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Supporting Information

To probe the performance of perovskite memory device: defects property and hysteresis

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Experimental

- Chemicals: The commercial chemical materials used are listed as follows: acetone (AR Beijing Chemical Works), ethanol (AR Beijing Chemical Works), isopropanol (99.99%, Aladdin Industrial Corporation), TiAc₂ (75 wt% in isopropanol, Sigma-Aldrich), Aminomethane (CP Beijing Chemical Works), Hydrogen iodide (57%, Alfa Aesar), Pbl₂ (99.999%, Sigma-Aldrich), N,N-dimethylformamide (99.99%, Sigma-Aldrich), dimethyl sulfoxide (AR Beijing Chemical Works), spiro-OMeTAD (Lumtec), and ITO substrates.
- 2. Preparation of perovskite solution:
- For perovskite of one-step devices, the Methylammonium iodide (MAI) and lead (II) iodide (PbI₂) at a 1:3 mass ratio were dissolved in N, N-dimethylformamide (DMF) at a concentration of 1070 mg/mL or in a mixture of DMF and DMSO (9:1 v/v) at a concentration of 881 mg/mL.
- 2) For perovskite of two-step devices, the MAI (MACI) or formamidine iodide (FAI) was dissolved in isopropanol (IPA) at a concentration of 50 or 60 mg/mL and PbI₂ was dissolved in DMF at a concentration of 450 mg/mL (or in a mixture of DMF and ACN (11:1 v/v) at a concentration of 458 mg/mL) or dissolved with caesium iodide (CsI) (10:1 m/m) in DMF at a concentration of 495 mg/mL.
- 3. Memory device fabrication:

The ITO substrate was sequentially washed with distilled water, ethanol, acetone and isopropanol. Then treated using UV/O_3 for 30 min.

- For perovskite of one-step devices, the solution was spin-coated on the ITO at 2000 rpm for 30 s or at 4000 rpm for 20 s. After a delay time, ether was quickly dropped onto the center of the substrate during spin-coating. The obtained films were annealed at 100 °C for 5 min. Finally, the device was finished by thermal evaporation of MoO₃ (50 nm) and Ag (150 nm) under vacuum.
- 2) For perovskite of two-step devices, the Pbl₂ (with ACN or Cs) was spin coated on ITO with 3000 rpm for 30 s and annealed at 75°C (or 110 °C) for 15 min in nitrogen glovebox. After the Pbl₂ coated substrates cooling to room temperature (25 °C) in nitrogen glovebox, MAI or FAI was spin coated with 3000 rpm for 30 s and annealed at 150

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°C for 15 min or 170 °C for 20 min in air. Finally, the device was finished by thermal evaporation of MoO_3 (50 nm) and Au (100 nm) or Ag (150 nm) under vacuum.

4. Solar cell device fabrication

The ITO substrate was sequentially washed with distilled water, ethanol, acetone and isopropanol. Then treated using UV/O_3 for 30 min. The ETL was subsequently coated on ITO substrate with the Titanium diisopropoxide bis (acetylacetonate) stabilized TiO₂ nanocrystal solution, and annealed at 150 °C for 30 min in air. Then the perovskite films fabrication is the same as memory device fabrication. But before the thermal evaporation, the HTM solution was deposited by spin coating at 3000 rpm for 30 s. The HTM solution, spiro-OMeTAD, doped with 4-tert-butylpyridine and lithium bis (trifluoromethylsulphonyl) imide in ACN in chlorobenzene. Finally, the device was finished by thermal evaporation of MoO₃ (10 nm) and Ag (150 nm) under vacuum.



Fig.S1 Current-Voltage (J-V) characteristics of memory device without MoO₃



Fig.S2 I-V characteristics with a compliance current of 0.01A



Fig.S3 I-V characteristics under the voltage sweep of $0 \rightarrow 1 \rightarrow 0 \rightarrow -1 \rightarrow 0$



Fig.S4 Retention performance of memory device at 0.1V at room temperature



Fig.S5 Endurance of the perovskite with MoO₃ interlayer after 240 hours





Fig.S6 The preparation of perovskite films from different deposition methods: (a) One-step solution process based on the mixture of PbI₂ and MAI. (b) Two-step solution process based on sequential coating of PbI₂ (ACN/Cs) and MAI or FAI.

Table S1. Photovoltaic parameters of the devices obtained at reverse scan (RS) and forward scan (FS)

Device No.	Sweep direction	Voc (V)	Jsc (mA/cm²)	FF	PCE (%)	H-Index
One-step	RS	1.064	19.48	62.99	13.04	0.38
	FS	0.927	19.15	46.72	8.28	
One-step DMSO	RS	1.006	21.70	60.54	13.22	0.27
	FS	0.970	21.66	42.83	9.00	
Two-step	RS	1.044	20.30	67.54	14.35	0.14
	FS	0.976	20.39	61.16	12.20	
Two-step ACN	RS	0.812	21.22	65.88	11.36	0.25
	FS	0.762	21.27	53.72	8.73	
Two-step FA _{0.9} Cs _{0.1} PbI ₃	RS	0.904	22.09	68.71	13.72	0.08
	FS	0.858	22.07	65.70	12.44	



Fig. S7 I-V characteristics of different devices. (a) ITO/CH₃NH₃PbI₃(CI)/MoO₃/Au (Two-step) (b)ITO/CH₃NH₃PbI₃/MoO₃/Ag (One-step) (c) ITO/CH₃NH₃PbI₃/MoO₃/Ag (One-step, with DMSO) (d) ITO/CH₃NH₃PbI₃(CI)/MoO₃/Ag (Two-step) (e) ITO/CH₃NH₃PbI₃(CI)/MoO₃/Ag (Two-step, with ACN) (f) ITO/FA_{0.9}Cs_{0.1}PbI₃/MoO₃/Ag (Two-step).



Fig. S8 I-V characteristics of devices with four different areas of Ag electrodes. "1" in the legend represents the device with primary area of 0.102 cm2. "0.75", "0.5" and "0.25" in the legend represent the device with 75%, 50% and 25% of the primary area, respectively.

At ON state (after SET), the device exhibits larger current by increasing the area of Ag electrode, indicating its lower resistance at ON state, as shown in Fig. S8. This means that the resistance is dependent on the device active area, which is consistent with the reported filamentary conduction mechanism.⁹



Fig. S9 ON/OFF ratio of the memory device based on one-step process with different metal anode (left: Au, right: Ag)