Supplementary Information

Designing zero-dimensional dimer-type all-inorganic perovskites for ultra-fast switching memory

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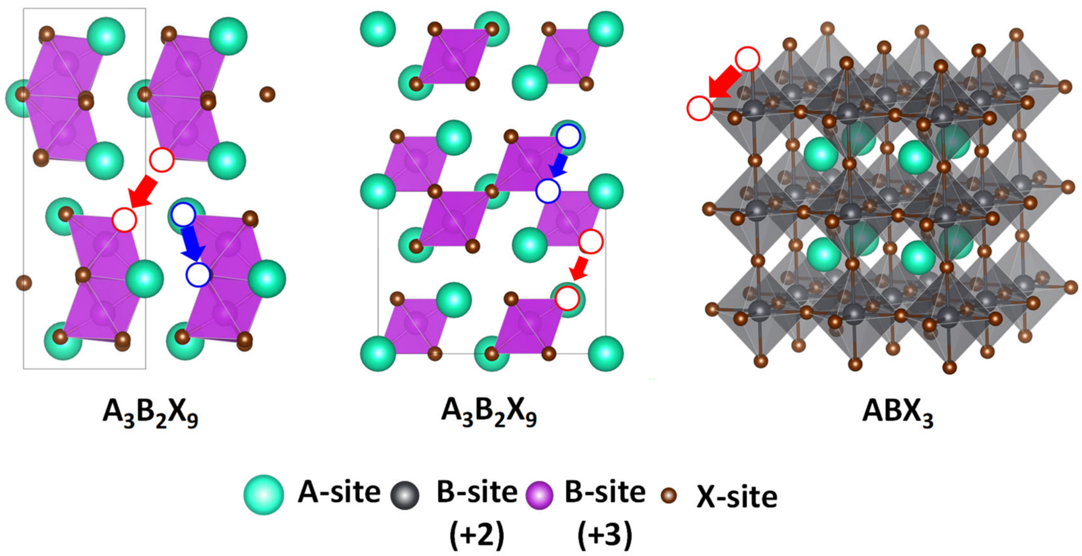
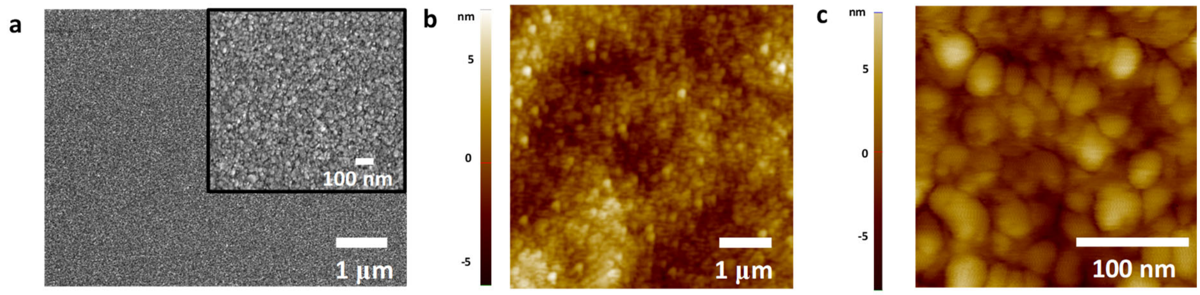
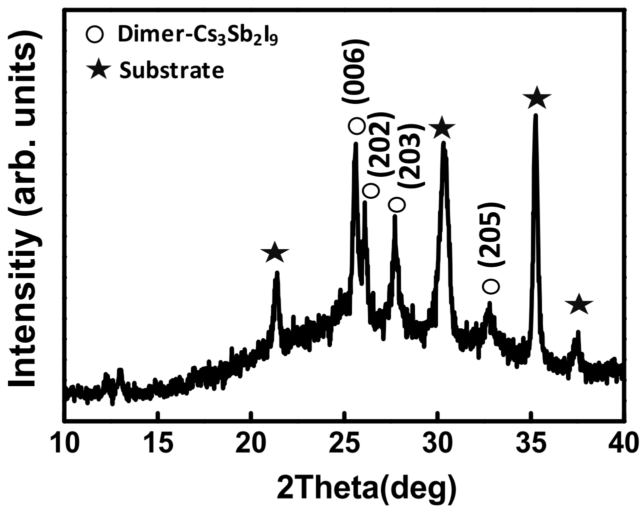


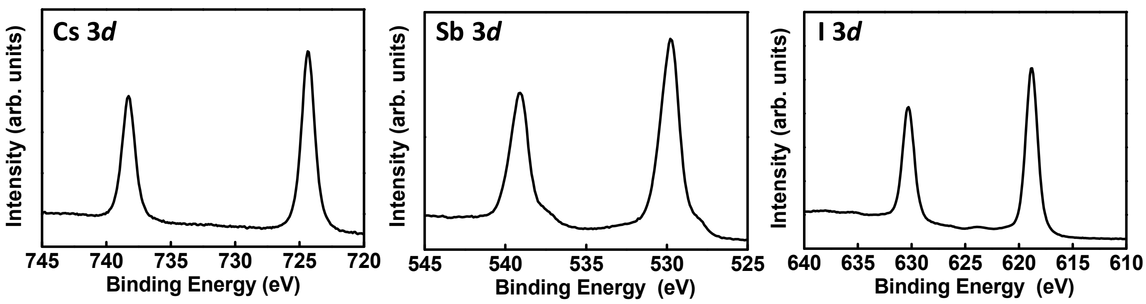
Supplementary Figure 1. Vacancy migration paths of HPs. Red circles and arrows represent inter-octahedron migration path. Blue circles and arrows represent intra-migration path. The rectangular boxes in schematic structures are the unit cell of crystal structure.



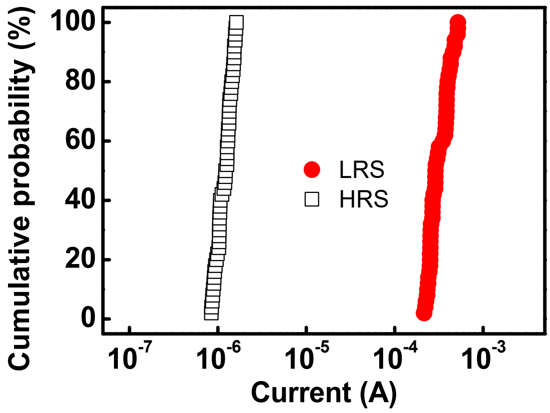
Supplementary Figure 2. (a) Plan-view SEM image of the dimer-$Cs_3Sb_2I_9$ thin film (inset: magnified image). (b) AFM image of the surface of the dimer-$Cs_3Sb_2I_9$. (c) Magnified AFM image of dimer-$Cs_3Sb_2I_9$.



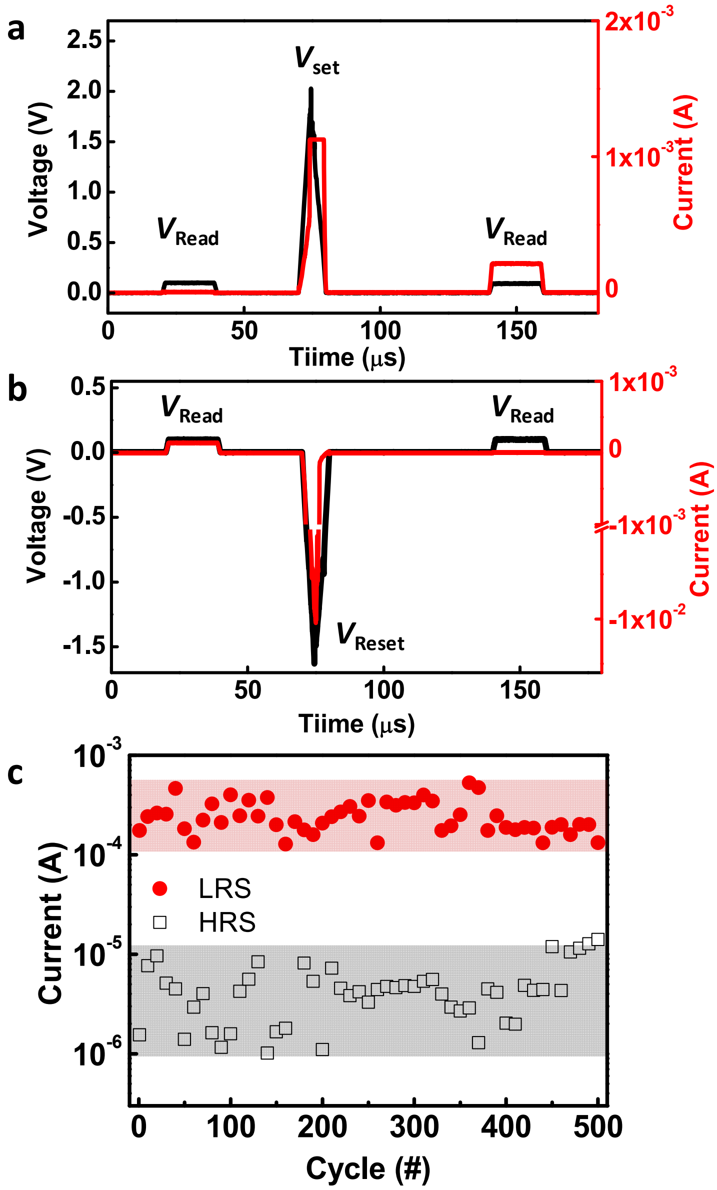
Supplementary Figure 3. XRD pattern of the dimer-$Cs_3Sb_2I_9$ thin film on ITO.



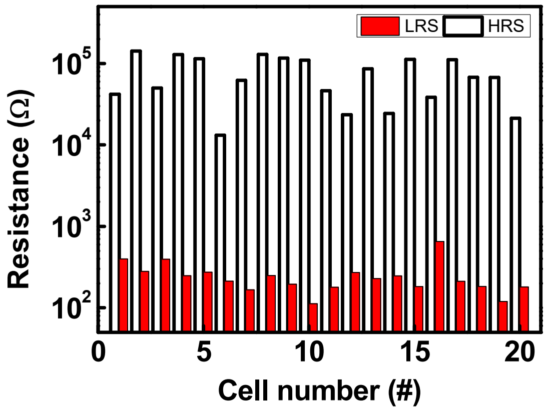
Supplementary Figure 4. XPS spectra of dimer-$Cs_3Sb_2I_9$ thin film.



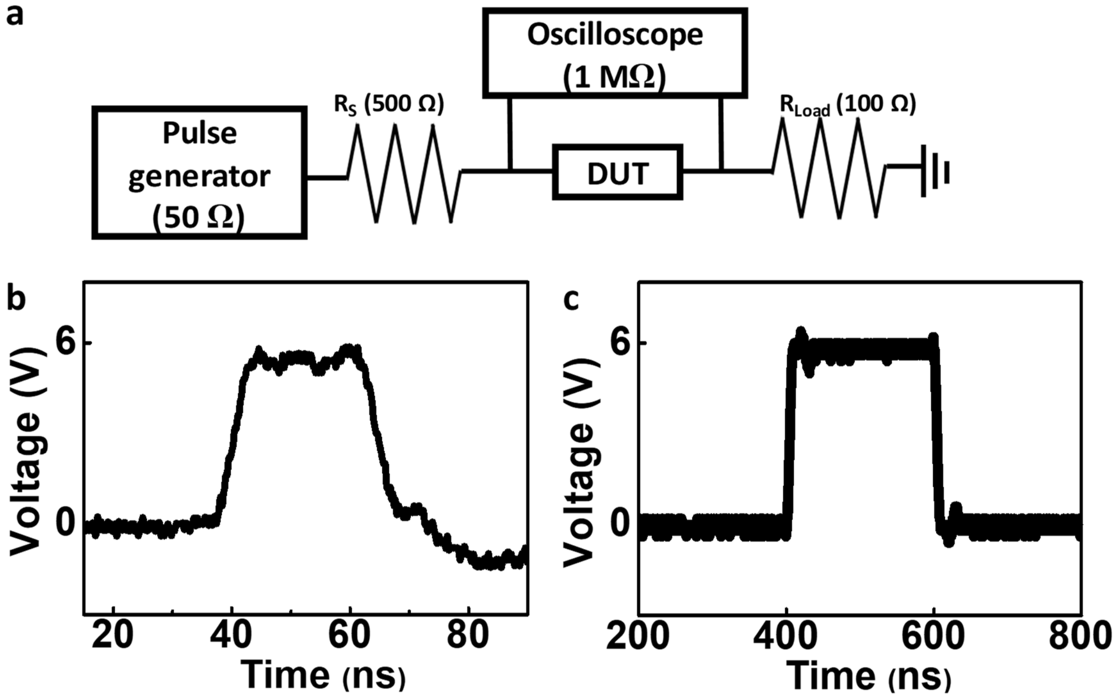
Supplementary Figure 5. Cumulative probability distributions of the resistance states during 50 consecutive voltage sweeps (0 V → 1 V → 0 V → -1 V → 0 V). The resistances of LRS and HRS are obtained at a read voltage of 0.1 V.



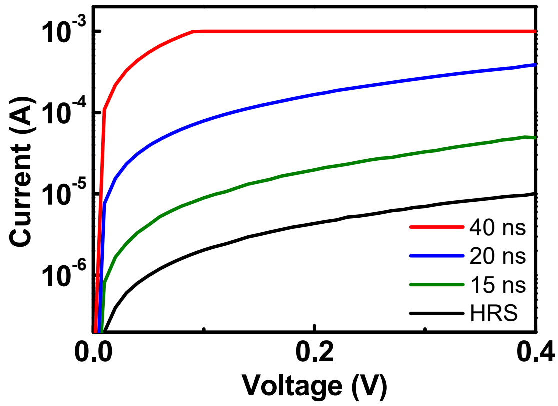
Supplementary Figure 6. Application of voltage pulses for (a) set and (b) reset processes. Positive triangular pulses that had a peak voltage of 2 V for the set process, and negative triangular pulses that had a peak of -1.6 V for the reset process, and the change of current level is measured under a read voltage of 0.1 V. (c) Endurance of dimer-$Cs_3Sb_2I_9$ RSM device.



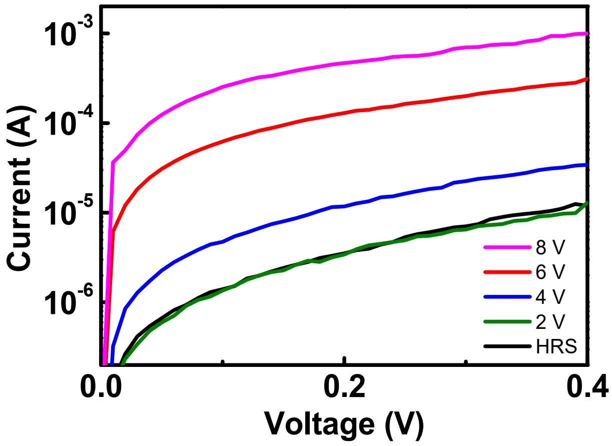
Supplementary Figure 7. Device-to-device variation of the dimer-$Cs_3Sb_2I_9$ RSM device. The resistance states of 20 devices are measured at a read voltage of 0.1 V.



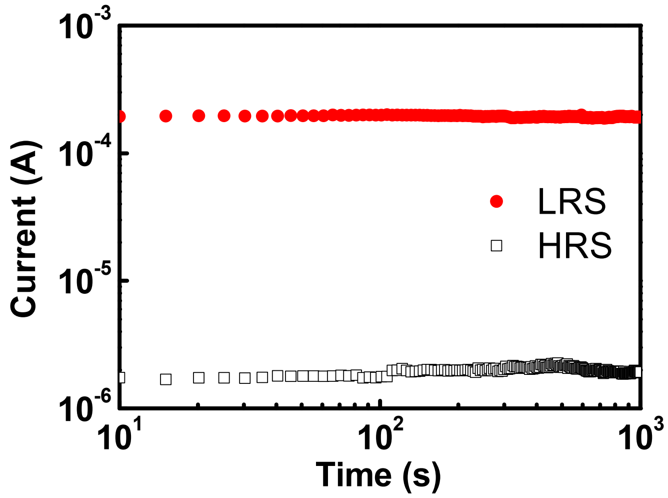
Supplementary Figure 8. a. Measurement scheme for pulse application. The waveforms of applied **b.** 20 ns and **c.** 200 ns pulses.



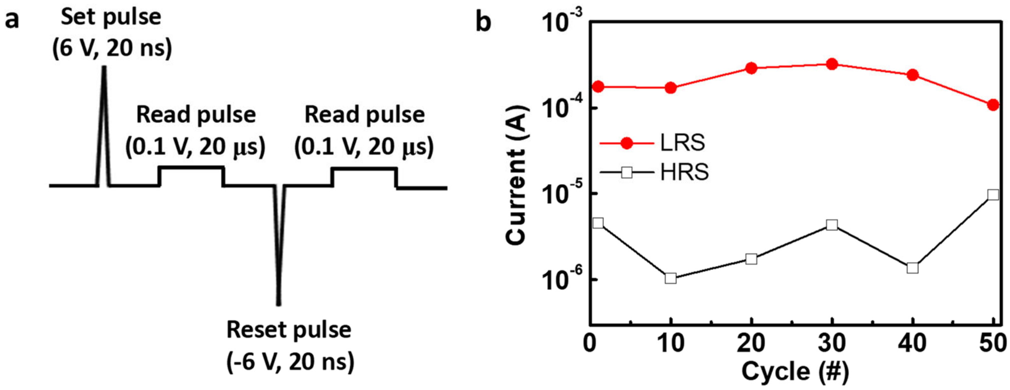
Supplementary Figure 9. *I-V* characteristics of the dimer-$Cs_3Sb_2I_9$ RSM by applying set pulses (6 V) with different pulse widths (15 ns, 20 ns, and 40 ns).



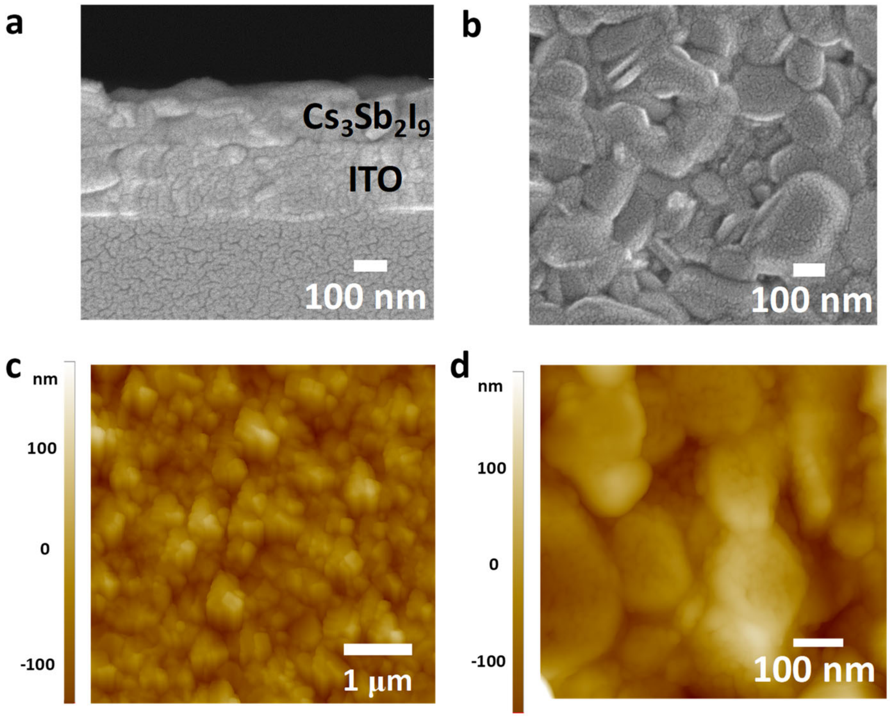
Supplementary Figure 10. *I-V* characteristics of the dimer-$Cs_3Sb_2I_9$ RSM by application of set pulses (20 ns) with different voltage amplitudes (2 V, 4 V, 6 V, and 8 V).



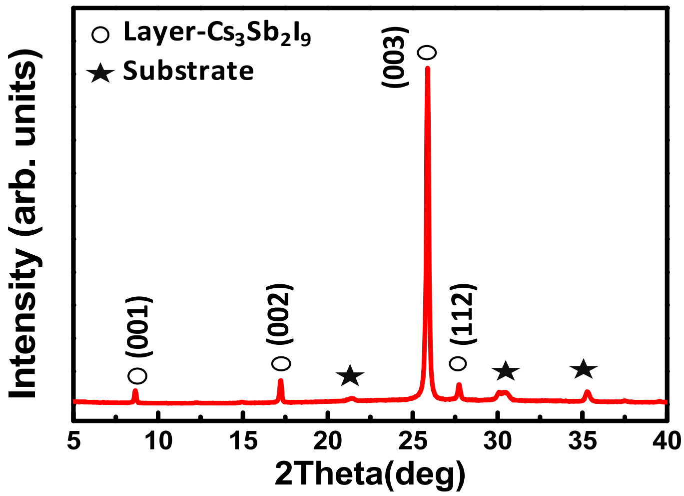
Supplementary Figure 11. Data retention characteristics of the dimer-$Cs_3Sb_2I_9$ RSM at LRS and HRS.



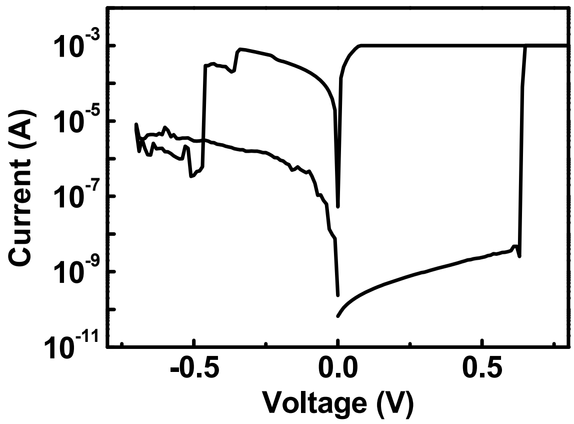
Supplementary Figure 12. (a) Applied pulse scheme. (b) Endurance of the dimer-$Cs_3Sb_2I_9$ RSM using 20 ns pulse application.



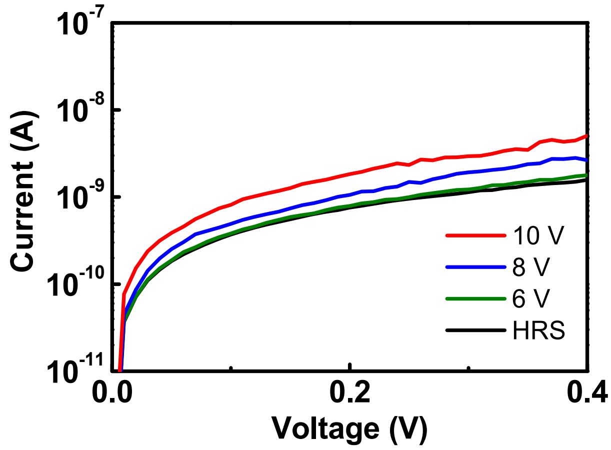
Supplementary Figure 13. (a) Cross-sectional SEM image of the layer-$Cs_3Sb_2I_9$ film. (b) Plan-view SEM image of the layer-$Cs_3Sb_2I_9$ film. (c) AFM image of the surface of the layer-$Cs_3Sb_2I_9$ film. (d) Magnified AFM image of layer-$Cs_3Sb_2I_9$.



Supplementary Figure 14. XRD pattern of layer-$Cs_3Sb_2I_9$ thin film on ITO.



Supplementary Figure 15. *I-V* characteristics of layer-$Cs_3Sb_2I_9$ RSM device.



Supplementary Figure 16. *I-V* characteristics of the layer-$Cs_3Sb_2I_9$ RSM by application of set pulse (20 ns) with different voltage amplitudes (6 V, 8 V, and 10 V).

Supplementary Table 1. Vacancy migration barriers of seven compounds according to two migration paths at final steps.

Compound name	Inter-octahedron barrier (eV)	Intra-octahedron barrier (eV)
$Cs_3Sb_2I_9$(layer)	0.54	0.24
$Cs_3Sb_2I_9$(dimer)	0.47	0.37
$Cs_3Bi_2I_9$(layer)	0.45	0.25
$FA_3Sb_2I_9$(dimer)	1.2	0.79
$CsPdBr_3$	0.44	0.14
$GA_3Sb_2Br_9$(layer)	0.96	0.34
$FA_3Sb_2I_9$(layer)	1.04	0.32

Supplementary Table 2. Switching speed of HP RSM devices.

Device structure	Switching speed	Reference
Au/$Cs_3Sb_2I_9$/ITO	20 ns	This work
Al/$CH_3NH_3PbI_3$:PVAm·HI/ITO	50 ns	1
Au/$CH_3NH_3PbI_3$/FTO	60 ns	2
Au/$(CH_3NH_3)_3Bi_2I_9$/ITO	100 ns	3
Pt/Zr-doped $CH_3NH_3PbI_3$/FTO	480 ns	4

1. Cao X., *et al.* Enhanced switching ratio and long-term stability of flexible RRAM by anchoring polyvinylammonium on perovskite grains. *ACS Appl. Mater. Interfaces* **11**, 35914-35923 (2019).

2. Ma H., *et al.* Interface state-induced negative differential resistance observed in hybrid perovskite resistive switching memory. *ACS Appl. Mater. Interfaces* **10**, 21755-21763 (2018).

3. Hwang B. & Lee J.-S. Lead-free, air-stable hybrid organic–inorganic perovskite resistive switching memory with ultrafast switching and multilevel data storage. *Nanoscale* **10**, 8578-8584 (2018).

4. He Y., *et al.* Impact of chemical doping on resistive switching behavior in zirconium-doped $CH_3NH_3PbI_3$ based RRAM. *Org. Electron.* **68**, 230-235 (2019).