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Water-Mediated Ionic Migration in Memristive Nanowires with a Tunable Resistive Switching Mechanism

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*ab initio*

*NW synthesis*

*Device fabrication*

OE

*Atmosphere-controlled electrical characterization*

*Density Functional Theory method*

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ZKLOH HOHFWURQÍLRQ LQWHUDFWLRQ ZDV G

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*Influence of moisture on electronic conduction*

~1.6  $\mu\text{m}$  and ~ 100 nm (aspect ratio of ~16) from the analysis of cross-sectional SEM images (Supplementary Information S1)



, *I-V*

*Influence of moisture on ionic conduction*

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*I-V*

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*i)*

*ii)*

*iii)*

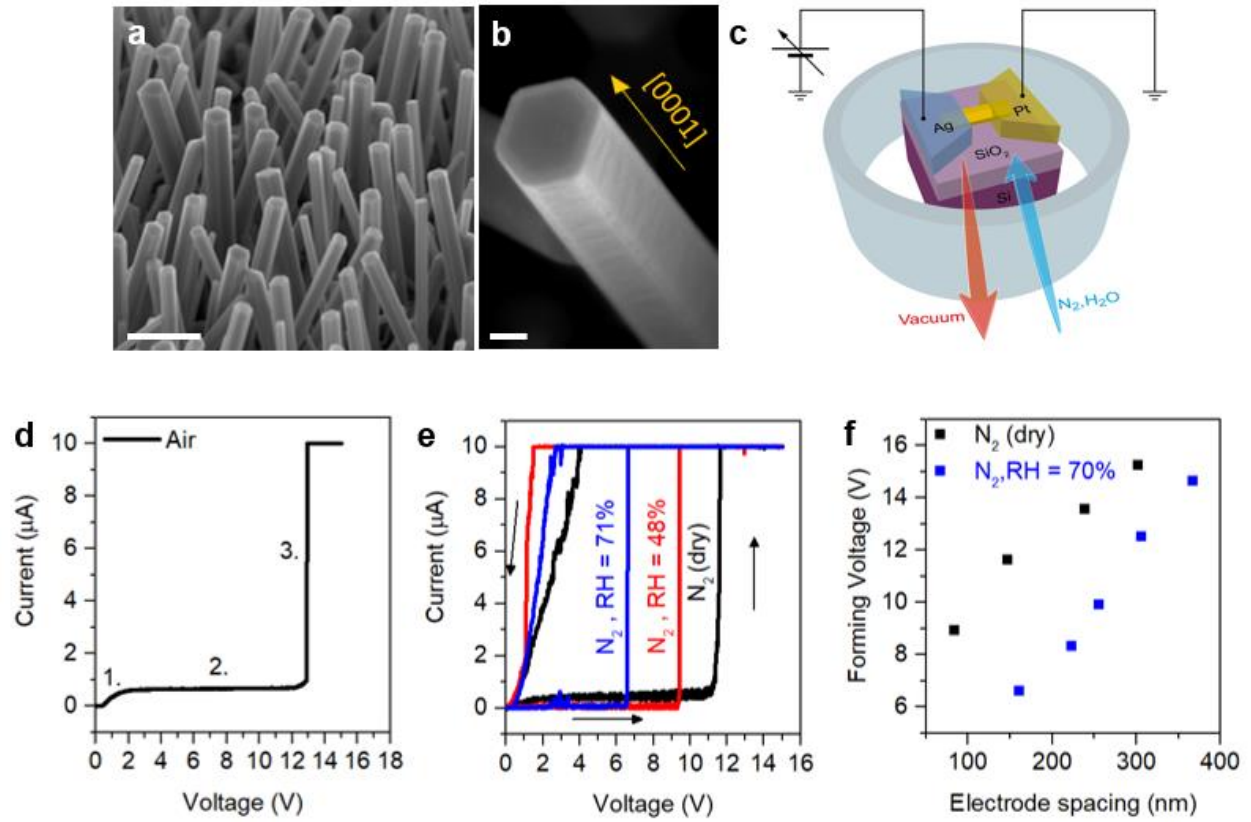
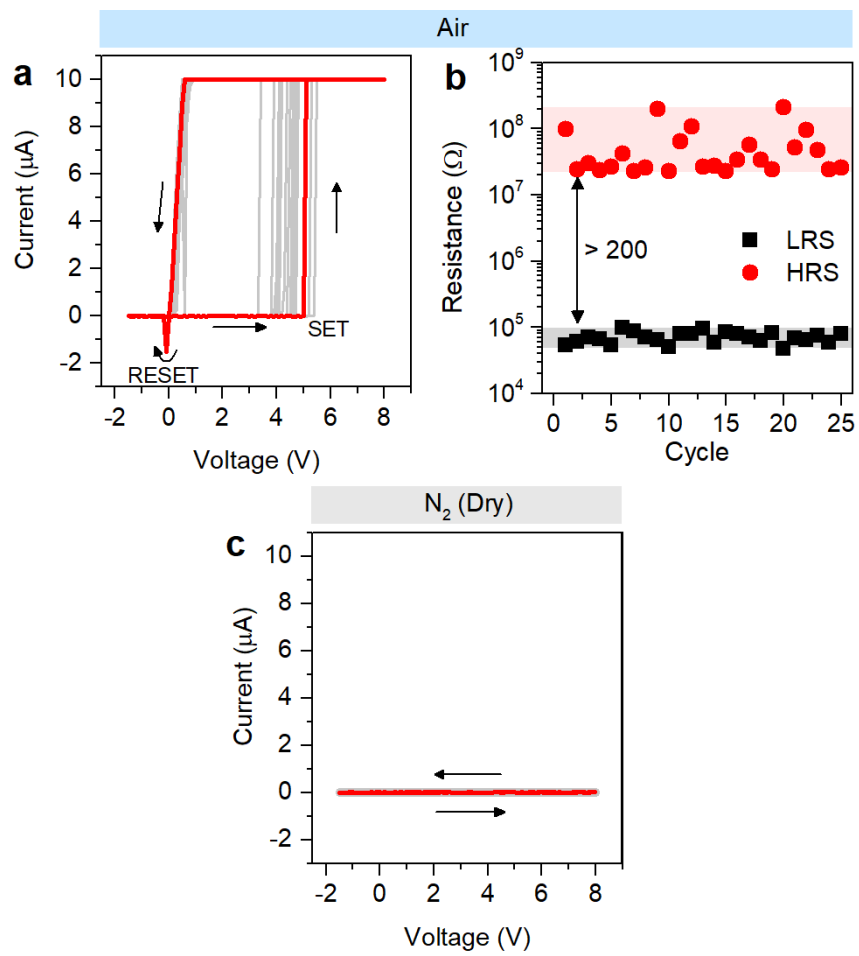


Figure. 1.

*Influence of moisture on resistive switching behavior*

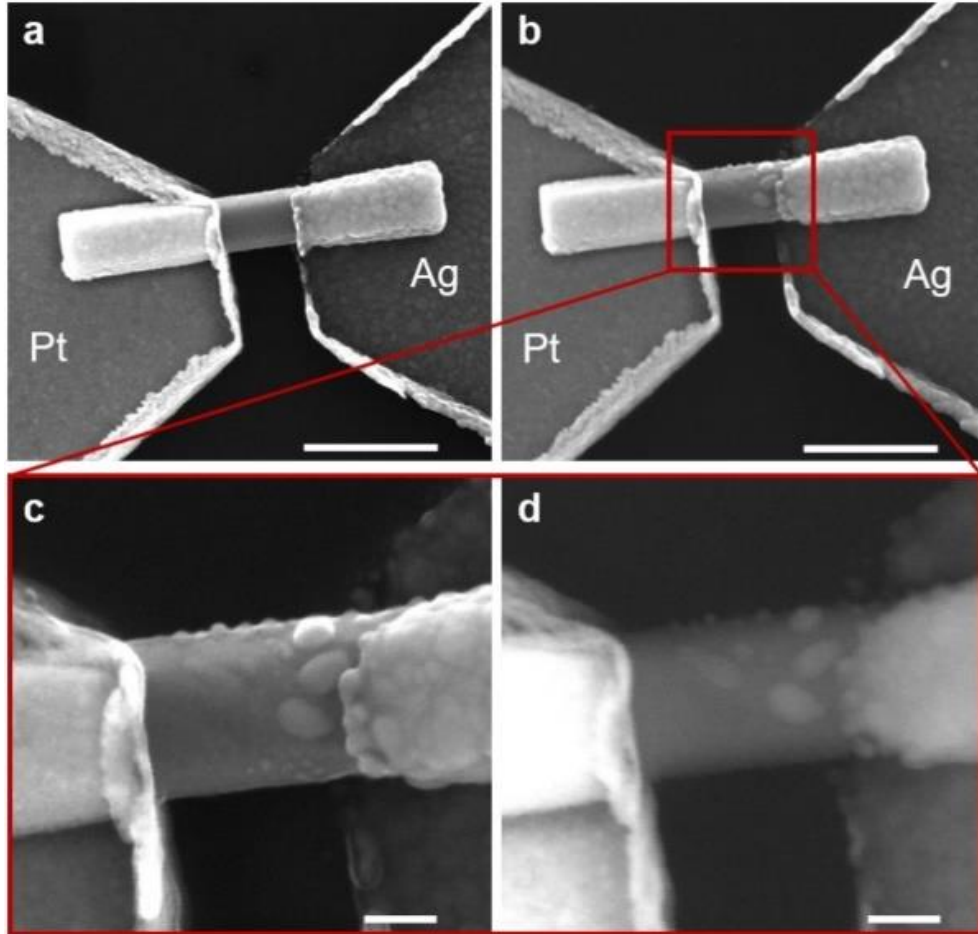


**Figure 2.**

*Morphological changes of the device after switching events*

wrinkled and corroded surface

(Supplementary Information S7), no significant corrosion effects were observed by exposing ZnO NWs to moisture and the smooth surface was preserved.



**Figure 3.**



*Density Functional Theory (DFT) simulations*

on the Zn atoms

belonging to the topmost surface layer

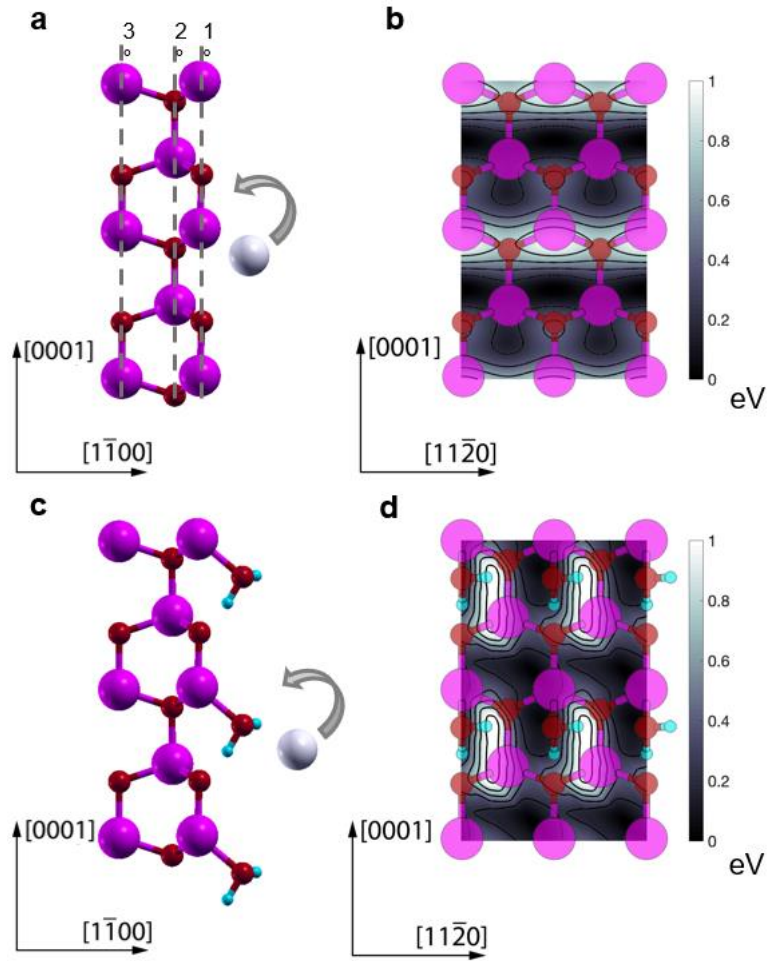
by the topmost Zn surface atoms

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the major effect of the adsorbed water is to avoid the repulsive interaction between the moving Ag atom and the topmost Zn atom. This is achieved by dissociation of water into  $\text{OH}^\ddagger + \text{H}^\ddagger$  (where  $\ddagger$  indicates a bond with the surface). The  $\text{OH}^\ddagger$  dangling bond is saturated by the Ag atom moving in the direction of the electric field ([0001]). As the OH group is located exactly between the Ag and the topmost Zn, the repulsive interaction between them is negated. Because the single  $\text{H}^\ddagger$  adsorbed on the surface plays no role, we can assume that the same mechanism holds for a surface covered with only OH groups. Based on these results, electrolysis is suggested not to influence ionic

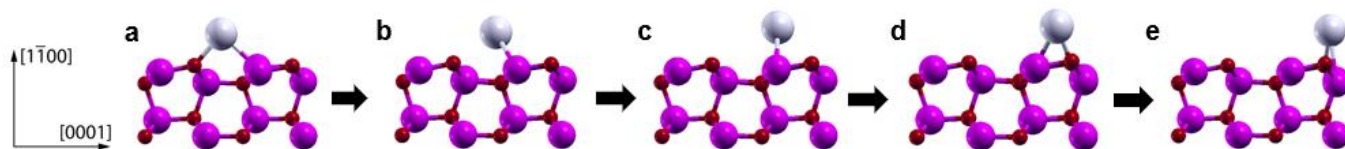
conduction mechanism even if it is not possible to exclude that this effect can be involved in regulating the memristive cell electrochemistry participating in the counter electrode reaction as reported in thin-film based devices, even if in our case we did not observe any bubbles or deformation of metallic electrodes related to electrolysis phenomena. Last, it should be remarked that moisture is expected to support corrosion of the Ag conductive path after its formation (i.e. in the ON state), leading to an expected poorer retention characteristic of the device.



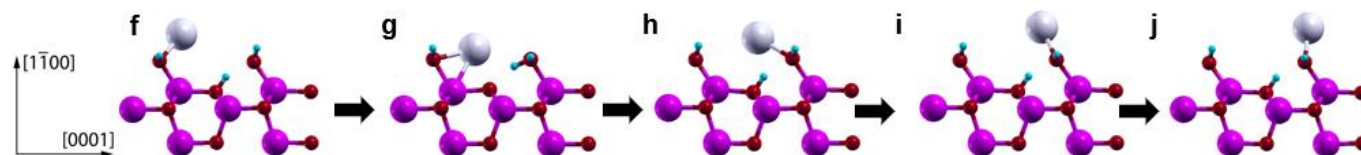
**Figure 4.**

In panel (a), 1°, 2° and 3° dashed line indicate the position of, respectively, the topmost, second highest and third highest ZnO layers, respectively

**ZnO bare surface**



**H<sub>2</sub>O-covered ZnO surface**



**Figure. 5**



## Supporting Information

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## Notes

There are no conflicts of interest to declare.

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*Resistive Switching - From Fundamentals of Nanoionic Redox Processes to Memristive Device Applications*

*Nano Lett.* **2010** 10 ±

:DQJ = -RVKL 6 6DYHO↑HY 6 ( -LDQJ +

*Nat. Mater.* **2017** 16 ±

*Nat. Commun.* **2018** 9

5LFFLDUGL & %UDLQ(,QVSLUHG 6WUXFWXUDO 3ODVWL  
0XOWL(7HUPLQDO 6HOI(2UJDQLJLQJ *Adv. Funct. Mater.* **2020** 1DQRZ

*Nat. Nanotechnol.* **2020**

*Adv. Electron. Mater.* **2017** 3

*Nature* **2015** 521 ±

:DQJ = -RVKL 6 6DYHO¶HY 6 6RQJ : 0LG\D 5 /L

*Nat.*

*Electron.* **2018** 1 ±

*Nat. Mach. Intell.* **2019** 1 ±

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*Nat. Electron.* **2019** 2 ±

*Nat. Electron.* **2020** 3 ±

*Nature* **2008** 453 ±

*Nat. Nanotechnol.*

**2008** 3 ±

*Nat.*

*Nanotechnol.* **2013** 8 ±

*Scr.*

*Mater.* **2020** 187 ±

*J. Alloys Compd.* **2020** 835

*Adv. Mater.* **2009** 21 ± ±

*J. Phys. D. Appl. Phys.* **2018** 51

*Adv.*

*Funct. Mater.* **2012** 22 ±

*ACS Nano* **2013** 7 ±

*Nano Energy* **2020**

*Nanotechnology* **2017** 28

*Adv. Electron. Mater.* **2018** 4

*Adv. Mater.* **2018** 30

*Adv.*

*Funct. Mater.* **2015** 25 ±

2

*Adv. Electron. Mater.* **2018** 4

*Nat. Commun.* **2013** 4

*Adv. Mater.* **2015** 27 ±

*Sci. Technol. Adv. Mater.* **2019**

20 ±

*Adv. Funct. Mater.* **2015** 25

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*Small*

**2017** 13

*Nano Lett.* **2011** 11 ±

*Nano Lett.* **2012** 12 ±

*Adv. Electron. Mater.* **2019** 5

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*J. Mater. Chem. C* **2017** 5 ±

*J. Phys. Chem. C* **2018** 122 ±

*Nanotechnology* **2019** 30

0L0DQR \* '¶2UWHQ]L / %H MWND . 0DQGULO H / \*L

*J. Phys. Chem. C* **2018** 122 ±

*J. Phys. Condens. Matter* **2009** 21

*Phys. Rev. Lett.* **1996** 77 ±

*Phys. Rev. B* **1990** 41 ±

*IEEE Trans. Nanotechnol.* **2020** 19 ±

*IEEE Trans. Nanotechnol.* **2020**

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*Adv. Mater. Interfaces* **2019** 6



*J. Phys. Chem. C* **2008** 112

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*Nanoscale* **2013** 5

*J. Mater. Chem.*

*C* **2015** 3

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*Nano*

*Lett.*

**2016**

16

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*Nat. Commun.* **2012** 3

*Nat.*

*Commun.* **2014** 5

*Adv.*

*Mater.* **2006** 18 ±

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*Phys.* *Chem.* *Chem.* *Phys.* **2017** 19 ±

*Solid State Ionics* **2017** 299 ±

