SUPPORTING INFORMATION

TITLE

Resistive Switching Dependence on Atomic Layer Deposition Parameters in HfO_2-based Memory Devices

AUTHORS

Raúl Zazpe[a], Mariana Ungureanu[a], Federico Golmar[a,b], †Pablo Stoliar[a,c,d], Roger Llopis[a], Fèlix Casanova[a,e], David F. Pickup[f,g], Celia Rogero[f,h], and Luis E. Hueso[a,e]*

[a] CIC nanoGUNE Consolider, Tolosa Hiribidea 76, 20018 Donostia - San Sebastian, Basque Country, Spain.

[b] I.N.T.I. - CONICET, Av. Gral. Paz 5445, Ed. 42, B1650JKA, San Martín, Bs As, Argentina.

[c] LPS, CNRS UMR 8502, Université Paris Sud, Bât 510, 91405 Orsay, France

[d] ECyT, UNSAM, Martín de Irigoyen 3100, B1650JKA, San Martín, Bs As, Argentina

[e] IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Basque Country, Spain.

[f] Material Physics Center (MPC), Centro de Física de Materiales (CSIC-UPV-EHU), 20018 San Sebastían, Spain.

[g] Departamento de Física Aplicada I, Universidad del País Vasco, 20018 San Sebastián, Spain.

[h] Donostia International Physics Center (DIPC), 20018 San Sebastián, Spain.

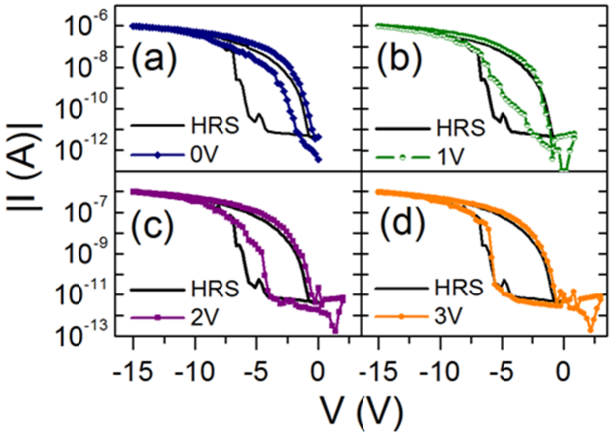

Fig. S1. Minor loops (a-d) obtained for the negative branch of the I-V curve for the Co/HfO_2/Ti device (HfO_2 deposited at 300 °C and 5 seconds of purge time) when applying different A voltage values indicated at the legend. The SET and RESET processes occur at opposite polarity indicating bipolar resistive switching behavior.

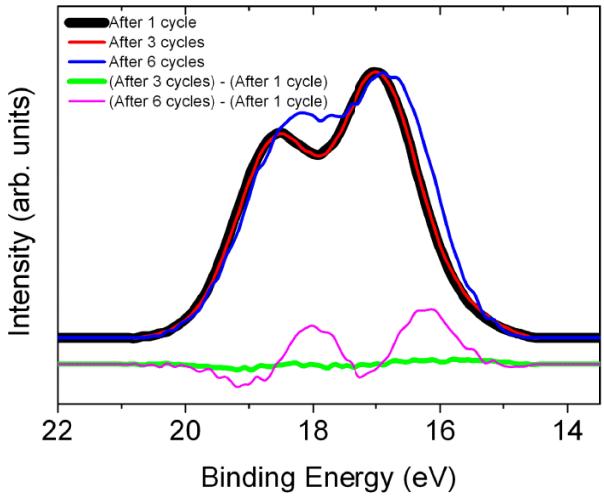

Fig. S2. XP Hf 4*f* spectra from a hysteretic 200°C, 5s purge sample are shown after various sputter cycles, with subtracted spectra to highlight the differences. The data shows that no changes in the Hf 4*f* spectra were observed whilst sputtering through the bulk of the HfO_2 film (up to 4 sputter cycles), as shown by comparing spectra after 1 (black line) and 3 (red line) sputter cycles and the corresponding

difference spectrum (green line). This indicates that we don´t have any preferential sputtering. Further sputtering cycles lead to the detection of the underlying Ti at which stage a significant change in the Hf 4*f* spectrum is observed as shown by the ´After 6 cycles´ spectrum (blue line) and the corresponding difference spectrum (pink line). This change is attributed to the presence of Hf suboxides.

Table S1. Relative nitrogen content in HfO_2 films in the bulk and at the underlying Ti interface from samples prepared under different deposition condition.

		% Nitrogen in the HfO_2 film	
Deposition Temperature (°C)	Purge Time (s)	Bulk	At Ti interface
125	5	23	25
200	5	8	6
	35	6	2.5
	70	9	7
300	5	10	10
	35	11	10

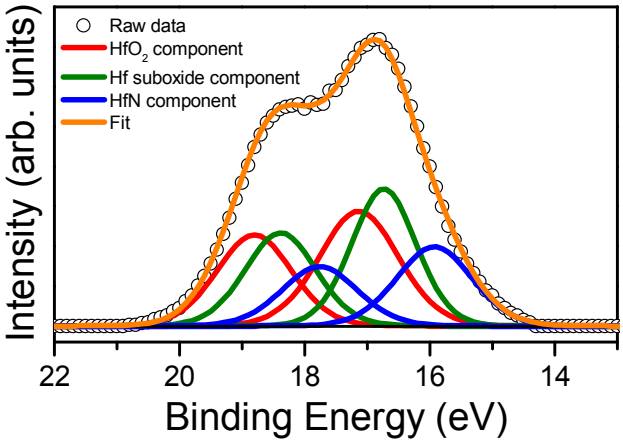

Fig. S3. XPS Hf 4f spectra of a sample deposited at 125 °C with a 5s purge time.

Table S2. Peak positions and spin-orbit splittings of the different components used to fit the Hf 4*f* and Ti 2*p* spectra.

XPS Region / Component	Binding Energy (eV) (±0.1eV)	Spin-orbit splitting (eV)	References
Hf $4f_{7/2}$			
HfO_2	17.1	1.7	1,2,3
HfO_{2-x}	16.5	1.7	4
Hf-N	15.9	1.9	2, 5
Ti $2p_{3/2}$			
Ti	453.7	5.9	6, 7
TiN	454.5	5.8	6, 8, 9
TiO_xN_y	456.4	5.5	6, 8, 10
TiO_2	458.2	5.6	6, 8, 10

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