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Supplementary Information

Solution processed metal oxo cluster for rewritable resistive memories

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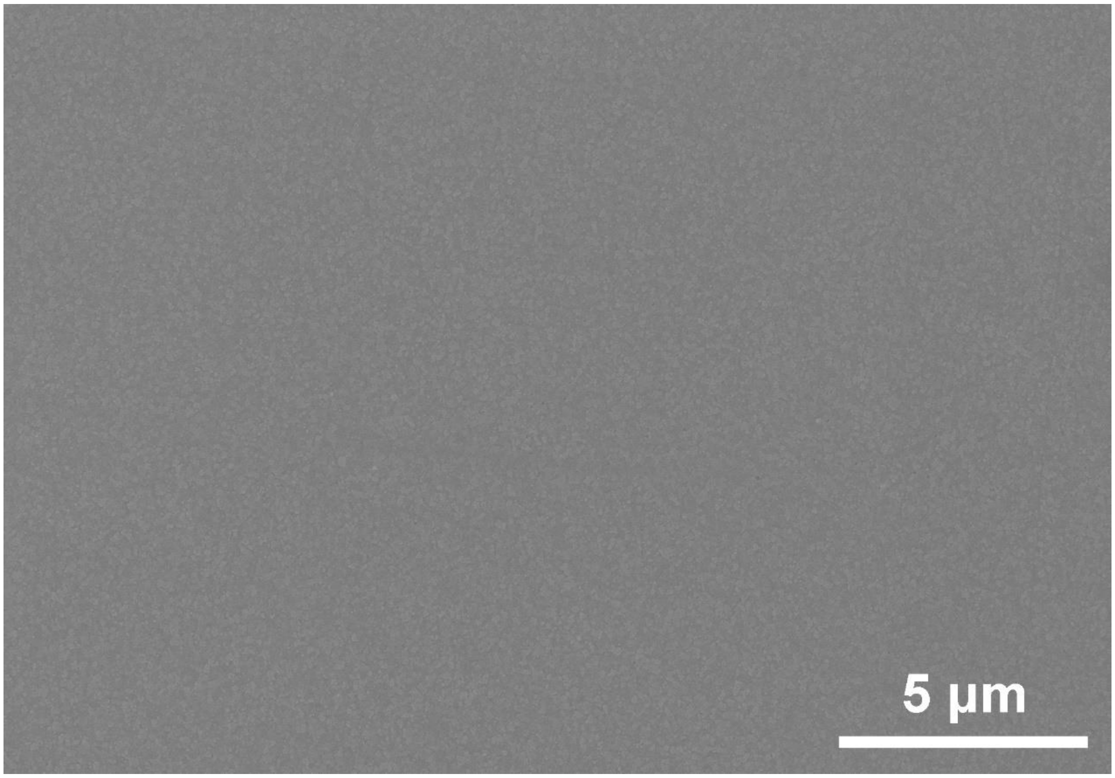

Figure S1. SEM of BiON NCs film on ITO glass.

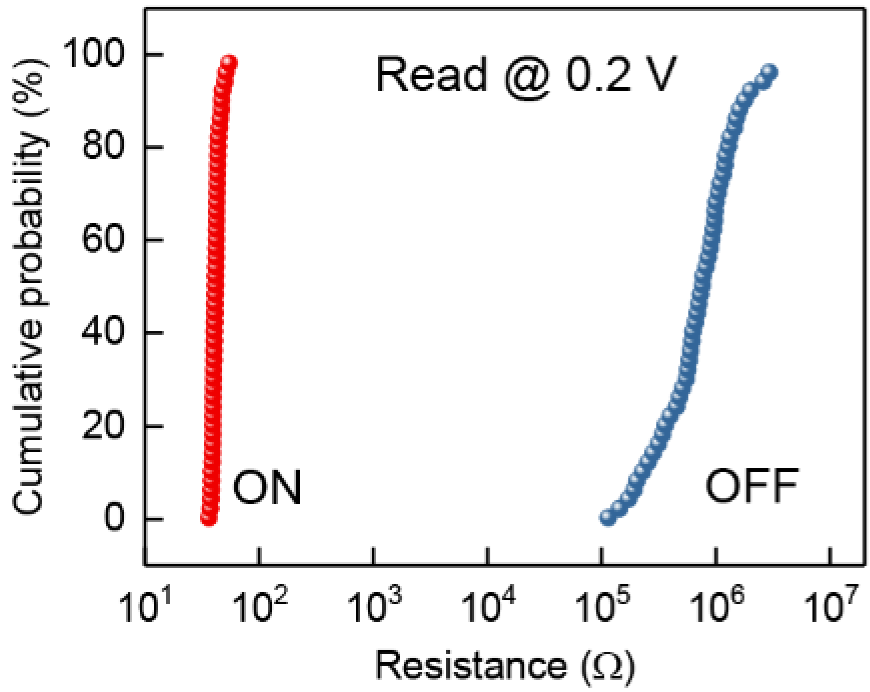

Figure S2 the cumulative resistance probability presents a centralized resistance distribution of LRS (35~55 Ω) and HRS (1.1×10^5~5.0×10^6 Ω) with a reliable 10^3 ON/OFF ratio.

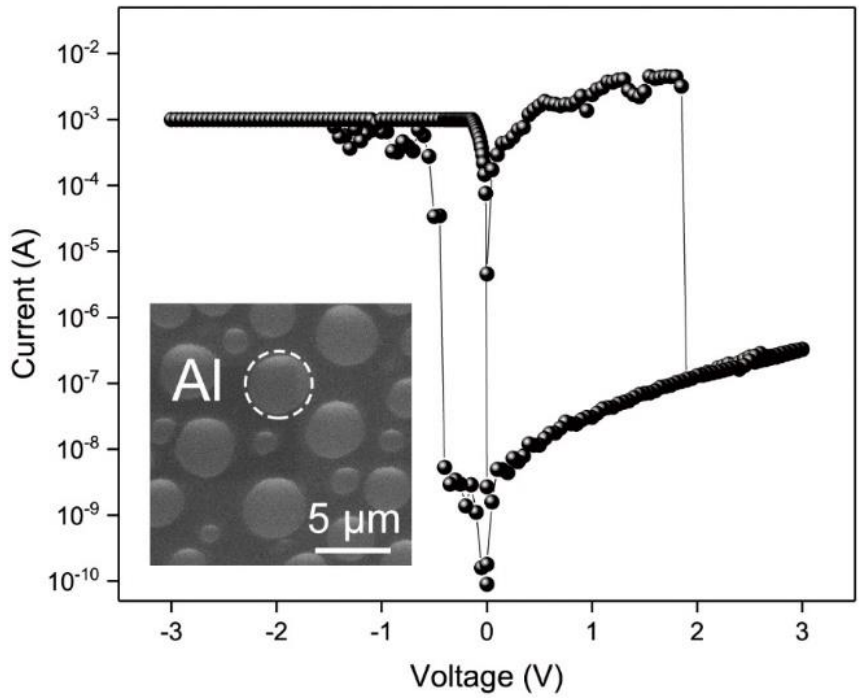

Figure S3 The *I*-*V* switching behavior of the micro-device with top electrode Al (diameter ~ 5 μm) by using an electric probe (diameter of probe tip ~ 1 μm)

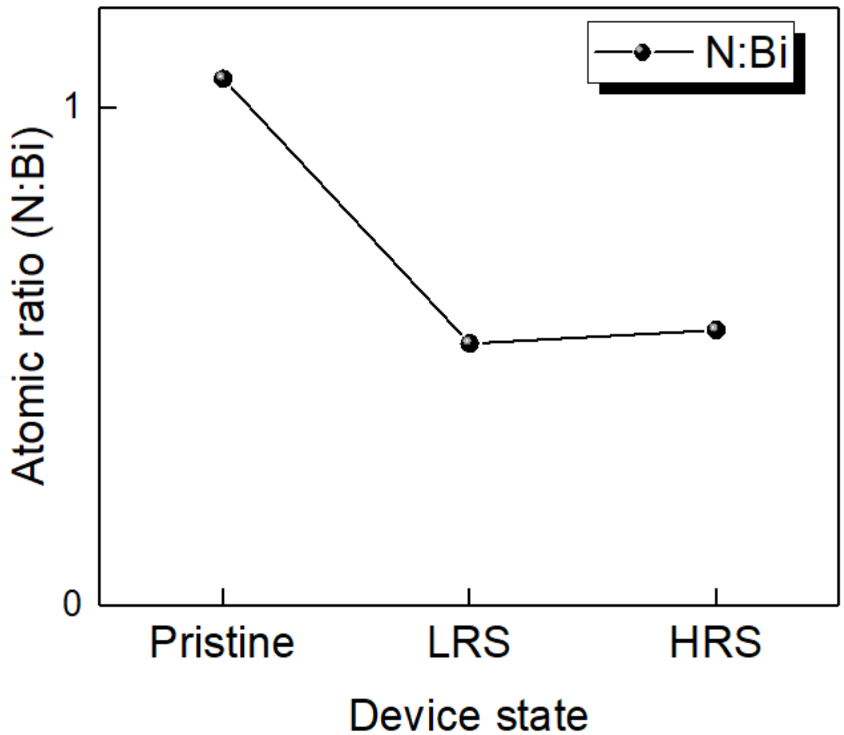

Figure S4 The atomic ratio of N: Bi for BiON in pristine state, LRS and HRS, respectively.

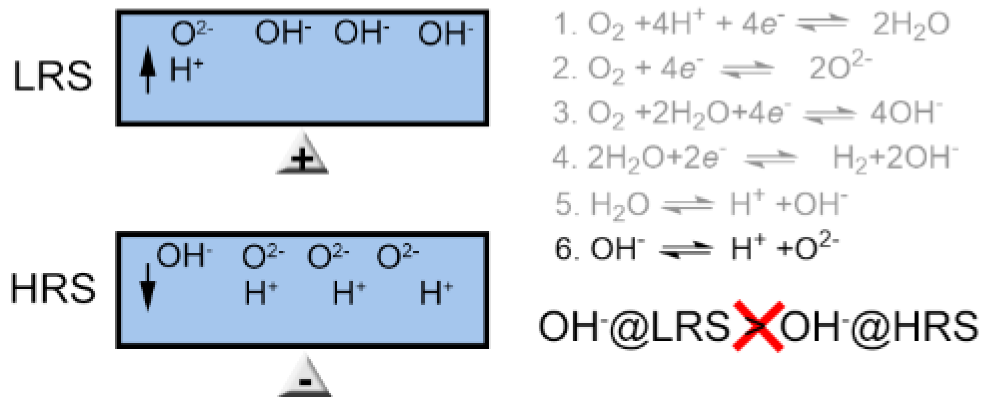

Figure S5. The schematic illustration of proton migration model. Reaction (1-5) could hardly occur since almost no water and oxygen can be observed in LRS and HRS (based on the result of XPS). Thus, the proton-induced OH^- content change should depend on Reaction (6). For example, an upward electric filed to simulate LRS, the H^+ migrate to top surface of the film and combine with O^{2-} to form more OH^-, leading to a higher OH^- content on the surface, i.e. OH^-@LRS > OH^-@HRS, which is contrary to XPS results (Table S1). In summary, the OH^- species was considered as the main mobile species rather than H^+.

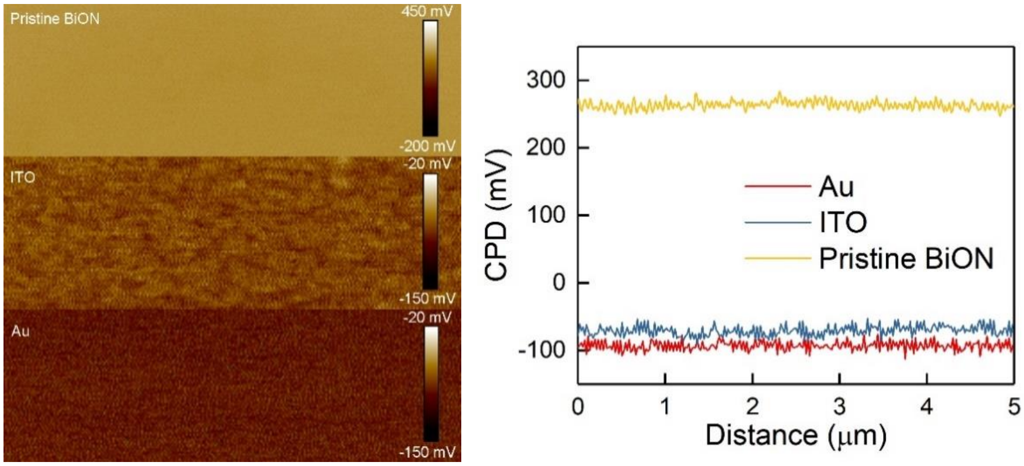

Figure S6. The work functions of the Au (φ_{Au} = 5.1 eV for calibration), bare ITO and pristine BiON film are measured by KPFM, the CPD values of which are about -90 mV, -70 mV and 260 mV, respectively. φ_{ITO} = 5.1-(-0.09)+(-0.07) =5.12 eV, φ_{BiON} = 5.1-(-0.09)+0.26 =5.45 eV.

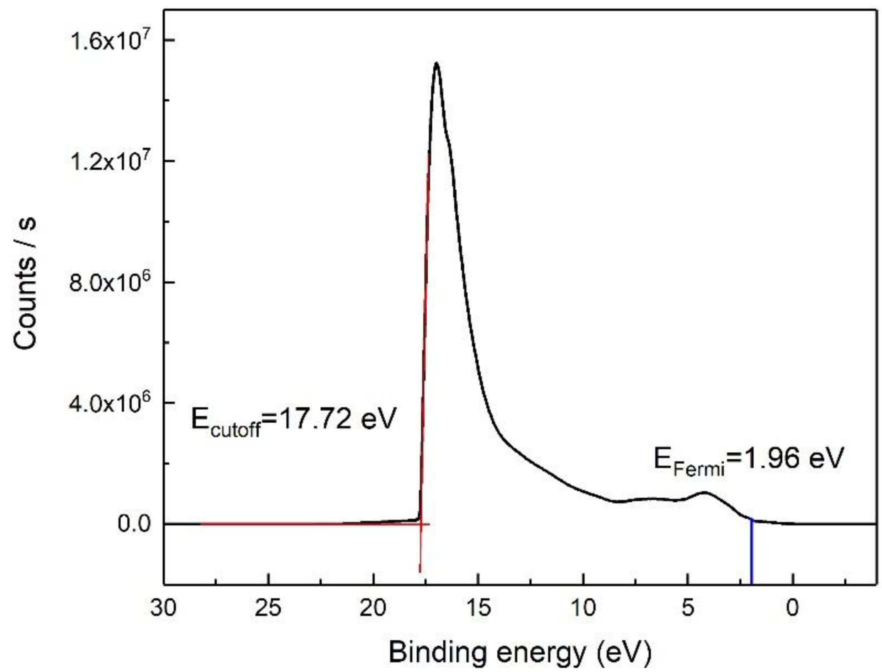

Figure S7. The ultraviolet photoelectron spectroscopy (UPS) of BiON nanocrystal, the work function φ_{BiON} = h_n-(E_{Cutoff} -E_F), φ_{BiON} = 21.2 eV- (17.72 eV- 1.96 eV) =5.44 eV

Table S1 the peak fitting of high resolution XPS of O1s

O1s	Peak	Position (eV)	Area	Area ratio	
				OH^- / NO_3^-	O^{2-} / NO_3^-
LRS	NO_3^-	531.66	42260.48		
	OH^-	530.02	21471.41		
	O^{2-}	529.0	14570.04		
				0.51	0.34
HRS	NO_3^-	531.96	38180.73		
	OH^-	530.11	31916.05		
	O^{2-}	529.31	11178.29		
				0.84	0.30

Table S2 XPS peaks identification and corresponding components in different depths of the device at LRS

Etching depth	Peaks identification	Corresponding components	Description
L1	O1s, Al2s, Al2p	AlO_x, Al	Top electrode with surface oxidation
L2	Al2s, Al2p	Al	Top electrode
L3	O1s, Bi4d, Bi4f, Al2s, Al2p	Al, AlO_x, BiON	Al/BiON interface
L4	O1s, Bi4d, Bi4f	BiON	BiON film
L5	O1s, Sn3d, In3d, Bi4d(weak), Bi4f	BiON, ITO	BiON/ITO interface
L6	O1s, Sn3d, In3d	ITO	Bottom electrode