T1R)	~10 ⁹	13	low
62	TiN/As-Ge-Te-Si-N/TiN (or Ni)	>10 ⁸	17	low
77	TiN/TiON/HfO _x /Pt	>10 ⁸	22	low
135	TiN/Ti/HfO ₂ /TiN	>10 ⁸	80	low
126	Pt/Au/MgO/Co ₃ O ₄ /Pt/Au	>10 ⁸	74	low
69	Al/PFCF-rGO/ITO	10 ⁸	9	low
70	Al/GO-PVK/ITO	10 ⁸	9	low
71	Al/TPAPAM-GO/ITO	10 ⁸	9	low
139	Al/PS- <i>b</i> -P4VP-GO/ITO	10 ⁸	25	low
72	Ti/AlN/Pt/Ti/SiO ₂ /Si	10 ⁸	9	low
148	TiN/AsTeGeSiN/TiN/Pt	10 ⁸	23	low
169	Si ₃ N ₄ /Ta/Si ₃ N ₄ /Ta/Si ₃ N ₄	10 ⁸	40	low
32	TiN/Ti/HfO _x /TiN	10 ⁸	~130	low
33	Cu/HfO ₂ /TiN/Ru	10 ⁸	8	low
134	ITO/HfO _x /ITO	5 × 10 ⁷	~150	low
117	Pt/SiO ₂ :Pt/Ta	3 × 10 ⁷	80	low
119	G/MoS _{2-x} O _x /G	>2 × 10 ⁷	70	low
144	TiN/RTO WO _x /W/TiN	>10 ⁷	~100	low
170	Cu/Cu-Te/GdO _x /W	>10 ⁷	44	low
73	TiN/a-C:H/Pt	10 ⁷	9	low
74	ITO/AlN/ITO	10 ⁷	9	low
152	TaN/ZrO ₂ /HfO ₂ /TiN	10 ⁷	63	low
154	Pt/Zn:SiO _x /TiN	10 ⁷	70	low
76	Cu/nanohole-graphene/HfO ₂ /Pt/Cu/HfO ₂ /Pt	10 ⁷	11	low
22	TiN/Hf/HfO ₂ /TiN	10 ⁷	27	low
76	Cu/Cu-doped SiO ₂ /W	10 ⁷	6	low
63	Al/P3BT:PCBM/Al/PET	10 ⁷	15	low
125	Pt/TiO ₂ /TiN/Pt	2 × 10 ⁶	24	low

Table 2. continued

ref	structure	endurance claimed	data points presented	reliability of the claim
13	Ag/GeSe/Si ₃ N ₄ /W	>10 ⁶	11	low
64	Pt/Nb-doped SrTiO ₃ /Pt	>10 ⁶	7	low
65	HZO/LSMO/LAO	>10 ⁶	13	low
66	Ag-GeSe-Si ₃ N ₄ -W	>10 ⁶	5	low
111	TiN/Hf/TaO/HfAlO/AlO/TiN	10 ⁶	21	low
32	TiN/Ti/HfO _x /TiN	10 ⁶	20	low
129	TiN/Ti/HfO _x /TiN	10 ⁶	~80	low
130	Au/Pt/Bi _{1-δ} FeO ₃ / SrRuO ₃ /SrTiO ₃	10 ⁶	55	low
132	Ru/Ta ₂ O ₅ /TiO ₂ /Ru	10 ⁶	~50	low
133	TiN/WO _x /TiN	10 ⁶	~70	low
137	noble metal/NiO/noble metal	10 ⁶	~80	low
142	TiN/Ti/HfO _x /TiN	10 ⁶	260	low
78	Pt/ZrO _x /HfO _x /TiN	10 ⁶	7	low
121	Pt/PCMO/Al	10 ⁶	37	low
153	Pt/ZnO/Pt	10 ⁶	60	low
166	Pt/Ni: SiO ₂ /TiN	10 ⁶	54	low
171	Al/TiN/Cu/TiW/Al ₂ O ₃ /W	10 ⁶	25	low
173	Pt/HfO _x /ZrN _x	10 ⁶	64	low
122	Cu-GST/TiTe _x /SiO ₂ /W	2 × 10 ⁵	24	low
138	Al/PMMA/MLG/PMMA/ITO/PET	1.5 × 10 ⁵	~200	low
79	Pt/ZrO _x /HfO _x /TiN	>10 ⁵	6	low
157	Pt/NP SiO _x /Pt	>10 ⁵	25	low
175	W/Ge _{0.4} Se _{0.6} /Cu/Al	>10 ⁵	48	low
80	IrO _x /Al ₂ O ₃ /Ge NW _S /SiO ₂ /Si MOS structure	>10 ⁵	12	low
81	Ni/HfO _x /n ⁺ Si	>10 ⁵	18	low
82	Ni/GeO _x /HfON/TaN	10 ⁵	12	low
83	Pt/SrTiO _x /Si	10 ⁵	5	low
84	Al/PMMA/GO/PMMA/ITO	10 ⁵	7	low
85	Al/TiO _x /Al	10 ⁵	11	low
86	Cu/pV ₃ D ₃ /Al	10 ⁵	9	low
124	TiN/HfO _x /AlO _x /Pt	10 ⁵	37	low
156	Pt/a-CO _x /SiO ₂ /W			
	Ti/a-CO _x /SiO ₂ /W	5 × 10 ⁴	40	low
	W/a-CO _x /SiO ₂ /W			

^aMany studies in the field of RS claimed endurance >10⁵ cycles based on plots that contain fewer than 50 data points, which are collected from one single device. Such claims need to be supported by much more abundant data, if possible, and collected on multiple devices. The rows with medium reliability are those for which, while showing abundant data, it looks like multiple cycles were not measured or the points were just skipped when plotted (which was not specified in the article).

474 convincing reason for not including such data in every RS
 475 publication. It is worth noting that the recommended method
 476 to characterize the endurance (Figure 3) requires reading the
 477 current during the pulsed voltage stress. The most common
 478 way to do this experiment is to (i) use an additional module of
 479 the SPA (e.g., Keysight B1500A and Keithley 4200) that can
 480 apply a PVS and record current simultaneously and (ii)
 481 connect the RS device in series with a resistor, apply the PVS
 482 with the SPA, and measure the voltage of the resistor using a
 483 mixed-signal oscilloscope (although the resolution of the
 484 oscilloscope is much worse than that of the SPA,⁹⁹ especially
 485 when trying to measure R_{HRS}). Some authors, when asked to
 486 provide the values of R_{HRS} and R_{LRS} in every cycle during
 487 revisions in journals, argue that they do not have such setups.
 488 In such cases, collaborating with other scientists or simply
 489 making no claim of endurance would be better than claiming a
 490 value without enough supported statistical data. Some papers
 491 showed a plot like the one in Figure 3a to demonstrate correct
 492 switching (which confirms that they have the required
 493 hardware), but they select not to measure R_{HRS} and R_{LRS} in
 494 every cycle.^{58,100–103} This practice should be avoided, as
 495 measuring 10⁶ cycles applying PVS with a duration of 1 μ s

would take only a few minutes or hours; if the device does not
 switch in 1 μ s, it is not useful for most RS technologies. 497

Regarding the number of devices to measure, it is obvious 498
 that the more the better (as for any other electronic device) 499
 and establishing a specific number to consider an article 500
 reliable would be subjective and controversial. As for any other 501
 product, controlling the quality of 100% of items fabricated for 502
 the entire required lifetime is not feasible. For this reason, 503
 quality controls in which only a small percentage of devices are 504
 tested are often applied. These processes bring an associated 505
 inherent trade-off: development time *vs* accuracy; and in most 506
 cases, companies do not reveal details about them. Never- 507
 theless, it is known that the standard qualification procedure 508
 for memory endurance in the industry requires statistical 509
 results from a sufficiently large memory array (>kilobit 510
 size).^{104,105} This characterizes endurance under the influences 511
 of cycle-to-cycle evolution and device-to-device variation. 512
 Therefore, a low bit error rate after cycling guarantees robust 513
 endurance. However, the array-level analysis with many devices 514
 is typically not available in the early development stage and in 515
 the academic research. Furthermore, the process control on the 516
 device variation is not optimized for most studies. In any case, 517

we note that publications in top journals exploring other electronic devices (e.g., FETs) often present data (i.e., mobility, subthreshold swing) for >50 devices;^{106–109} however, in the field of RS, the number of devices analyzed is (in general) strikingly lower, and most publications present data for one or few (i.e., < 5) devices, especially in terms of endurance. Combined with the aforementioned low number of data points presented in many cases, the claim of high endurance is particularly unreliable. We think that presenting endurance data for at least 10 devices would be more reliable and at the same time feasible. This number is in line with the statistical reproducibility statements of some relevant journals.¹¹⁰ A useful validation metric would be to plot the distributions of resistance states obtained for steps 1–4 and compare them with the distributions obtained for step 5, which would allow detecting any onset of degradation mechanisms.¹¹¹ Some companies also use verified-endurance tests, using algorithms like increasing-step-pulse-programming.¹⁰¹

Finally, we would like to point out some much less reliable claims of high endurances presented in some articles, consisting of measuring few cycles and drawing extended dashed lines (see Figure Sa,b).¹¹² Similar unwarranted extrapolations have also been done for HRS and LRS retention times (Figure 5 c,d).^{40,113} Presenting a few current vs voltage (I – V) curves corresponding to a handful of selected cycles (see Figures 5e) also leads to underestimations of cycle-to-cycle variability.¹¹⁴ With this method, the devices show a low variability of switching voltages and state currents, when the device currents could be experiencing larger variations from one cycle to another. The best variability in the switching voltages and state resistances demonstrated by visualizing multiple cycles in state-of-the-art RS devices fabricated at industrial facilities is always much larger (see Figures 5f,g).¹¹⁵

Another important aspect when analyzing the endurance of RS devices is the lateral size. One should always keep in mind that achieving a very high endurance (> 10^8 cycles) in RS devices with a large lateral size (> $25\ \mu\text{m}^2$) does not mean that smaller ($\sim 100\ \text{nm}^2$) RS devices with similar composition will exhibit a similar endurance.¹¹⁵ First, narrower metallic wires could easily melt if the currents in LRS are too high; and second the concentration of native defects (necessary to promote RS)^{1,2,6,44} may be much lower in smaller devices, and if it reaches a critical value, the devices may not switch. Note that the switching voltages and state currents strongly depend on the device size, as shown in Figure 5h,i.²² It may happen for large cells that when the filament degrades and gets stuck into a high-resistive state during the endurance test, another filament is created and takes over the cycling. This phenomenon might occur several times, and its probability to happen is much higher for large than for scaled RS devices.

Some of the most prominent researchers in the field of RS usually present values of R_{HRS} and R_{LRS} in every cycle when making endurance claims.^{90,116,117} This is also the method used by the industry before trying to commercialize RS devices for different technologies, independently if they show that in papers or not (sometimes articles from companies do not disclose enough information due to confidentiality issues). Therefore, it would be much better if the entire community working on RS devices always support endurance claims by measuring R_{HRS} and R_{LRS} in every cycle (see Figure 1e,f and Figure 3c,d). To provide the readers with a practical summary of the endurances of RS devices, in Table 2 we have summarize all the studies that we found in the literature claiming an

endurance > 10^5 cycles and indicate the number of data points presented.^{172,174}

CONCLUSION

The cycling endurance of RS devices has been mainly characterized using two different methods. The first one consists of stressing the devices and measuring the values of R_{HRS} and R_{LRS} after every set/reset transition; this method is highly reliable and should always be employed, at least for the first 10^6 – 10^7 cycles. The second one consists of stressing the devices without measuring R_{HRS} and R_{LRS} and stopping the stress after some cycles to read the values of R_{HRS} and R_{LRS} ; this method is not recommended because it cannot confirm the switching of the device in every cycle, it introduces delays affecting the RS properties, it underestimates cycle-to-cycle variability of switching voltages and state currents, and it also overestimates the endurance lifetime. Table 2 presents a summary of the highest endurances ever claimed for RS devices and the number of data points presented in those studies. In our opinion, authors of studies in the field of RS reporting endurance plots should always describe in detail their measuring protocol, specifically indicating if they read R_{HRS} and R_{LRS} in each cycle and how many data points they are displaying. Clear communication of these critical experimental details will help readers to understand the real cycling endurance of the RS devices developed in each study and should accelerate the commercialization of RS technologies.

AUTHOR INFORMATION

Corresponding Author

Mario Lanza – Physical Science and Engineering Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia; orcid.org/0000-0003-4756-8632; Email: mario.lanza@kaust.edu.sa

Authors

Rainer Waser – Peter-Grünberg-Institut (PGI-7), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany; Peter-Grünberg-Institut (PGI-10), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany; Institut für Werkstoffe der Elektrotechnik 2 (IWE2), RWTH Aachen University, Aachen 52074, Germany

Daniele Ielmini – Dipartimento di Elettronica, Informazione e Bioingegneria, Politecnico di Milano and IUNET, Milano 20133, Italy

J. Joshua Yang – Department of Electrical and Computer Engineering, University of Southern California, Los Angeles, California 90089, United States

Ludovic Goux – Imec, 3001 Leuven, Belgium

Jordi Suñe – Departament d'Enginyeria Electrònica, Universitat Autònoma de Barcelona, Barcelona 08193, Spain

Anthony Joseph Kenyon – Department of Electronic and Electrical Engineering, University College London, London WC1E 7JE, United Kingdom

Adnan Mehonic – Department of Electronic and Electrical Engineering, University College London, London WC1E 7JE, United Kingdom

Sabina Spiga – CNR-IMM, Unit of Agrate Brianza, Agrate Brianza (MB) 20864, Italy; orcid.org/0000-0001-7293-7503

Vikas Rana – Peter-Grünberg-Institut (PGI-10), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany

- 641 **Stefan Wiefels** – Peter-Grünberg-Institut (PGI-7),
642 Forschungszentrum Jülich GmbH, 52425 Jülich, Germany
- 643 **Stephan Menzel** – Peter-Grünberg-Institut (PGI-7),
644 Forschungszentrum Jülich GmbH, 52425 Jülich, Germany;
645 orcid.org/0000-0002-4258-2673
- 646 **Iliia Valov** – Peter-Grünberg-Institut (PGI-7),
647 Forschungszentrum Jülich GmbH, 52425 Jülich, Germany;
648 orcid.org/0000-0002-0728-7214
- 649 **Marco A. Villena** – Applied Materials Inc., Reggio Emilia 74L
650 42122, Italy
- 651 **Enrique Miranda** – Departament d'Enginyeria Electrònica,
652 Universitat Autònoma de Barcelona, Barcelona 08193, Spain
- 653 **Xu Jing** – School of Materials Science and Engineering, Jiangsu
654 Key Laboratory of Advanced Metallic Materials, Southeast
655 University, Nanjing 211189, China
- 656 **Francesca Campabadal** – Institut de Microelectrònica de
657 Barcelona-Centre Nacional de Microelectrònica, Consejo
658 Superior de Investigaciones Científicas, Bellaterra 08193,
659 Spain
- 660 **Mireia Gonzalez** – Institut de Microelectrònica de Barcelona-
661 Centre Nacional de Microelectrònica, Consejo Superior de
662 Investigaciones Científicas, Bellaterra 08193, Spain
- 663 **Fernando Aguirre** – Unidad de Investigación y Desarrollo de
664 las Ingenierías-CONICET, Facultad Regional Buenos Aires,
665 Universidad Tecnológica Nacional (UIDI-CONICET/
666 FRBA-UTN), Medrano 951 (C1179AAQ), Argentina
- 667 **Felix Palumbo** – Unidad de Investigación y Desarrollo de las
668 Ingenierías-CONICET, Facultad Regional Buenos Aires,
669 Universidad Tecnológica Nacional (UIDI-CONICET/
670 FRBA-UTN), Medrano 951 (C1179AAQ), Argentina
- 671 **Kaichen Zhu** – Physical Science and Engineering Division,
672 King Abdullah University of Science and Technology
673 (KAUST), Thuwal 23955-6900, Saudi Arabia
- 674 **Juan Baustista Roldan** – Departamento de Electrónica y
675 Tecnología de Computadores, Facultad de Ciencias,
676 Universidad de Granada, Granada 18071, Spain
- 677 **Francesco Maria Puglisi** – Dipartimento di Ingegneria “Enzo
678 Ferrari”, Università di Modena e Reggio Emilia, Modena
679 41125, Italy
- 680 **Luca Larcher** – Applied Materials Inc., Reggio Emilia 74L
681 42122, Italy
- 682 **Tuo-Hung Hou** – Department of Electronics Engineering and
683 Institute of Electronics, National Yang Ming Chiao Tung
684 University, Hsinchu 300, Taiwan; orcid.org/0000-0002-9686-7076
- 685
- 686 **Themis Prodromakis** – Centre for Electronics Frontiers,
687 University of Southampton, Southampton SO171BJ, United
688 Kingdom
- 689 **Yuchao Yang** – Key Laboratory of Microelectronic Devices and
690 Circuits (MOE), Department of Micro/nanoelectronics,
691 Peking University, Beijing 100871, China; orcid.org/0000-0003-4674-4059
- 692
- 693 **Peng Huang** – Key Laboratory of Microelectronic Devices and
694 Circuits (MOE), Department of Micro/nanoelectronics,
695 Peking University, Beijing 100871, China; orcid.org/0000-0003-3280-0099
- 696
- 697 **Tianqing Wang** – Department of Applied Physics, The Hong
698 Kong Polytechnic University, Kowloon, Hong Kong
- 699 **Yang Chai** – Department of Applied Physics, The Hong Kong
700 Polytechnic University, Kowloon, Hong Kong
- 701 **Kin Leong Pey** – Engineering Product Development,
702 Singapore University of Technology and Design (SUTD),
703 487372, Singapore; orcid.org/0000-0002-0066-091X
- Nagarajan Raghavan** – Engineering Product Development,
Singapore University of Technology and Design (SUTD),
487372, Singapore; orcid.org/0000-0001-6735-3108
- Salvador Dueñas** – Department of Electronics, University of
Valladolid, Valladolid E-47011, Spain
- Tao Wang** – Institute of Functional Nano and Soft Materials
(FUNSOM), Collaborative Innovation Center of Suzhou
Nano Science and Technology, Soochow University, Suzhou
215123, China
- Qiangfei Xia** – Department of Electrical and Computer
Engineering, University of Massachusetts, Amherst,
Massachusetts 01003-9292, United States
- Sebastian Pazos** – Physical Science and Engineering Division,
King Abdullah University of Science and Technology
(KAUST), Thuwal 23955-6900, Saudi Arabia
- Complete contact information is available at:
<https://pubs.acs.org/10.1021/acsnano.1c06980>
- Notes**
The authors declare no competing financial interest.
- ACKNOWLEDGMENTS**
This work has been supported by the generous Baseline
funding program of the King Abdullah University of Science
and Technology (KAUST).
- VOCABULARY**
Resistive switching device, electronic device that can alter
their electrical resistance when an electrical stress is applied.
They normally have a metal/insulator/metal structure (*i.e.*, two
electrodes), although some three-electrode configurations
(with a third electrode adjacent to the insulator) have been
proposed. The changes in the electrical resistance are normally
nonvolatile, although a small portion of devices also exhibit
volatile resistive switching; **memristor**, short for “memory” and
“resistor”, this is the fourth fundamental electronic component
(together with the resistor, capacitor and inductor), and it
relates electric charge and magnetic flux. The word memristor
has been often employed as a synonym of resistive switching
device, although one portion of the community working in this
field thinks that is not correct; **nonvolatile memory (NVM)**, a
type of computer memory that can store information even after
power is removed. This type of memory is used to store
information for long periods >10 years. The most employed
NVM is the NAND Flash. Emerging NVMs are phase-change
memories, resistive random-access memories, and magnetic
random-access memories, among others; **switching endurance**,
for devices exhibiting two stable states, endurance is
defined as the maximum number of programming cycles that a
device can undergo before its electrical characteristics start to
deviate outside the allowed ranges, even if that happens only
temporarily during one cycle. For multistate devices, there is
no consensus on what endurance is, although several authors
defined it as the number of transitions from the most to the
least resistive states stopping at all intermediate states and back
to the most resistive state; **crossbar array**, circuit containing
two groups of metallic bars separated by one insulator. First,
one group of parallel metallic bars is deposited on an insulating
substrate, then an insulator is deposited on the bars, and finally
the second group of bars (parallel to each other but
perpendicular to the first group) is deposited on top. This
configuration leads to multiple metal/insulator/metal cells at

763 the intersections between the bars, which serve as a resistive
764 switching device

765 REFERENCES

- 766 (1) Ielmini, D.; Waser, R. *Resistive Switching: From Fundamentals of*
767 *Nanoionic Redox Processes to Memristive Device Applications*; Wiley-
768 VCH: Weinheim, 2016.
- 769 (2) Zhang, W.; Gao, B.; Tang, J.; Li, X.; Wu, W.; Qian, H.; Wu, H.
770 Analog-Type Resistive Switching Devices for Neuromorphic Comput-
771 ing. *Phys. Status Solidi RRL* **2019**, *13*, 1900204.
- 772 (3) Jo, S. H.; Kumar, T.; Narayanan, S.; Lu, W. D.; Nazarian, H. 3D-
773 Stackable Crossbar Resistive Memory Based on Field Assisted
774 Superlinear Threshold (Fast) Selector. Proceedings from the 2014
775 *IEEE International Electron Devices Meeting*, San Francisco, CA,
776 December 15–17, 2014; IEEE: New York, 2014; pp 160–163.
- 777 (4) Chen, W.-H.; Li, K.-X.; Lin, W.-Y.; Hsu, K.-H.; Li, P.-Y.; Yang,
778 C.-H.; Xue, C.-X.; Yang, E.-Y.; Chen, Y.-K.; Chang, Y.-S. A 65nm
779 1Mb Nonvolatile Computing-in-Memory ReRAM Macro with Sub-
780 16ns Multiply-and-Accumulate for Binary DNN AI Edge Processors.
781 Proceedings from the *International Solid-State Circuits Conference*
782 (*ISSCC*), San Francisco, CA, February 11–15, 2018; IEEE: New
783 York, 2018; pp 494–496.
- 784 (5) Wang, W.; Pedretti, G.; Milo, V.; Carboni, R.; Calderoni, A.;
785 Ramaswamy, N.; Spinelli, A. S.; Ielmini, D. Learning of
786 Spatiotemporal Patterns in a Spiking Neural Network with Resistive
787 Switching Synapses. *Sci. Adv.* **2018**, *4*, 4752.
- 788 (6) Guo, Y.; Wu, H.; Gao, B.; Qian, H. Unsupervised Learning on
789 Resistive Memory Array Based Spiking Neural Networks. *Front.*
790 *Neurosci.* **2019**, *13*, 812.
- 791 (7) Kim, M.; Pallecchi, E.; Ge, R.; Wu, X.; Ducournau, G.; Lee, J. C.;
792 Happy, H.; Akinwande, D. Analogue Switches Made from Boron
793 Nitride Monolayers for Application in 5G and Terahertz Communi-
794 cation Systems. *Nat. Electron.* **2020**, *3*, 479–485.
- 795 (8) Pi, S.; Ghadiri-Sadrabadi, M.; Bardin, J. C.; Xia, Q. Nanoscale
796 Memristive Radiofrequency Switches. *Nat. Commun.* **2015**, *6*, 7519.
- 797 (9) Wang, Y.; Wu, F.; Liu, X.; Lin, J.; Chen, J.-Y.; Wu, W.-W.; Wei,
798 J.; Liu, Y.; Liu, Q.; Liao, L. High On/Off Ratio Black Phosphorus
799 Based Memristor with Ultra-Thin Phosphorus Oxide Layer. *Appl.*
800 *Phys. Lett.* **2019**, *115*, 193503.
- 801 (10) Balatti, S.; Ambrogio, S.; Ielmini, D. Normally-Off Logic Based
802 on Resistive Switches—Part I: Logic Gates. *IEEE Trans. Electron*
803 *Devices* **2015**, *62*, 1831–1838.
- 804 (11) Siemon, A.; Drabinski, R.; Schultis, M. J.; Hu, X.; Linn, E.;
805 Heitmann, A.; Waser, R.; Querlioz, D.; Menzel, S.; Friedman, J. S.
806 Stateful Three-Input Logic with Memristive Switches. *Sci. Rep.* **2019**,
807 *9*, 14618.
- 808 (12) Ielmini, D.; Wong, H.-S. P. In-Memory Computing with
809 Resistive Switching Devices. *Nat. Electron.* **2018**, *1*, 333–343.
- 810 (13) Gaba, S.; Sheridan, P.; Zhou, J.; Choi, S.; Lu, W. Stochastic
811 Memristive Devices for Computing and Neuromorphic Applications.
812 *Nanoscale* **2013**, *5*, 5872–5878.
- 813 (14) Liu, T. A 130.7 mm² 2-Layer 32 Gb ReRAM Memory Device
814 in 24 nm Technology. Proceedings from the *International Solid-State*
815 *Circuits Conference Digest of Technical Papers*, San Francisco, CA,
816 February 17–21, 2013; IEEE: New York, 2013; pp 210–212.
- 817 (15) Fackenthal, R.; Kitagawa, M.; Otsuka, W.; Prall, K.; Mills, D.;
818 Tsutsui, K.; Javanifard, J.; Tedrow, K.; Tsushima, T.; Shibahara, Y.
819 19.7 A 16 Gb ReRAM with 200 MB/s Write and 1 GB/s Read in 27
820 nm Technology. Proceedings from the *International Solid-State*
821 *Circuits Conference Digest of Technical Papers*, San Francisco, CA,
822 February 9–13, 2014; IEEE: New York, 2014; pp 338–340.
- 823 (16) Ito, S.; Hayakawa, Y.; Wei, Z.; Muraoka, S.; Kawashima, K.;
824 Kotani, H.; Kouno, K.; Nakamura, M.; Du, G. A.; Chen, J. F. ReRAM
825 Technologies for Embedded Memory and Further Applications.
826 Proceedings from the *International Memory Workshop (IMW)*, Kyoto,
827 Japan, May 13–16, 2018; IEEE: New York, 2018.
- 828 (17) Yao, P.; Wu, H.; Gao, B.; Tang, J.; Zhang, Q.; Zhang, W.; Yang,
829 J. J.; Qian, H. Fully Hardware-Implemented Memristor Convolutional
830 Neural Network. *Nature* **2020**, *577*, 641–646.
- (18) Cai, F.; Correll, J. M.; Lee, S. H.; Lim, Y.; Bothra, V.; Zhang, Z.;
831 Flynn, M. P.; Lu, W. D. A Fully Integrated Reprogrammable
832 Memristor-CMOS System for Efficient Multiply-Accumulate Oper-
833 ations. *Nat. Electron.* **2019**, *2*, 290–299. 834
- (19) Tian, H.; Guo, Q.; Xie, Y.; Zhao, H.; Li, C.; Cha, J. J.; Xia, F.;
835 Wang, H. Anisotropic Black Phosphorus Synaptic Device for
836 Neuromorphic Applications. *Adv. Mater.* **2016**, *28*, 4991–4997. 837
- (20) Sangwan, V. K.; Lee, H.-S.; Bergeron, H.; Balla, I.; Beck, M. E.;
838 Chen, K.-S.; Hersam, M. C. Multi-Terminal Memtransistors from
839 Polycrystalline Monolayer Molybdenum Disulfide. *Nature* **2018**, *554*,
840 500–504. 841
- (21) *International Roadmap for Devices and Systems 2020 Edition*;
842 IEEE: New York, 2020. <https://irds.ieee.org/editions/2020> (accessed
843 2021-05-25). 844
- (22) Govoreanu, B.; Kar, G.; Chen, Y.; Paraschiv, V.; Kubicek, S.;
845 Fantini, A.; Radu, I.; Goux, L.; Clima, S.; Degraeve, R. 10 × 10 nm²
846 Hf/HfO_x Crossbar Resistive RAM with Excellent Performance,
847 Reliability and Low-Energy Operation. Proceedings from the
848 *International Electron Devices Meeting (IEDM)*, Washington, DC,
849 December 5–7, 2011; IEEE: New York, 2011; pp 729–732. 850
- (23) Fujii, S.; Incorvia, J. A. C.; Yuan, F.; Qin, S.; Hui, F.; Shi, Y.;
851 Chai, Y.; Lanza, M.; Wong, H.-S. P. Scaling the CBRAM Switching
852 Layer Diameter to 30 nm Improves Cycling Endurance. *IEEE Electron*
853 *Device Lett.* **2018**, *39*, 23–26. 854
- (24) Levisse, A.; Giraud, B.; Noel, J.-P.; Moreau, M.; Portal, J.-M.
855 Rram Crossbar Arrays for Storage Class Memory Applications:
856 Throughput and Density Considerations. Proceedings from the
857 *Conference on Design of Circuits and Integrated Systems (DCIS)*,
858 Lyon, France, November 14–16, 2018; IEEE: New York, 2018. 859
- (25) Deng, Y.; Chen, H.-Y.; Gao, B.; Yu, S.; Wu, S.-C.; Zhao, L.;
860 Chen, B.; Jiang, Z.; Liu, X.; Hou, T.-H. Design and Optimization
861 Methodology for 3D RRAM Arrays. Proceedings from the *Internation-*
862 *al Electron Devices Meeting (IEDM)*, Washington, DC, December
863 9–11, 2013; IEEE: New York, 2013; pp 629–632. 864
- (26) Lanza, M.; Wong, H. S. P.; Pop, E.; Ielmini, D.; Strukov, D.;
865 Regan, B. C.; Larcher, L.; Villena, M. A.; Yang, J. J.; Goux, L.;
866 Belmonte, A.; Yang, Y.; Puglisi, F. M.; Kang, J.; Magyari-Köpe, B.;
867 Yalon, E.; Kenyon, A.; Buckwell, M.; Mehonic, A.; Shluger, A.; et al.
868 Recommended Methods to Study Resistive Switching Devices. *Adv.*
869 *Electron. Mater.* **2019**, *5*, 1800143. 870
- (27) Chen, Y. Y.; Govoreanu, B.; Goux, L.; Degraeve, R.; Fantini, A.;
871 Kar, G. S.; Wouters, D. J.; Groeseneken, G.; Kittl, J. A.; Jurczak, M.
872 Balancing SET/RESET Pulse for 10¹⁰ Endurance in HfO₂/Hf 1T1R
873 Bipolar RRAM. *IEEE Trans. Electron Devices* **2012**, *59*, 3243–3249. 874
- (28) Milo, V.; Anzalone, F.; Zambelli, C.; Pérez, E.; Mahadevaiah,
875 M.; Ossorio, Ó.; Olivo, P.; Wenger, C.; Ielmini, D. Optimized
876 Programming Algorithms for Multilevel RRAM in Hardware Neural
877 Networks. Proceedings from the *International Reliability Physics*
878 *Symposium (IRPS)*, Monterey, CA, March 21–25, 2021; IEEE: New
879 York, 2021. 880
- (29) Grenouillet, L.; Castellani, N.; Persico, A.; Meli, V.; Martin, S.;
881 Billoint, O.; Segaud, R.; Bernasconi, S.; Pellissier, C.; C. Jahan, C.;
882 CharpinNicollet, C.; Dezest, P.; Carabasse, C.; Besombes, P.; Ricavy,
883 S.; Tran, N.-P.; Magalhaes-Lucas, A.; Roman, A.; Boixaderas, C.;
884 Magis, T.; Bedjaoui, M.; Tessaie, A.; Seignard, F.; Mazon, S.; Landis,
885 M.; Vianello, E.; Molas, G.; Gaillard, F.; Arcamone, J.; Nowak, E.
886 16kbit 1T1R OxRAM Arrays Embedded in 28 nm FDSOI
887 Technology Demonstrating Low BER, High Endurance, and
888 Compatibility with Core Logic Transistors. Proceedings from the
889 *International Memory Workshop (IMC)*, Dresden, Germany, May 16–
890 19, 2021; IEEE: New York, 2021. 891
- (30) Nail, C.; Molas, G.; Blaise, P.; Piccolboni, G.; Sklenard, B.;
892 Cagli, C.; Bernard, M.; Roule, A.; Azzaz, M.; Vianello, E.; Carabasse,
893 C.; Berthier, R.; Cooper, D.; Pelissier, C.; Magis, T. Understanding
894 RRAM Endurance, Eetention and Window Margin Trade-Off Using
895 Experimental Results and Simulations. Proceedings from the *Internation-*
896 *al Electron Devices Meeting (IEDM)*, San Francisco, CA, December
897 3–7, 2016; IEEE: New York, 2016; pp 95–98. 898

- (31) Grossi, A.; Vianello, E.; Zambelli, C.; Royer, P.; Noel, J.; Giraud, B.; Perniola, L.; Olivo, P.; Nowak, E. Experimental Investigation of 4-kb RRAM Arrays Programming Conditions Suitable for TCAM. In *2018 IEEE Transactions on Very Large Scale Integration (VLSI) Systems* **2018**, *26*, 2599–2607.
- (32) Chen, Y.-S.; Lee, H.-Y.; Chen, P.-S.; Liu, W.-H.; Wang, S.-M.; Gu, P.-Y.; Hsu, Y.-Y.; Tsai, C.-H.; Chen, W.-S.; Chen, F.; et al. Robust High-Resistance State and Improved Endurance of HfO_x Resistive Memory by Suppression of Current Overshoot. *IEEE Electron Device Lett.* **2011**, *32*, 1585–1587.
- (33) Cao, R.; Liu, S.; Liu, Q.; Zhao, X.; Wang, W.; Zhang, X.; Wu, F.; Wu, Q.; Wang, Y.; Lv, H.; Long, S.; Liu, M. Improvement of Device Reliability by Introducing a BEOL-Compatible TiN Barrier Layer in CBRAM. *IEEE Electron Device Lett.* **2017**, *38*, 1371–1374.
- (34) Balatti, S.; Ambrogio, S.; Wang, Z.; Sills, S.; Calderoni, A.; Ramaswamy, N.; Ielmini, D. Voltage-Controlled Cycling Endurance of HfO_x-Based Resistive-Switching Memory. *IEEE Trans. Electron Devices* **2015**, *62*, 3365–3372.
- (35) Stathopoulos, S.; Khat, A.; Trapatseli, M.; Cortese, S.; Serb, A.; Valov, I.; Prodromakis, T. Multibit Memory Operation of Metal-Oxide Bi-Layer Memristors. *Sci. Rep.* **2017**, *7*, 17532.
- (36) Chen, S.; Mahmoodi, M. R.; Shi, Y.; Mahata, C.; Yuan, B.; Liang, X.; Wen, C.; Hui, F.; Akinwande, D.; Strukov, D. B.; et al. Wafer-Scale Integration of Two-Dimensional Materials in High-Density Memristive Crossbar Arrays for Artificial Neural Networks. *Nat. Electron.* **2020**, *3*, 638–645.
- (37) Mehonic, A.; Joksas, D.; Ng, W. H.; Buckwell, M.; Kenyon, A. J. Simulation of Inference Accuracy Using Realistic RRAM Devices. *Front. Neurosci.* **2019**, *13*, 593.
- (38) Ielmini, D.; Ambrogio, S. Emerging Neuromorphic Devices. *Nanotechnology* **2020**, *31*, 092001.
- (39) Hwan Kim, G.; Ho Lee, J.; Yeong Seok, J.; Ji Song, S.; Ho Yoon, J.; Jean Yoon, K.; Hwan Lee, M.; Min Kim, K.; Dong Lee, H.; Wook Ryu, S.; et al. Improved Endurance of Resistive Switching TiO₂ Thin Film by Hourglass Shaped Magnéli Filaments. *Appl. Phys. Lett.* **2011**, *98*, 262901.
- (40) Banerjee, W.; Maikap, S.; Lai, C.-S.; Chen, Y.-Y.; Tien, T.-C.; Lee, H.-Y.; Chen, W.-S.; Chen, F. T.; Kao, M.-J.; Tsai, M.-J.; et al. Formation Polarity Dependent Improved Resistive Switching Memory Characteristics Using Nanoscale (1.3 nm) Core-Shell IrO_x Nano-Dots. *Nanoscale Res. Lett.* **2012**, *7*, 194–205.
- (41) Wen, C.; Li, X.; Zanotti, T.; Puglisi, F. M.; Shi, Y.; Saiz, F.; Antidormi, A.; Roche, S.; Zheng, W.; Liang, X.; et al. Advanced Data Encryption Using 2D Materials. *Adv. Mater.* **2021**, *33*, 2100185.
- (42) Li, X.; Zanotti, T.; Wang, T.; Zhu, K.; Puglisi, F. M.; Lanza, M. Random Telegraph Noise in Metal-Oxide Memristors for True Random Number Generators: A Materials Study. *Adv. Funct. Mater.* **2021**, *31*, 2102172.
- (43) Wei, Z.; Katoh, Y.; Ogasahara, S.; Yoshimoto, Y.; Kawai, K.; Ikeda, Y.; Eriguchi, K.; Ohmori, K.; Yoneda, S. True Random Number Generator Using Current Difference Based on a Fractional Stochastic Model in 40-nm Embedded ReRAM. Proceedings from the *International Electron Devices Meeting (IEDM)*, San Francisco, CA, December 3–7, 2016; IEEE: New York, 2016; pp 107–110.
- (44) Palumbo, F.; Wen, C.; Lombardo, S.; Pazos, S.; Aguirre, F.; Eizenberg, M.; Hui, F.; Lanza, M. A Review on Dielectric Breakdown in Thin Dielectrics: Silicon Dioxide, High-k, and Layered Dielectrics. *Adv. Funct. Mater.* **2020**, *30*, 1900657.
- (45) Yang, Y.; Takahashi, Y.; Tsurumaki-Fukuchi, A.; Arita, M.; Moors, M.; Buckwell, M.; Mehonic, A.; Kenyon, A. Probing Electrochemistry at the Nanoscale: In Situ TEM and STM Characterizations of Conducting Filaments in Memristive Devices. *J. Electroceram.* **2017**, *39*, 73–93.
- (46) Roldán, J. B.; González-Cordero, G.; Picos, R.; Miranda, E.; Palumbo, F.; Jiménez-Molinos, F.; Moreno, E.; Maldonado, D.; Balmori, S. B.; Moner Al Chawa, M.; et al. On the Thermal Models for Resistive Random Access Memory Circuit Simulation. *Nanomaterials* **2021**, *11*, 1261.
- (47) Strukov, D. B. Endurance-Write-Speed Tradeoffs in Nonvolatile Memories. *Appl. Phys. A: Mater. Sci. Process.* **2016**, *122*, 302.
- (48) Stathopoulos, S.; Michalas, L.; Khat, A.; Serb, A.; Prodromakis, T. An Electrical Characterisation Methodology for Benchmarking Memristive Device Technologies. *Sci. Rep.* **2019**, *9*, 19412.
- (49) Zhao, M.; Gao, B.; Xi, Y.; Xu, F.; Wu, H.; Qian, H. Endurance and Retention Degradation of Intermediate Levels in Filamentary Analog RRAM. *IEEE J. Electron Devices Soc.* **2019**, *7*, 1239–1247.
- (50) Shi, Y.; Liang, X.; Yuan, B.; Chen, V.; Li, H.; Hui, F.; Yu, Z.; Yuan, F.; Pop, E.; Wong, H.-S. P.; et al. Electronic Synapses Made of Layered Two-Dimensional Materials. *Nat. Electron.* **2018**, *1*, 458–465.
- (51) Cho, H.; Kim, S. Enhancing Short-Term Plasticity by Inserting a Thin TiO₂ Layer in WO_x-Based Resistive Switching Memory. *Coatings* **2020**, *10*, 908.
- (52) Rana, A. M.; Akbar, T.; Ismail, M.; Ahmad, E.; Hussain, F.; Talib, I.; Imran, M.; Mehmood, K.; Iqbal, K.; Nadeem, M. Y. Endurance and Cycle-to-Cycle Uniformity Improvement in Tri-Layered CeO₂/Ti/CeO₂ Resistive Switching Devices by Changing Top Electrode Material. *Sci. Rep.* **2017**, *7*, 39539.
- (53) Ismail, M.; Ahmed, E.; Rana, A.; Hussain, F.; Talib, I.; Nadeem, M.; Panda, D.; Shah, N. Improved Endurance and Resistive Switching Stability in Ceria Thin Films Due to Charge Transfer Ability of Al Dopant. *ACS Appl. Mater. Interfaces* **2016**, *8*, 6127–6136.
- (54) Kim, K. M.; Yang, J. J.; Strachan, J. P.; Grafals, E. M.; Ge, N.; Melendez, N. D.; Li, Z.; Williams, R. S. Voltage Divider Effect for the Improvement of Variability and Endurance of TaO_x Memristor. *Sci. Rep.* **2016**, *6*, 20085.
- (55) Lin, C.-L.; Lin, T.-Y. Superior Unipolar Resistive Switching in Stacked ZrO_x/ZrO₂/ZrO_x Structure. *AIP Adv.* **2016**, *6*, 035103.
- (56) Liu, S.; Lu, N.; Zhao, X.; Xu, H.; Banerjee, W.; Lv, H.; Long, S.; Li, Q.; Liu, Q.; Liu, M. Eliminating Negative-SET Behavior by Suppressing Nanofilament Overgrowth in Cation-Based Memory. *Adv. Mater.* **2016**, *28*, 10623.
- (57) Jana, D.; Samanta, S.; Roy, S.; Lin, Y. F.; Maikap, S. Observation of Resistive Switching Memory by Reducing Device Size in a New Cr/CrO_x/TiO_x/TiN Structure. *Nano-Micro Lett.* **2015**, *7*, 392–399.
- (58) Huber, B.; Popp, P.; Kaiser, M.; Ruediger, A.; Schindler, C. Fully Inkjet Printed Flexible Resistive Memory. *Appl. Phys. Lett.* **2017**, *110*, 143503.
- (59) Lienig, J. Electromigration and Its Impact on Physical Design in Future Technologies. Proceedings of the *2013 ACM International symposium on Physical Design*, Nevada, March 24–27, 2013; ACM: New York, 2013; pp 33–40.
- (60) Kozicki, M. N.; Park, M.; Mitkova, M. Nanoscale Memory Elements Based on Solid-Electrolytes. *IEEE Trans. Nanotechnol.* **2005**, *4*, 331–338.
- (61) Lv, H.; Xu, X.; Liu, H.; Liu, R.; Liu, Q.; Banerjee, W.; Sun, H.; Long, S.; Li, L.; Liu, M. Evolution of Conductive Filament and Its Impact on Reliability Issues in Oxide-Electrolyte Based Resistive Random Access Memory. *Sci. Rep.* **2015**, *5*, 7764.
- (62) Lee, M.-J.; Lee, D.; Cho, S.-H.; Hur, J.-H.; Lee, S.-M.; Seo, D. H.; Kim, D.-S.; Yang, M.-S.; Lee, S.; Hwang, E.; et al. A Plasma-Treated Chalcogenide Switch Device for Stackable Scalable 3D Nanoscale Memory. *Nat. Commun.* **2013**, *4*, 2629.
- (63) Lai, Y. C.; Hsu, F. C.; Chen, J. Y.; He, J. H.; Chang, T. C.; Hsieh, Y. P.; Lin, T. Y.; Yang, Y. J.; Chen, Y. F. Transferable and Flexible Label-Like Macromolecular Memory on Arbitrary Substrates with High Performance and a Facile Methodology. *Adv. Mater.* **2013**, *25*, 2733–2739.
- (64) Seong, D.-J.; Jo, M.; Lee, D.; Hwang, H. HPHA Effect on Reversible Resistive Switching of Pt/Nb-Doped SrTiO₃ Schottky Junction for Nonvolatile Memory Application. *Electrochem. Solid-State Lett.* **2007**, *10*, H168.
- (65) Yoong, H. Y.; Wu, H.; Zhao, J.; Wang, H.; Guo, R.; Xiao, J.; Zhang, B.; Yang, P.; Pennycook, S. J.; Deng, N.; et al. Epitaxial Ferroelectric Hf_{0.3}Zr_{0.5}O₂ Thin Films and Their Implementations in Memristors for Brain-Inspired Computing. *Adv. Funct. Mater.* **2018**, *28*, 1806037.

- (66) Kund, M.; Beitel, G.; Pinnow, C. U.; Röhr, T.; Schumann, J.; Symanczyk, R.; Ufert, K.; Muller, G. Conductive-Bridging RAM (CBRAM): An Emerging Non-Volatile Memory Technology Scalable to Sub 20 nm. Proceedings from the *International Electron Devices Meeting (IEDM)*, Washington, DC, December 5, 2005; IEEE: New York, 2005; pp 754–757.
- (67) Chand, U.; Huang, C.-Y.; Jieng, J.-H.; Jang, W.-Y.; Lin, C.-H.; Tseng, T.-Y. Suppression of Endurance Degradation by Utilizing Oxygen Plasma Treatment in HfO₂ Resistive Switching Memory. *Appl. Phys. Lett.* **2015**, *106*, 153502.
- (68) Zhang, R.; Chang, K.; Chang, T.; Tsai, T.; Huang, S.; Chen, W.; Chen, K.; Lou, J.; Chen, J.; Young, T.; Chen, M.; Chen, H.; Liang, S.; Syu, Y.; Sze, S. M. Characterization of Oxygen Accumulation in Indium-Tin-Oxide for Resistance Random Access Memory. *IEEE Electron Device Lett.* **2014**, *35*, 630–632.
- (69) Zhang, B.; Liu, G.; Chen, Y.; Zeng, L. J.; Zhu, C. X.; Neoh, K. G.; Wang, C.; Kang, E. T. Conjugated Polymer-Grafted Reduced Graphene Oxide for Nonvolatile Rewritable Memory. *Chem. - Eur. J.* **2011**, *17*, 13646–13652.
- (70) Liu, G.; Zhuang, X.; Chen, Y.; Zhang, B.; Zhu, J.; Zhu, C.-X.; Neoh, K.-G.; Kang, E.-T. Bistable Electrical Switching and Electronic Memory Effect in a Solution-Processable Graphene Oxide-Donor Polymer Complex. *Appl. Phys. Lett.* **2009**, *95*, 253301.
- (71) Zhuang, X. D.; Chen, Y.; Liu, G.; Li, P. P.; Zhu, C. X.; Kang, E. T.; Noeh, K. G.; Zhang, B.; Zhu, J. H.; Li, Y. X. Conjugated-Polymer-Functionalized Graphene Oxide: Synthesis and Nonvolatile Rewritable Memory Effect. *Adv. Mater.* **2010**, *22*, 1731–1735.
- (72) Kim, H.-D.; An, H.-M.; Lee, E. B.; Kim, T. G. Stable Bipolar Resistive Switching Characteristics and Resistive Switching Mechanisms Observed in Aluminum Nitride-Based ReRAM Devices. *IEEE Trans. Electron Devices* **2011**, *58*, 3566–3573.
- (73) Chen, Y.-J.; Chen, H.-L.; Young, T.-F.; Chang, T.-C.; Tsai, T.-M.; Chang, K.-C.; Zhang, R.; Chen, K.-H.; Lou, J.-C.; Chu, T.-J.; et al. Hydrogen Induced Redox Mechanism in Amorphous Carbon Resistive Random Access Memory. *Nanoscale Res. Lett.* **2014**, *9*, 52.
- (74) Kim, H.-D.; An, H.-M.; Seo, Y.; Kim, T. G. Transparent Resistive Switching Memory Using ITO/AlN/ITO Capacitors. *IEEE Electron Device Lett.* **2011**, *32*, 1125–1127.
- (75) Zhao, X.; Liu, S.; Niu, J.; Liao, L.; Liu, Q.; Xiao, X.; Lv, H.; Long, S.; Banerjee, W.; Li, W.; et al. Confining Cation Injection to Enhance CBRAM Performance by Nanopore Graphene layer. *Small* **2017**, *13*, 1603948.
- (76) Schindler, C.; Therman, S. C. P.; Waser, R.; Kozicki, M. N. Bipolar and Unipolar Resistive Switching in Cu-Doped SiO₂. *IEEE Trans. Electron Devices* **2007**, *54*, 2762.
- (77) Yu, S.; Chen, H.-Y.; Gao, B.; Kang, J.; Wong, H.-S. P. HfO_x-Based Vertical Resistive Switching Random Access Memory Suitable for Bit-Cost-Effective Three-Dimensional Cross-Point Architecture. *ACS Nano* **2013**, *7*, 2320–2325.
- (78) Lee, J.; Shin, J.; Lee, D.; Lee, W.; Jung, S.; Jo, M.; Park, J.; Biju, K. P.; Kim, S.; Park, S. Diode-Less Nano-Scale ZrO_x/HfO_x RRAM Device with Excellent Switching Uniformity and Reliability for High-Density Cross-Point Memory Applications. Proceedings from the *International Electron Devices Meeting (IEDM)*, San Francisco, CA, December 6–8, 2010; IEEE: New York, 2010; pp 452–455.
- (79) Lee, J.; Bourim, E. M.; Lee, W.; Park, J.; Jo, M.; Jung, S.; Shin, J.; Hwang, H. Effect of ZrO_x/HfO_x Bilayer Structure on Switching Uniformity and Reliability in Nonvolatile Memory Applications. *Appl. Phys. Lett.* **2010**, *97*, 172105.
- (80) Prakash, A.; Maikap, S.; Rahaman, S. Z.; Majumdar, S.; Manna, S.; Ray, S.-K. Resistive Switching Memory Characteristics of Ge/GeO_x Nanowires and Evidence of Oxygen Ion Migration. *Nanoscale Res. Lett.* **2013**, *8*, 220.
- (81) Tran, X.-A.; Yu, H.-Y.; Yeo, Y.-C.; Wu, L.; Liu, W.-J.; Wang, Z.-R.; Fang, Z.; Pey, K.-L.; Sun, X.-W.; Du, A.-Y.; Nguyen, B.-Y.; Li, M.-F. A High-Yield HfO_x-Based Unipolar Resistive RAM Employing Ni Electrode Compatible with Si-Diode Selector for Crossbar Integration. *IEEE Electron Device Lett.* **2011**, *32*, 396–398.
- (82) Cheng, C. H.; Yeh, F. S.; Chin, A. Low-Power High-Performance Non-Volatile Memory on a Flexible Substrate with Excellent Endurance. *Adv. Mater.* **2011**, *23*, 902–905.
- (83) Choi, D.; Lee, D.; Sim, H.; Chang, M.; Hwang, H. Reversible Resistive Switching of SrTiO_x Thin Films for Nonvolatile Memory Applications. *Appl. Phys. Lett.* **2006**, *88*, 082904.
- (84) Valanarasu, S.; Kulandaisamy, I.; Kathalingam, A.; Rhee, J.-K.; Vijayan, T.; Chandramohan, R. High-Performance Memory Device Using Graphene Oxide Flakes Sandwiched Polymethylmethacrylate Layers. *J. Nanosci. Nanotechnol.* **2013**, *13*, 6755–6759.
- (85) Kim, S.; Yarimaga, O.; Choi, S. J.; Choi, Y. K. Highly Durable and Flexible Memory Based on Resistance Switching. *Solid-State Electron.* **2010**, *54*, 392–396.
- (86) Jang, B. C.; Seong, H.; Kim, S. K.; Kim, J. Y.; Koo, B. J.; Choi, J.; Yang, S. Y.; Im, S. G.; Choi, S. Flexible Nonvolatile Polymer Memory Array on Plastic Substrate via Initiated Chemical Vapor Deposition. *ACS Appl. Mater. Interfaces* **2016**, *8*, 12951.
- (87) Gonzalez, M. B.; Martin-Martinez, J.; Rodriguez, R.; Acero, M. C.; Nafria, M.; Campabadal, F.; Aymerich, X. Dedicated Random Telegraph Noise Characterization of Ni/HfO₂-Based RRAM Devices. *Microelectron. Eng.* **2015**, *147*, 59–62.
- (88) Berdan, R.; Serb, A.; Khiat, A.; Regoutz, A.; Papavassiliou, C.; Prodromakis, T. A μ -Controller-Based System for Interfacing Selector-Less RRAM Crossbar Arrays. *IEEE Trans. Electron Devices* **2015**, *62*, 2190–2196.
- (89) Hennen, T.; Wichmann, E.; Elias, A.; Lille, J.; Mosendz, O.; Waser, R.; Wouters, D. J.; Bedau, D. Current-Limiting Amplifier for High Speed Measurement of Resistive Switching Data. *Rev. Sci. Instrum.* **2021**, *92*, 054701.
- (90) Jo, S. H.; Kumar, T.; Zitlaw, C.; Nazarian, H. Self-Limited RRAM with ON/OFF Resistance Ratio Amplification. Proceedings from the *Symposium on VLSI Technology (VLSI Technology)*, Kyoto, Japan, June 16–18, 2015; IEEE: New York, 2015; pp 128–129.
- (91) Yang, J. J.; Zhang, M.-X.; Strachan, J. P.; Miao, F.; Pickett, M. D.; Kelley, R. D.; Medeiros-Ribeiro, G.; Williams, R. S. High Switching Endurance in TaO_x Memristive Devices. *Appl. Phys. Lett.* **2010**, *97*, 232102.
- (92) Xu, X.; Tai, L.; Gong, T.; Yin, J.; Huang, P.; Yu, J.; Luo, Q.; Liu, J.; Yu, Z.; Zhu, X. 40 × Retention Improvement by Eliminating Resistance Relaxation with High Temperature Forming in 28 nm RRAM Chip. Proceedings from the *International Electron Devices Meeting (IEDM)*, San Francisco, CA, December 1–5, 2018; IEEE: New York, 2018; pp 464–467.
- (93) Chen, C. Y.; Fantini, A.; Degraeve, R.; Redolfi, A.; Groeseneken, G.; Goux, L.; Kar, G. S. Statistical Investigation of the Impact of Program History and Oxide-Metal Interface on OxRRAM Retention. Proceedings from the *IEEE International Electron Devices Meet. (IEDM)*, San Francisco, CA, December 3–7, 2016; IEEE: New York, 2016; pp 99–102.
- (94) Kinoshita, K.; Tsunoda, K.; Sato, Y.; Noshiro, H.; Yagaki, S.; Aoki, M.; Sugiyama, Y. Reduction in the Reset Current in a Resistive Random Access Memory Consisting of NiOx Brought About by Reducing a Parasitic Capacitance. *Appl. Phys. Lett.* **2008**, *93*, 033506.
- (95) Sato, Y.; Tsunoda, K.; Kinoshita, K.; Noshiro, H.; Aoki, M.; Sugiyama, Y. Sub-100- μ A Reset Current of Nickel Oxide Resistive Memory Through Control of Filamentary Conductance by Current Limit of MOSFET. *IEEE Trans. Electron Devices* **2008**, *55*, 1185–1191.
- (96) Nardi, F.; Ielmini, D.; Cagli, C.; Spiga, S.; Fanciulli, M.; Goux, L.; Wouters, D. J. Control of Filament Size and Reduction of Reset Current below 10 μ A in NiO Resistance Switching Memories. *Solid-State Electron.* **2011**, *58*, 42–47.
- (97) Lee, H.; Chen, P.; Wu, T.; Chen, Y.; Wang, C.; Tzeng, P.; Lin, C.; Chen, F.; Lien, C.; Tsai, M.-J. Low Power and High Speed Bipolar Switching with a Thin Reactive Ti Buffer Layer in Robust HfO₂ Based RRAM. Proceedings from the *IEEE International Electron Devices Meet. (IEDM)*, San Francisco, CA, December 15–17, 2008; IEEE: New York, 2008.

- 1172 (98) Wiefels, S.; Von Witzleben, M.; Huttemann, M.; Bottger, U.;
1173 Waser, R.; Menzel, S. Impact of the Ohmic Electrode on the
1174 Endurance of Oxide Based Resistive Switching Memory. *IEEE Trans.*
1175 *Electron Devices* **2021**, *68*, 1024–1030.
- 1176 (99) Montesi, L.; Buckwell, M.; Zarudnyi, K.; Garnett, L.; Hudziak,
1177 S.; Mehonic, A.; Kenyon, A. J. Nanosecond Analog Programming of
1178 Substoichiometric Silicon Oxide Resistive RAM. *IEEE Trans.*
1179 *Nanotechnol.* **2016**, *15*, 428–434.
- 1180 (100) Nikam, R. D.; Rajput, K. G.; Hwang, H. Single-Atom
1181 Quantum-Point Contact Switch Using Atomically Thin Hexagonal
1182 Boron Nitride. *Small* **2021**, *17*, 2006760.
- 1183 (101) Zhao, X.; Ma, J.; Xiao, X.; Liu, Q.; Shao, L.; Chen, D.; Liu, S.;
1184 Niu, J.; Zhang, X.; Wang, Y.; et al. Breaking the Current-Retention
1185 Dilemma in Cation-Based Resistive Switching Devices Utilizing
1186 Graphene with Controlled Defects. *Adv. Mater.* **2018**, *30*, 1705193.
- 1187 (102) Wu, Z.; Zhao, X.; Yang, Y.; Wang, W.; Zhang, X.; Wang, R.;
1188 Cao, R.; Liu, Q.; Banerjee, W. Transformation of Threshold Volatile
1189 Switching to Quantum Point Contact Originated Nonvolatile
1190 Switching in Graphene Interface Controlled Memory Devices.
1191 *Nanoscale Adv.* **2019**, *1*, 3753–3760.
- 1192 (103) Chen, Y.-S.; Wu, T.-Y.; Tzeng, P.-J.; Chen, P.-S.; Lee, H.-Y.;
1193 Lin, C.-H.; Chen, F.; Tsai, M.-J. Forming-Free HfO₂ Bipolar RRAM
1194 Device with Improved Endurance and High Speed Operation.
1195 Proceedings from the 2009 International Symposium on VLSI
1196 Technology, Systems, and Applications, Hsinchu, Taiwan, April 27–
1197 29, 2009; IEEE: New York, 2009; pp 37–38.
- 1198 (104) Golonzka, O.; Arslan, U.; Bai, P.; Bohr, M.; Baykan, O.;
1199 Chang, Y.; Chaudhari, A.; Chen, A.; Clarke, J.; Connor, C.; Das, N.;
1200 English, C.; Ghani, T.; Hamzaoglu, F.; Hentges, P.; Jain, P.; Jezewski,
1201 C.; Karpov, I.; Kothari, H.; Kotlyar, R.; Lin, B.; Metz, M.; Odonnell,
1202 J.; Ouellette, D.; Park, J.; Pirkle, A.; Quintero, P.; Seghete, D.; Sekhar,
1203 M.; Sen Gupta, A.; Seth, M.; Strutt, N.; Wiegand, C.; Yoo, H. J.;
1204 Fischer, K. Non-Volatile RRAM Embedded into 22FFL FinFET
1205 Technology. Proceedings from the 2019 Symposium on VLSI
1206 Technology Digest of Technical Papers, Kyoto, Japan, June 9–14,
1207 2019; IEEE: New York, 2019; pp 230–231.
- 1208 (105) Chou, C.; Lin, Z.; Lai, C.; Su, C.; Tseng, P.; Chen, W.; Tsai,
1209 W.; Chu, W.; Ong, T.; Chuang, H.; Chih, Y.; Chang, T. J. A 22nm
1210 96KX144 RRAM Macro with a Self-Tracking Reference and a Low
1211 Ripple Charge Pump to Achieve a Configurable Read Window and a
1212 Wide Operating Voltage Range. Proceedings from the 2020 IEEE
1213 Symposium on VLSI Circuits, Honolulu, HI, June 16–19, 2020; IEEE:
1214 New York, 2020.
- 1215 (106) Illarionov, Y. Y.; Bانشchikov, A. G.; Polyushkin, D. K.;
1216 Wachter, S.; Knobloch, T.; Thesberg, M.; Mennel, L.; Paur, M.;
1217 Stöger-Pollach, M.; Steiger-Thirsfeld, A.; Vexler, M. I.; Walzl, M.;
1218 Sokolov, N. S.; Mueller, T.; Grasser, T. A Ultrathin Calcium Fluoride
1219 Insulators for Two-Dimensional Field-Effect Transistors. *Nat.*
1220 *Electron.* **2019**, *2*, 230–235.
- 1221 (107) Smithe, K. K.; Suryavanshi, S. V.; Muñoz Rojo, M.; Tedjarati,
1222 A. D.; Pop, E. Low Variability in Synthetic Monolayer MoS₂ Devices.
1223 *ACS Nano* **2017**, *11*, 8456–8463.
- 1224 (108) Lin, Z.; Liu, Y.; Halim, U.; Ding, M.; Liu, Y.; Wang, Y.; Jia, C.;
1225 Chen, P.; Duan, X.; Wang, C.; et al. Solution-Processable 2D
1226 Semiconductors for High-Performance Large-Area Electronics. *Nature*
1227 **2018**, *562*, 254–258.
- 1228 (109) Shi, Y.; Groven, B.; Serron, J.; Wu, X.; Nalin Mehta, A.; Minj,
1229 A.; Sergeant, S.; Han, H.; Asselberghs, I.; Lin, D.; et al. Engineering
1230 Wafer-Scale Epitaxial Two-Dimensional Materials through Sapphire
1231 Template Screening for Advanced High-Performance Nanoelectronics.
1232 *ACS Nano* **2021**, *15*, 9482–9494.
- 1233 (110) Nature website, author guidelines. <https://www.nature.com/nature/for-authors/initial-submission> (accessed 2021-06-11).
- 1234 (111) Chen, C. Y.; Goux, L.; Fantini, A.; Degraeve, R.; Redolfi, A.;
1235 Groeseneken, G.; Jurczak, M. Stack Optimization of Oxide-Based
1236 RRAM for Fast Write Speed (<1 fs) at Low Operating Current (<10
1237 μA). *Solid-State Electron.* **2016**, *125*, 198–203.
- 1238 (112) Rahaman, S. Z.; Maikap, S.; Tien, T.-C.; Lee, H.-Y.; Chen, W.-
1239 S.; Chen, F. T.; Kao, M.-J.; Tsai, M.-J. Excellent Resistive Memory
1240 Characteristics and Switching Mechanism Using a Ti Nanolayer at the
1241 Cu/TaO_x Interface. *Nanoscale Res. Lett.* **2012**, *7*, 345–355.
- 1242 (113) Fang, R.-C.; Sun, Q.-Q.; Zhou, P.; Yang, W.; Wang, P.-F.;
1243 Zhang, D. W. High-Performance Bilayer Flexible Resistive Random
1244 Access Memory Based on Low-Temperature Thermal Atomic Layer
1245 Deposition. *Nanoscale Res. Lett.* **2013**, *8*, 92.
- 1246 (114) Cheng, P.; Sun, K.; Hu, Y. H. Memristive Behavior and Ideal
1247 Memristor of 1T Phase MoS₂ Nanosheets. *Nano Lett.* **2016**, *16*, 572–
1248 576.
- 1249 (115) Fantini, A.; Goux, L.; Redolfi, A.; Degraeve, R.; Kar, G.; Chen,
1250 Y. Y.; Jurczak, M. Lateral and Vertical Scaling Impact on Statistical
1251 Performances and Reliability of 10 nm TiN/Hf(Al)O/Hf/TiN
1252 RRAM Devices. Proceedings from the 2014 Symposium on VLSI
1253 Technology (VLSI-Technology): Digest of Technical Papers, Honolulu,
1254 HI, June 9–12, 2014; IEEE: New York, 2014.
- 1255 (116) Strukov, D. B.; Snider, G. S.; Stewart, D. R.; Williams, R. S.
1256 The Missing Memristor Found. *Nature* **2008**, *453*, 80–83.
- 1257 (117) Choi, B. J.; Torrezan, A. C.; Norris, K. J.; Miao, F.; Strachan, J.
1258 P.; Zhang, M.-X.; Ohlberg, D. A.; Kobayashi, N. P.; Yang, J. J.;
1259 Williams, R. S. Electrical Performance and Scalability of Pt Dispersed
1260 SiO₂ Nanometallic Resistance Switch. *Nano Lett.* **2013**, *13*, 3213–
1261 3217.
- 1262 (118) Quesada, E. P.; ío Romero-Zalaz, R.; Pérez, E.; Mahadevaiah,
1263 M. K.; Reuben, J.; Schubert, M. A.; Jiménez-Molinos, F.; Roldán, J. B.;
1264 Wenger, C. Toward Reliable Compact Modeling of Multilevel 1T-1R
1265 RRAM Devices for Neuromorphic Systems. *Electronics* **2021**, *10*, 645.
- 1266 (119) Wang, M.; Cai, S.; Pan, C.; Wang, C.; Lian, X.; Zhuo, Y.; Xu,
1267 K.; Cao, T.; Pan, X.; Wang, B.; et al. Robust Memristors Based on
1268 Layered Two-Dimensional Materials. *Nat. Electron.* **2018**, *1*, 130–136.
- 1269 (120) Kim, Y.-B.; Lee, S. R.; Lee, D.; Lee, C. B.; Chang, M.; Hur, J.
1270 H.; Lee, M.-J.; Park, G.-S.; Kim, C. J.; Chung, U.-I. Bi-Layered RRAM
1271 with Unlimited Rndurance and Extremely Uniform Switching.
1272 Proceedings from the 2011 Symposium on VLSI Technology-Digest of
1273 Technical Papers, Kyoto, Japan, June 14–16, 2011; IEEE: New York,
1274 2011; pp 52–53.
- 1275 (121) Seong, D.-J.; Park, J.; Lee, N.; Hasan, M.; Jung, S.; Choi, H.;
1276 Lee, J.; Jo, M.; Lee, W.; Park, S. Effect of Oxygen Migration and
1277 Interface Engineering on Resistance Switching Behavior of Reactive
1278 Metal/Polycrystalline Pr_{0.7}Ca_{0.3}MnO₃ Device for Nonvolatile Memory
1279 Applications. *International Electron Devices Meeting (IEDM)*;
1280 IEEE: Baltimore, MD, USA, 7–9 Dec 2009; pp 101–104.
- 1281 (122) Lin, Y.-Y.; Lee, F.-M.; Chen, Y.-C.; Chien, W.-C.; Yeh, C.-W.;
1282 Hsieh, K.-Y.; Lu, C.-Y. A Novel Tite Buffered Cu-GeSbTe/SiO₂
1283 Electrochemical Resistive Memory (ReRAM). Proceedings from the
1284 2010 Symposium on VLSI Technology, Honolulu, HI, June 15–17,
1285 2010; IEEE: New York, 2010; pp 91–92.
- 1286 (123) Lee, M.-J.; Lee, C. B.; Lee, D.; Lee, S. R.; Chang, M.; Hur, J.
1287 H.; Kim, Y.-B.; Kim, C.-J.; Seo, D. H.; Seo, S.; et al. A Fast, High-
1288 Endurance and Scalable Non-Volatile Memory Device Made from
1289 Asymmetric Ta₂O_{5-x}/TaO_{2-x} Bilayer Structures. *Nat. Mater.* **2011**, *10*,
1290 625–630.
- 1291 (124) Yu, S.; Wu, Y.; Jeyasingh, R.; Kuzum, D.; Wong, H.-S. P. An
1292 Electronic Synapse Device Based on Metal Oxide Resistive Switching
1293 Memory for Neuromorphic cComputation. *IEEE Trans. Electron*
1294 *Devices* **2011**, *58*, 2729–2737.
- 1295 (125) Yoshida, C.; Tsunoda, K.; Noshiro, H.; Sugiyama, Y. High
1296 Speed Resistive Switching in Pt/TiO₂/TiN Film for Nonvolatile
1297 Memory Application. *Appl. Phys. Lett.* **2007**, *91*, 223510.
- 1298 (126) Nagashima, K.; Yanagida, T.; Oka, K.; Taniguchi, M.; Kawai,
1299 T.; Kim, J.-S.; Park, B. H. Resistive Switching Multistate Nonvolatile
1300 Memory Effects in a Single Cobalt Oxide Nanowire. *Nano Lett.* **2010**,
1301 *10*, 1359–1363.
- 1302 (127) Yan, Z.; Guo, Y.; Zhang, G.; Liu, J. M. High-Performance
1303 Programmable Memory Devices Based on Co-Doped BaTiO₃. *Adv.*
1304 *Mater.* **2011**, *23*, 1351–1355.
- 1305 (128) Chen, Y. Y.; Goux, L.; Clima, S.; Govoreanu, B.; Degraeve, R.;
1306 Kar, G. S.; Fantini, A.; Groeseneken, G.; Wouters, D. J.; Jurczak, M.
1307 Endurance/Retention Trade-Off on HfO₂/Metal Cap 1T1R Bipolar
1308 RRAM. *IEEE Trans. Electron Devices* **2013**, *60*, 1114–1121.
- 1309

- (129) Lee, H.; Chen, Y.; Chen, P.; Wu, T.; Chen, F.; Wang, C.; Tzeng, P.; Tsai, M.-J.; Lien, C. Low-Power and Nanosecond Switching in Robust Hafnium Oxide Resistive Memory with a Thin Ti Cap. *IEEE Electron Device Lett.* **2010**, *31*, 44–46.
- (130) Tsurumaki, A.; Yamada, H.; Sawa, A. Impact of Bi Deficiencies on Ferroelectric Resistive Switching Characteristics Observed at p-Type Schottky-Like Pt/Bi_{1-x}FeO₃ Interfaces. *Adv. Funct. Mater.* **2012**, *22*, 1040–1047.
- (131) Wei, Z.; Kanzawa, Y.; Arita, K.; Katoh, Y.; Kawai, K.; Muraoka, S.; Mitani, S.; Fujii, S.; Katayama, K.; Iijima, M. Highly Reliable TaO_x ReRAM and Direct Evidence of Redox Reaction Mechanism. Proceedings from the *International Electron Devices Meeting (IEDM)*, San Francisco, CA, December 15–17, 2008; IEEE: New York, 2008.
- (132) Terai, M.; Sakotsubo, Y.; Saito, Y.; Kotsuji, S.; Hada, H. Effect of Bottom Electrode of ReRAM with Ta₂O₅/TiO₂ Stack on RTN and Retention. Proceedings from the *International Electron Devices Meeting (IEDM)*, Baltimore, MD, December 7–9, 2009; IEEE: New York, 2009; pp 775–778.
- (133) Chien, W.; Chen, Y.; Chen, Y.; Chuang, A. T.; Lee, F.; Lin, Y.; Lai, E.; Shih, Y.; Hsieh, K.; Lu, C.-Y. A Forming-Free WO_x Resistive Memory Using a Novel Self-Aligned Field Enhancement Feature with Excellent Reliability and Scalability. Proceedings from the *International Electron Devices Meeting (IEDM)*, San Francisco, CA, December 6–8, 2010; IEEE: USA, 2010; pp 440–443.
- (134) Shang, J.; Liu, G.; Yang, H.; Zhu, X.; Chen, X.; Tan, H.; Hu, B.; Pan, L.; Xue, W.; Li, R. W. Thermally Stable Transparent Resistive Random Access Memory Based on All-Oxide Heterostructures. *Adv. Funct. Mater.* **2014**, *24*, 2171–2179.
- (135) Garbin, D.; Vianello, E.; Bichler, O.; Raffay, Q.; Gamrat, C.; Ghibauda, G.; DeSalvo, B.; Perniola, L. HfO₂-Based OxRAM Devices as Synapses for Convolutional Neural Networks. *IEEE Trans. Electron Devices* **2015**, *62*, 2494–2501.
- (136) Goswami, S.; Matula, A. J.; Rath, S. P.; Hedström, S.; Saha, S.; Annamalai, M.; Sengupta, D.; Patra, A.; Ghosh, S.; Jani, H.; et al. Robust Resistive Memory Devices Using Solution-Processable Metal-Coordinated Azo Aromatics. *Nat. Mater.* **2017**, *16*, 1216–1224.
- (137) Baek, I.; Lee, M.; Seo, S.; Lee, M.; Seo, D.; Suh, D.-S.; Park, J.; Park, S.; Kim, H.; Yoo, I. Highly Scalable Nonvolatile Resistive Memory Using Simple Binary Oxide Driven by Asymmetric Unipolar Voltage Pulses. Proceedings from the *International Electron Devices Meeting (IEDM)*, San Francisco, CA, December 13–15, 2004; IEEE: New York, 2004; pp 587–590.
- (138) Son, D. I.; Kim, T. W.; Shim, J. H.; Jung, J. H.; Lee, D. U.; Lee, J. M.; Park, W. I.; Choi, W. K. Flexible Organic Bistable Devices Based on Graphene Embedded in an Insulating Poly (methyl Methacrylate) Polymer Layer. *Nano Lett.* **2010**, *10*, 2441–2447.
- (139) Yu, A.-D.; Liu, C.-L.; Chen, W.-C. Supramolecular Block Copolymers: Graphene Oxide Composites for Memory Device Applications. *Chem. Commun.* **2012**, *48*, 383–385.
- (140) Chen, Y. Y.; Govoreanu, B.; Goux, L.; Degraeve, R.; Fantini, A.; Kar, G. S.; Wouters, D. J.; Groeseneken, G.; Kittl, J. A.; Jurczak, M.; et al. Balancing SET/RESET Pulse for 10¹⁰ Endurance in HfO₂/Hf 1T1R Bipolar RRAM. *IEEE Trans. Electron Devices* **2012**, *59*, 3243–3249.
- (141) Oligschlaeger, R.; Waser, R.; Meyer, R.; Karthäuser, S.; Dittmann, R. Resistive Switching and Data Reliability of Epitaxial (Ba, Sr) TiO₃ Thin Films. *Appl. Phys. Lett.* **2006**, *88* (4), 042901.
- (142) Chen, Y.-S.; Lee, H.-Y.; Chen, P.-S.; Wu, T.-Y.; Wang, C.-C.; Tzeng, P.-J.; Chen, F.; Tsai, M.-J.; Lien, C. An Ultrathin Forming-Free HfO_x Resistance Memory with Excellent Electrical Performance. *IEEE Electron Device Lett.* **2010**, *31*, 1473–1475.
- (143) Min Son, J.; Seung Song, W.; Ho Yoo, C.; Yeol Yun, D.; Whan Kim, T. Electrical Stabilities and Memory Mechanisms of Organic Bistable Devices Fabricated Utilizing a Poly (3, 4-Ethylene-Dioxythiophene): Poly (styrenesulfonate) Layer with a Poly (methyl Methacrylate) Buffer Layer. *Appl. Phys. Lett.* **2012**, *100*, 183303.
- (144) Chien, W.; Chen, Y.; Lai, E.; Yao, Y.; Lin, P.; Horng, S.; Gong, J.; Chou, T.; Lin, H.; Chang, M.; et al. Unipolar Switching Behaviors of RTO WO_x RRAM. *IEEE Electron Device Lett.* **2010**, *31*, 126–128.
- (145) Jiang, H.; Han, L.; Lin, P.; Wang, Z.; Jang, M. H.; Wu, Q.; Barnell, M.; Yang, J. J.; Xin, H. L.; Xia, Q. Sub-10 nm Ta Channel Responsible for Superior Performance of a HfO₂ Memristor. *Sci. Rep.* **2016**, *6*, 28525.
- (146) You, T.; Shuai, Y.; Luo, W.; Du, N.; Bürger, D.; Skorupa, I.; Hübner, R.; Henker, S.; Mayr, C.; Schüffny, R.; et al. Exploiting Memristive BiFeO₃ Bilayer Structures for Compact Sequential Logics. *Adv. Funct. Mater.* **2014**, *24*, 3357–3365.
- (147) Boyn, S.; Girod, S.; Garcia, V.; Fusil, S.; Xavier, S.; Deranlot, C.; Yamada, H.; Carrétéro, C.; Jacquet, E.; Bibes, M.; et al. High-Performance Ferroelectric Memory Based on Fully Patterned Tunnel Junctions. *Appl. Phys. Lett.* **2014**, *104*, 052909.
- (148) Lee, M.-J.; Lee, D.; Kim, H.; Choi, H.-S.; Park, J.-B.; Kim, H. G.; Cha, Y.-K.; Chung, U.-I.; Yoo, I.-K.; Kim, K. Highly-Scalable Threshold Switching Select Device Based on Chalcogenide Glasses for 3D Nanoscaled Memory Arrays. Proceedings from the *International Electron Devices Meeting (IEDM)*, San Francisco, CA, December 10–13, 2012; IEEE: New York, 2012; pp 33–35.
- (149) Bai, Y.; Wu, H.; Wu, R.; Zhang, Y.; Deng, N.; Yu, Z.; Qian, H. Study of Multi-Level Characteristics for 3D Vertical Resistive Switching Memory. *Sci. Rep.* **2015**, *4*, 5780.
- (150) Lee, H.; Chen, Y.; Chen, P.; Gu, P.; Hsu, Y.; Wang, S.; Liu, W.; Tsai, C.; Sheu, S.; Chiang, P. Evidence and Solution of Over-RESET Problem for HfO_x Based Resistive Memory with Sub-nm Switching Speed and High Endurance. Proceedings from the *International Electron Devices Meeting (IEDM)*, San Francisco, CA, December 6–8, 2010; IEEE: New York, 2010; pp 460–463.
- (151) Merced-Grafals, E. J.; Dávila, N.; Ge, N.; Williams, R. S.; Strachan, J. P. Repeatable, Accurate, and High Speed Multi-Level Programming of Memristor 1T1R Arrays for Power Efficient Analog Computing Applications. *Nanotechnology* **2016**, *27*, 365202.
- (152) Huang, C.-Y.; Huang, C.-Y.; Tsai, T.-L.; Lin, C.-A.; Tseng, T.-Y. Switching Mechanism of Double Forming Process Phenomenon in ZrO_x/HfO_y Bilayer Resistive Switching Memory Structure with Large Endurance. *Appl. Phys. Lett.* **2014**, *104*, 062901.
- (153) Chiu, F.-C.; Li, P.-W.; Chang, W.-Y. Reliability Characteristics and Conduction Mechanisms in Resistive Switching Memory Devices Using ZnO Thin Films. *Nanoscale Res. Lett.* **2012**, *7*, 178.
- (154) Chang, K.-C.; Tsai, T.-M.; Chang, T.-C.; Wu, H.-H.; Chen, J.-H.; Syu, Y.-E.; Chang, G.-W.; Chu, T.-J.; Liu, G.-R.; Su, Y.-T.; et al. Characteristics and Mechanisms of Silicon-Oxide-Based Resistance Random Access Memory. *IEEE Electron Device Lett.* **2013**, *34*, 399–401.
- (155) Jo, S. H.; Kumar, T.; Narayanan, S.; Nazarian, H. Cross-Point Resistive RAM Based on Field-Assisted Superlinear Threshold Selector. *IEEE Trans. Electron Devices* **2015**, *62*, 3477–3481.
- (156) Santini, C. A.; Sebastian, A.; Marchiori, C.; Jonnalagadda, V. P.; Dellmann, L.; Koelmans, W. W.; Rossell, M. D.; Rossel, C. P.; Eleftheriou, E. Oxygenated Amorphous Carbon for Resistive Memory Applications. *Nat. Commun.* **2015**, *6*, 8600.
- (157) Wang, G.; Yang, Y.; Lee, J.-H.; Abramova, V.; Fei, H.; Ruan, G.; Thomas, E. L.; Tour, J. M. Nanoporous Silicon Oxide Memory. *Nano Lett.* **2014**, *14*, 4694–4699.
- (158) Sangwan, V. K.; Jariwala, D.; Kim, I. S.; Chen, K.-S.; Marks, T. J.; Lauhon, L. J.; Hersam, M. C. Gate-Tunable Memristive Phenomena Mediated by Grain Boundaries in Single-Layer MoS₂. *Nat. Nanotechnol.* **2015**, *10*, 403–406.
- (159) Zhu, X.; Li, D.; Liang, X.; Lu, W. D. Ionic Modulation and Ionic Coupling Effects in MoS₂ Devices for Neuromorphic Computing. *Nat. Mater.* **2019**, *18*, 141–148.
- (160) Ge, R.; Wu, X.; Kim, M.; Shi, J.; Sonde, S.; Tao, L.; Zhang, Y.; Lee, J. C.; Akinwande, D. Atomristor: Nonvolatile Resistance Switching in Atomic Sheets of Transition Metal Dichalcogenides. *Nano Lett.* **2018**, *18*, 434–441.
- (161) Yang, Y.; Du, H.; Xue, Q.; Wei, X.; Yang, Z.; Xu, C.; Lin, D.; Jie, W.; Hao, J. Three-Terminal Memtransistors Based on Two-

- 1446 Dimensional Layered Gallium Selenide Nanosheets for Potential
1447 Low-Power Electronics Applications. *Nano Energy* **2019**, *57*, 566–
1448 573.
- 1449 (162) Li, D.; Wu, B.; Zhu, X.; Wang, J.; Ryu, B.; Lu, W. D.; Lu, W.;
1450 Liang, X. MoS₂ Memristors Exhibiting Variable Switching Character-
1451 istics toward Biorealistic Synaptic Emulation. *ACS Nano* **2018**, *12*,
1452 9240–9252.
- 1453 (163) Xue, F.; He, X.; Retamal, J. R. D.; Han, A.; Zhang, J.; Liu, Z.;
1454 Huang, J. K.; Hu, W.; Tung, V.; He, J. H.; et al. Gate-Tunable and
1455 Multidirection-Switchable Memristive Phenomena in a van der Waals
1456 Ferroelectric. *Adv. Mater.* **2019**, *31*, 1901300.
- 1457 (164) Xu, R.; Jang, H.; Lee, M.-H.; Amanov, D.; Cho, Y.; Kim, H.;
1458 Park, S.; Shin, H.-J.; Ham, D. Vertical MoS₂ Double-Layer Memristor
1459 with Electrochemical Metallization as an Atomic-Scale Synapse with
1460 Switching Thresholds Approaching 100 mV. *Nano Lett.* **2019**, *19*,
1461 2411–2417.
- 1462 (165) Wu, X.; Ge, R.; Chen, P. A.; Chou, H.; Zhang, Z.; Zhang, Y.;
1463 Banerjee, S.; Chiang, M. H.; Lee, J. C.; Akinwande, D. Thinnest
1464 Nonvolatile Memory Based on Monolayer h-BN. *Adv. Mater.* **2019**,
1465 *31*, 1806790.
- 1466 (166) Midya, R.; Wang, Z.; Zhang, J.; Savel'ev, S. E.; Li, C.; Rao, M.;
1467 Jang, M. H.; Joshi, S.; Jiang, H.; Lin, P.; Norris, K.; Ge, N.; Wu, Q.;
1468 Barnell, M.; Li, Z.; Xin, H. L.; Williams, R. S.; Xia, Q.; Yang, J. J.
1469 Anatomy of Ag/Hafnia-Based Selectors with 10¹⁰ Nonlinearity. *Adv.*
1470 *Mater.* **2017**, *29*, 1604457.
- 1471 (167) Hsu, C.-W.; Wang, Y.-F.; Wan, C.-C.; Wang, I.-T.; Chou, C.-
1472 T.; Lai, W.-L.; Lee, Y.-J.; Hou, T.-H. Homogeneous Barrier
1473 Modulation of TaO_x/TiO₂ Bilayers for Ultra-High Endurance
1474 Three-Dimensional Storage-Class Memory. *Nanotechnology* **2014**,
1475 *25*, 165202.
- 1476 (168) Wu, H.; Li, X.; Wu, M.; Huang, F.; Yu, Z.; Qian, H. Resistive
1477 Switching Performance Improvement of Ta₂O_{5-x}/TaO_y Bilayer
1478 ReRAM Devices by Inserting AlO_δ Barrier Layer. *IEEE Electron*
1479 *Device Lett.* **2014**, *35*, 39–41.
- 1480 (169) Yu, M.; Cai, Y.; Wang, Z.; Fang, Y.; Liu, Y.; Yu, Z.; Pan, Y.;
1481 Zhang, Z.; Tan, J.; Yang, X.; Li, M.; Huang, R. Novel Vertical 3d
1482 Structure of TaO_x-Based RRAM with Self-Localized Switching Region
1483 by Sidewall Electrode Oxidation. *Sci. Rep.* **2016**, *6*, 21020.
- 1484 (170) Aratani, K.; Ohba, K.; Mizuguchi, T.; Yasuda, S.; Shiimoto, T.;
1485 Tsushima, T.; Sone, T.; Endo, K.; Kouchiyama, A.; Sasaki, S.;
1486 Maesaka, A.; Yamada, N.; Narisawa, H. A Novel Resistance Memory
1487 with High Scalability and Nanosecond Switching. Proceedings from
1488 the *International Electron Devices Meeting (IEDM)*, Washington, DC,
1489 December 10–12, 2007; IEEE: New York, 2007; pp 783–786.
- 1490 (171) Tsai, T.-M.; Chang, K.-C.; Chang, T.-C.; Syu, Y.-E.; Chuang,
1491 S.-L.; Chang, G.-W.; Liu, G.-R.; Chen, M.-C.; Huang, H.-C.; Liu, S.-
1492 K.; Tai, Y.-H.; Gan, D.-S.; Yang, Y.-L.; Young, T.-F.; Tseng, B.-H.;
1493 Chen, K.-H.; Tsai, M.-J.; Ye, C.; Wang, H.; Sze, S. M.; et al. Bipolar
1494 Resistive RAM Characteristics Induced by Nickel Incorporated into
1495 Silicon Oxide Dielectrics for IC Applications. *IEEE Electron Device*
1496 *Lett.* **2012**, *33*, 1696.
- 1497 (172) Belmonte, A.; Kim, W.; Chan, B. T.; Heylen, N.; Fantini, A.;
1498 Houssa, M.; Jurczak, M.; Goux, L. 90 nm W/Al₂O₃/TiW/Cu 1T1R
1499 CBRAM Cell Showing Low-Power, Fast and Disturb-Free Operation.
1500 Proceedings from the *IEEE International Memory Workshop (IMW)*,
1501 Monterey, CA, May 26–29, 2013; IEEE: New York, 2013; pp 26–30.
- 1502 (173) Zhou, Q.; Zhai, J. HfO_x Bipolar Resistive Memory with
1503 Robust Endurance Using ZrN_x as Bottom Electrode. *Appl. Surf. Sci.*
1504 **2013**, *284*, 644–650.
- 1505 (174) Vianello, E.; Molas, G.; Longnos, F.; Blaise, P.; Souchier, E.;
1506 Cagil, C.; Palma, G.; Guy, J.; Bernard, M.; Reyboz, M.; Rodriguez, G.;
1507 Roule, A.; Carabasse, C.; Delaye, V.; Jousseume, V.; Maitrejean, S.;
1508 Reimbold, G.; De Salvo, B.; Dahmani, F.; Verrier, P.; Bretegnier, D.;
1509 Liebault, J. Sb-Doped GeS₂ as Performance and Reliability Booster in
1510 Conductive Bridge RAM. Proceedings from the *International Electron*
1511 *Devices Meeting (IEDM)*, San Francisco, CA, December 10–13, 2012;
1512 IEEE: New York, 2012; pp 741–744.
- 1513 (175) Rahaman, S. Z.; Maikap, S.; Chiu, H.-C.; Lin, C.-H.; Wu, T.-
1514 Y.; Chen, Y.-S.; Tzeng, P.-J.; Chen, F.; Kao, M.-J.; Tsai, M. J. Bipolar
Resistive Switching Memory Using Cu Metallic Filament in Ge_{0.4}Se_{0.6}
Solid Electrolyte. *Electrochem. Solid-State Lett.* **2010**, *13*, H159–H162.
- (176) Zaffora, A.; Cho, D.-Y.; Lee, K.-S.; Di Quarto, F.; Waser, R.;
Santamaria, M.; Valov, I. Electrochemical Tantalum Oxide for
Resistive Switching Memories. *Adv. Mater.* **2017**, *29*, 1703357.
- (177) Wang, G.; Long, S.; Yu, Z.; Zhang, M.; Ye, T.; Li, Y.; Xu, D.;
Lv, H.; Liu, Q.; Wang, M.; Xu, X.; Liu, H.; Yang, B.; Suñé, J.; Liu, M.
Improving Resistance Uniformity and Endurance of Resistive
Switching Memory by Accurately Controlling the Stress Time of
Pulse Program Operation. *Appl. Phys. Lett.* **2015**, *106*, 092103.
- (178) Kim, K.-H.; Hyun Jo, S.; Gaba, S.; Lu, W. Nanoscale Resistive
Memory with Intrinsic Diode Characteristics and Long Endurance.
Appl. Phys. Lett. **2010**, *96*, 053106.
- (179) Kempen, T.; Waser, R.; Rana, V. 50x Endurance Improvement
in TaO_x RRAM by Extrinsic Doping. Proceedings from the *IEEE*
International Memory Workshop (IMW), Dresden, Germany, May 16–
19, 2021; IEEE: New York, 2021.