ADVANCED MATERIALS

Supporting Information

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Iodine Vacancy Redistribution in Organic–Inorganic Halide Perovskite Films and Resistive Switching Effects

*Xiaojian Zhu, Jihang Lee, and Wei D. Lu**

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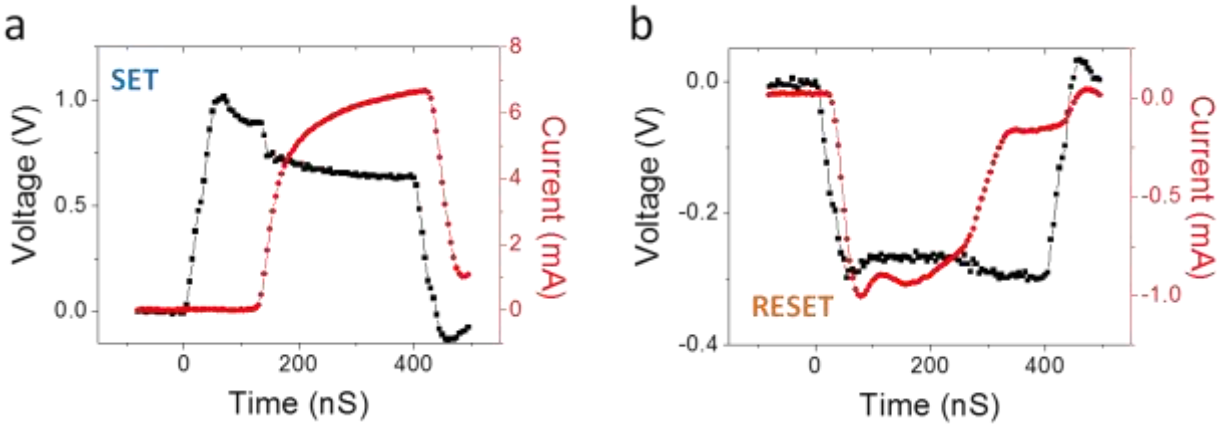


Figure S1. Transient switching characteristics of a Au/$MAPbI_3$/Au device during the (a) SET and (b) RESET processes.

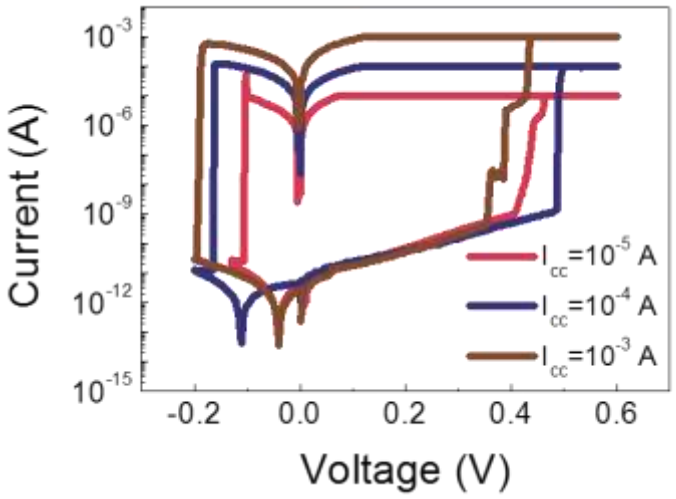


Figure S2. *I-V* characteristics of a Au/$MAPbI_3$/Au device during RS, with different SET current compliances (I_{cc}).

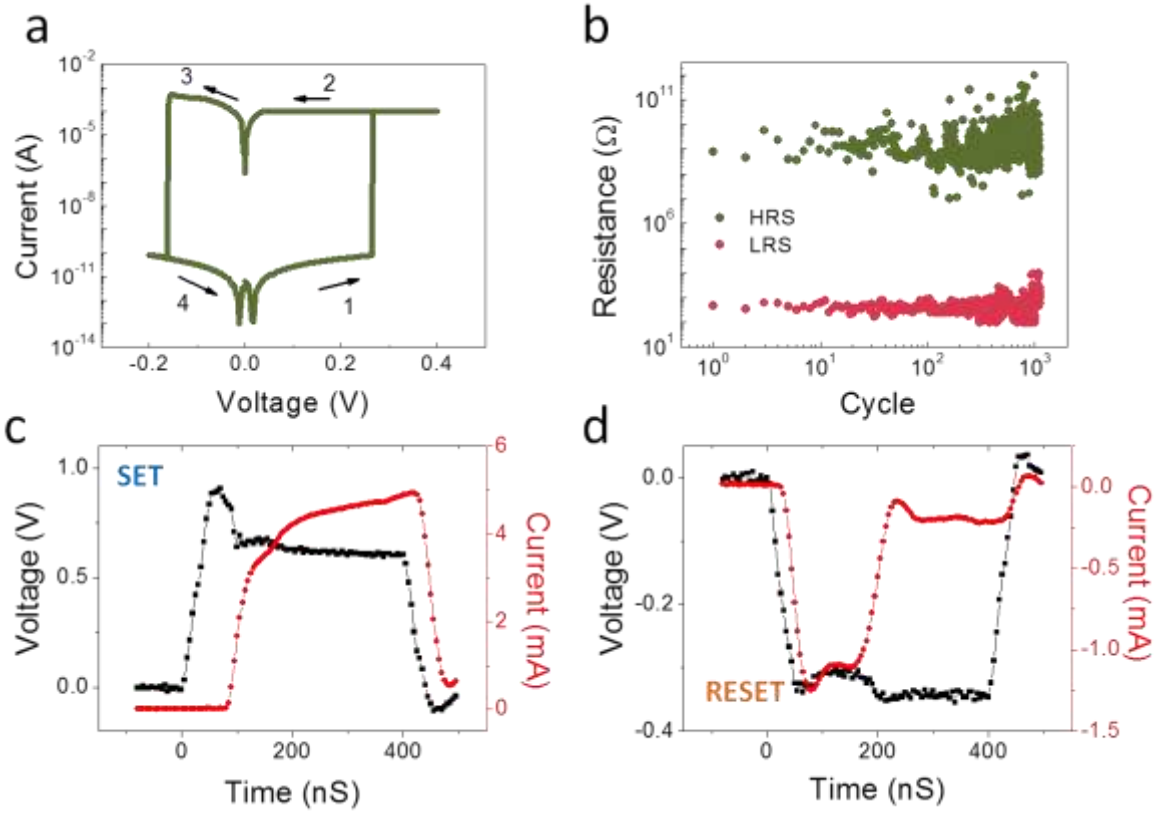
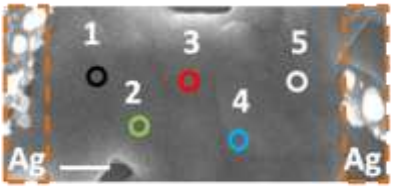


Figure S3. RS effects in a Ag/$MAPbI_3$/Au device, showing (a) *I-V* characteristics, (b) write/erase endurance test results, and transient switching characteristics during the (c) SET and (d) RESET processes.

	SET voltage (V)	RESET voltage (V)	HRS resistance (10^9 Ω)	LRS resistance (10^2 Ω)	HRS/LRS resistance ratio (10^6)
Au/$MAPbI_3$/Au	0.27 ± 0.10	-0.15 ± 0.09	3.46 ± 5.99	8.44 ± 6.01	4.11
Ag/$MAPbI_3$/Au	0.20 ± 0.07	-0.14 ± 0.08	5.45± 3.50	5.11 ± 6.97	11

Table S1. Comparison of RS performance, including SET voltage, RESET voltage, resistances at HRS and LRS and HRS/LRS resistance ratio of Au/$MAPbI_3$/Au and Ag/$MAPbI_3$/Au devices.



	1	2	3	4	5	Average
I:Pb	3.04	2.97	3.02	2.94	2.98	2.99±0.04

Figure S4. EDX analysis of the I:Pb distributions in an as-fabricated Ag/$MAPbI_3$/Ag device, at different positions across the film. Scale bar: 300 nm.

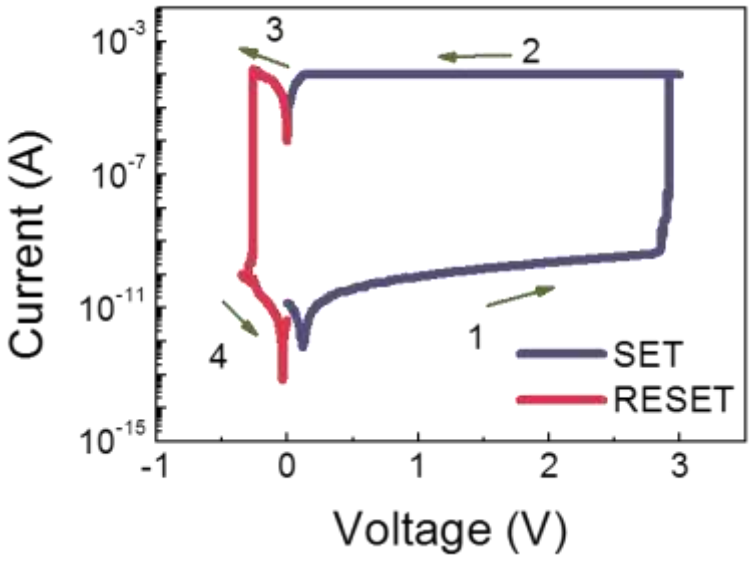


Figure S5. *I-V* characteristics of a Ag/$MAPbI_3$/Ag planar device during RS. During SET, a compliance current of 1×10^{-4} A was applied to prevent the device from hard breakdown.

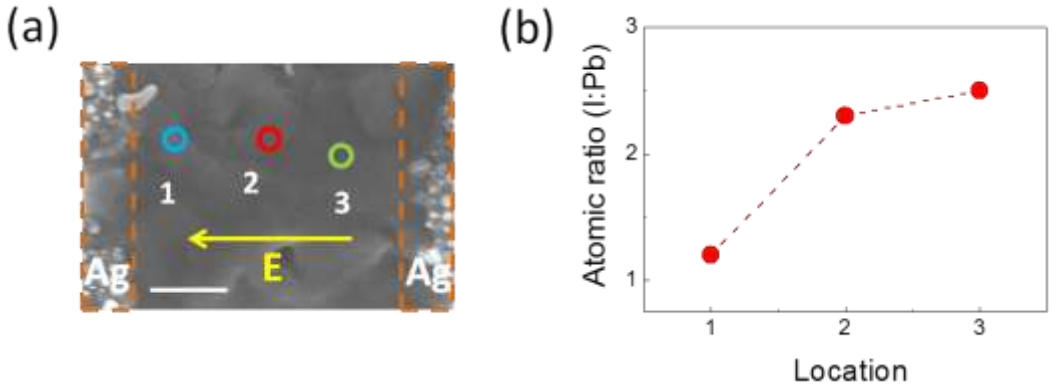


Figure S6. EDX analysis of the elemental distributions in a Ag/$MAPbI_3$/Ag device at LRS. The device was switched using the right electrode as the anode. a) SEM image of the device. Locations 1, 2 and 3 mark the positions where the EDX analyses were performed. The arrow illulstrates the electric field direction during SET. Scale bar: 300 nm. b) I:Pb atomic concentration ratios at locations 1-3.

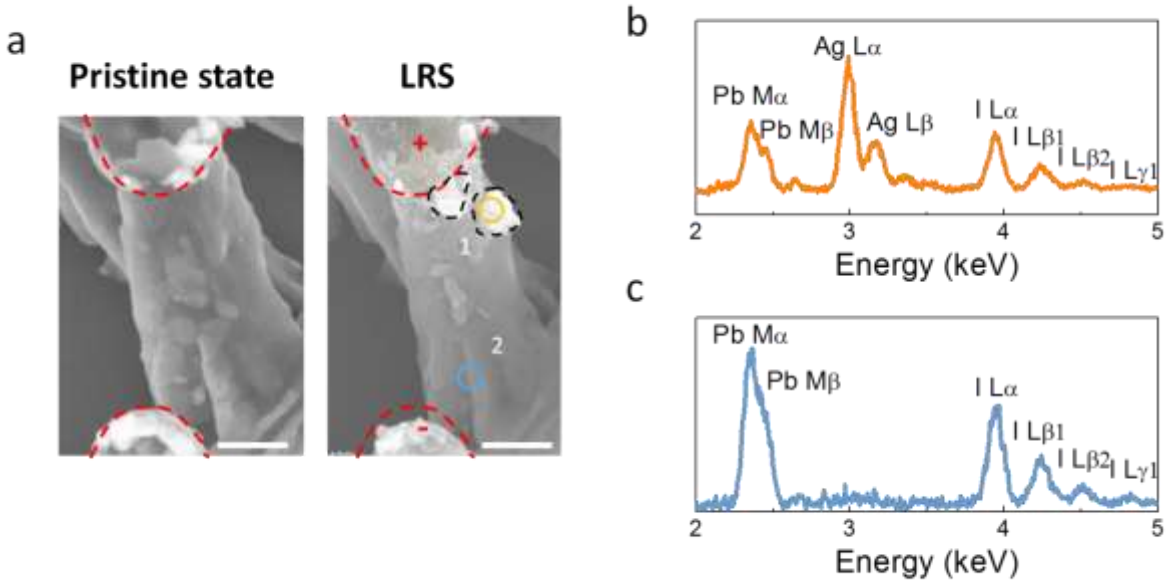


Figure S7. a) SEM images of a device at pristine state and LRS (~30 Ω) after being programmed with high compliamce current I_{cc} = 8 mA. The red and black dashed lines mark the locations of the Ag electrodes and the formed Ag clusters, respectively. Scale bar: 600 nm. b,c) EDS spectra collected at (b) location 1 (orange circle) and (c) location 2 (blue circle) marked in (a).

We found that when the device was programmed under a high SET compliance current (>=8 mA), clusters formed near the anode can be occasionally observed. The spectrum collected at location 1 (Figure S7b) showed pronounced Ag characteristic X-ray peaks, suggesting that Ag migrated into $MAPbI_3$ film and formed Ag clusters. On the other hand, from the SEM image, one can see that these Ag clusters only accumulated near the anode and did not form filaments throughout the whole film. This observation was further supported by the absence of Ag characteristic X-ray peaks in the EDS spectrum collected at location 2 near the cathode (Figure S7c).

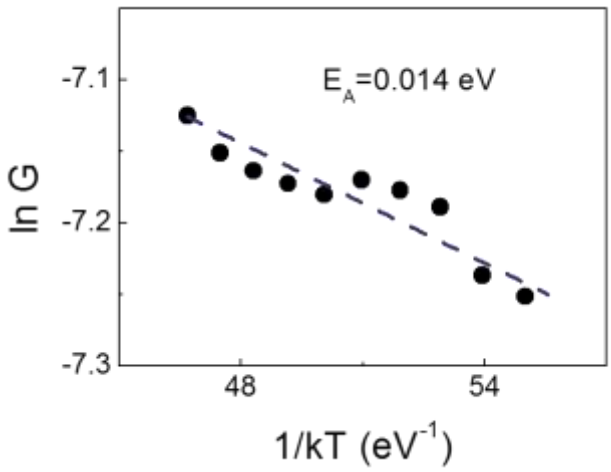


Figure S8. Temperature dependence of the Ag/$MAPbI_3$/Au device conductance at LRS, showing a thermal-activation behavior.

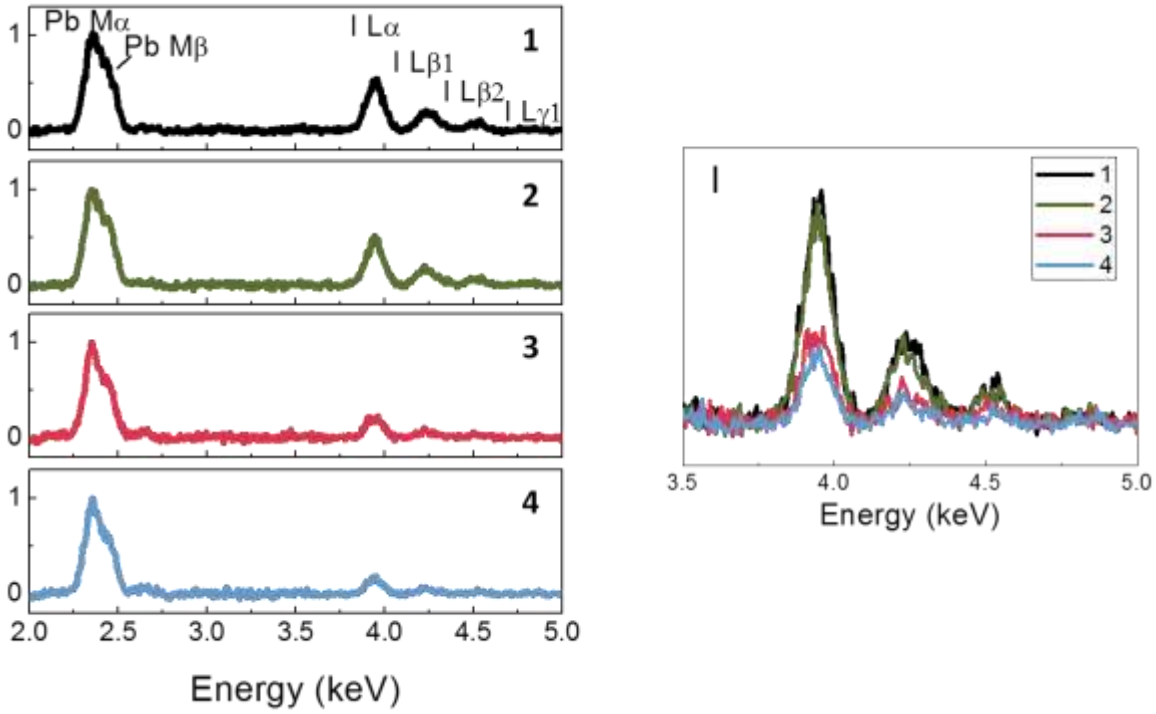


Figure S9. Left: EDX spectra of the device at HRS, showing the main characteristic X-ray peaks of Pb (M series) and I (L series) collected at locations 1-4 in Figure 2(b). The spectra have been normalized against the intensity of the characteristic Pb M series peaks. Right: Comparison of the I peaks at locations 1-4.

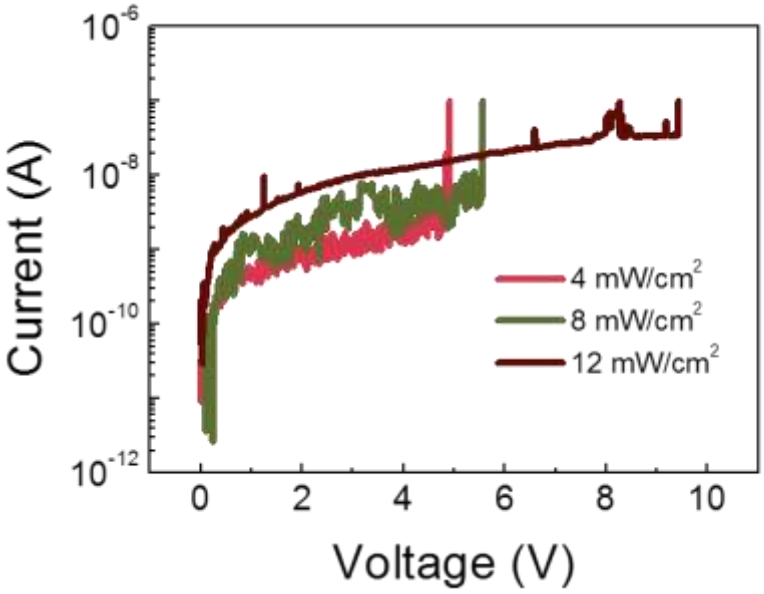


Figure S10. *I-V* characteristics showing SET processes of a Ag/$MAPbI_3$/Au planar structure device measured under laser light (525 nm) illumination at intensity of 4 mW/cm^2, 8 mW/cm^2 and 12 mW/cm^2 respectively. Higher voltages are required to form the device as the light illumination intensity is increased.

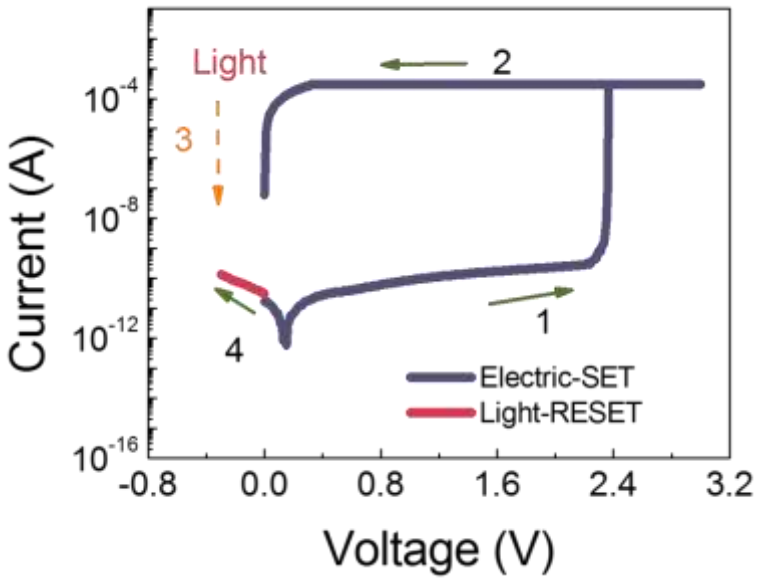


Figure S11. *I-V* characteristics of a Ag/$MAPbI_3$/Au planar structure device demonstrating electric-SET and light-RESET. The device was electrically switched to the LRS in the dark at first. After being exposed to laser illumination (4 mw/cm^2, 525 nm) for a few seconds (step 3), the device switched to the HRS, as reflected in subsequent sweep (step 4).