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In-situ Observation of Out-of-plane Switching Filament in 2D Halide $(PbI_2)_{1-x}(BiI_3)_x$ Memristor Under Operando Biasing

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Memristive switching devices (i.e., memristor), voltage-induced modulating memresistance (M) with nonlinear bipolar I-V hysteresis, have been considered as next-generation computing and memory devices because conventional devices have been confronting obstacles due to physical challenge. [1-2] The memresistance alteration between a high resistive state (HRS) and a low resistive state (LRS) is reversible so that the two digital states can be stored as binary code (0 or 1). The memristive behavior is observed in 2D metal chalcogenides semiconductors such as MoS_2, TaS_2, WS_2, $MoSe_2$, and WSe_2 through in-plane anion migration or local phase transition [3-5]. Herein, we introduce an Iodide-based memristor composed of the two-dimensional $(PbI_2)_{1-x}(BiI_3)_x$. As Bi content (x) increases, $(PbI_2)_{1-x}(BiI_3)_x$ structure exhibits brick wall-like phase separation (Pb-rich vs. Bi-rich). A "brick wall" motif of two phases (Pb-rich or Bi-rich) in Figures 1a-b generates extensive dislocation defects at the side phase boundaries due to mainly out-of-plane lattice mismatch. The memristive switching mechanism by migration of halogen ions (=Iodine) toward out-of-plane direction of 2D planes at the high Bi composition (>= 50 at%) depends on the defect concentration (i.e., degree of phase separation), because no switching is revealed at the lower degree of the interfacial defects at low Bi composition (Bi=20 at%) in Figure 1c. Reversible conducting Iodine filament under sweep biasing is unveiled using an in-situ electrical biasing TEM holder (Nanofactory) shown in Figure 2. Furthermore, we found that the integration between our Pb rich and Bi rich type phases affects the optical properties, where trapping of Bi in PbI2 like clusters changes bandgap. The observation that interfacial defect engineering can limit or boost ionic charge transport through phase boundaries toward the out-of-plane direction of halide-based memristors would pave the road to develop a brand-new memristor system. This work made use of the EPIC facility of Northwestern University's NUANCE Center, which has received support from the Soft and Hybrid Nanotechnology Experimental (SHyNE) Resource (NSF ECCS-1542205); the MRSEC program (NSF DMR-1720139) at the Materials Research Center; the International Institute for Nanotechnology (IIN); the Keck Foundation; and the State of Illinois, through the IIN.

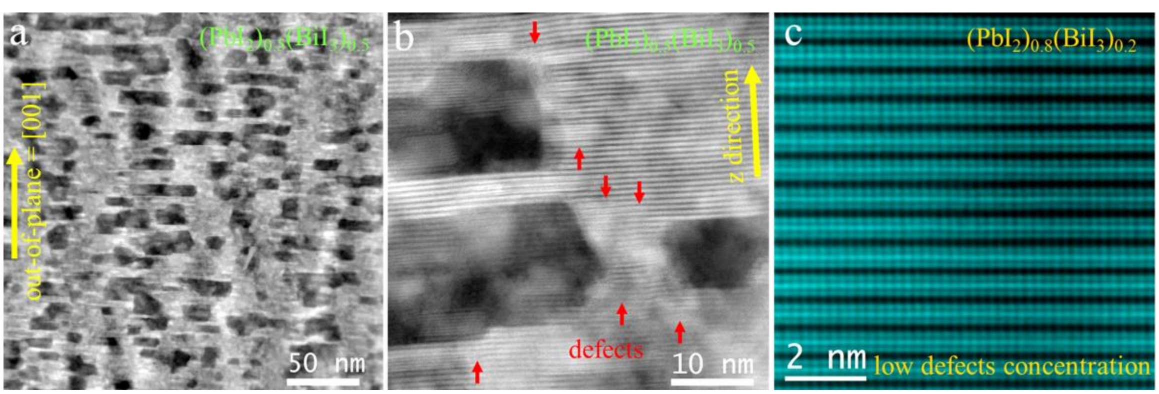

Figure 1. a and b. HAADF images of (PbI2)0.5(BiI3)0.5: Overall brick wall-like phase separation (Pb-rich and Bi-rich) in a, and interfacial defects in b (red arrows) at phase boundaries due to difference in their lattice parameters. c. Atomically resolved HAADF of (PbI2)0.8(BiI3)0.2 with low defects concentration. Please note that only a high concentration of interfacial defects shows memristive behavior.

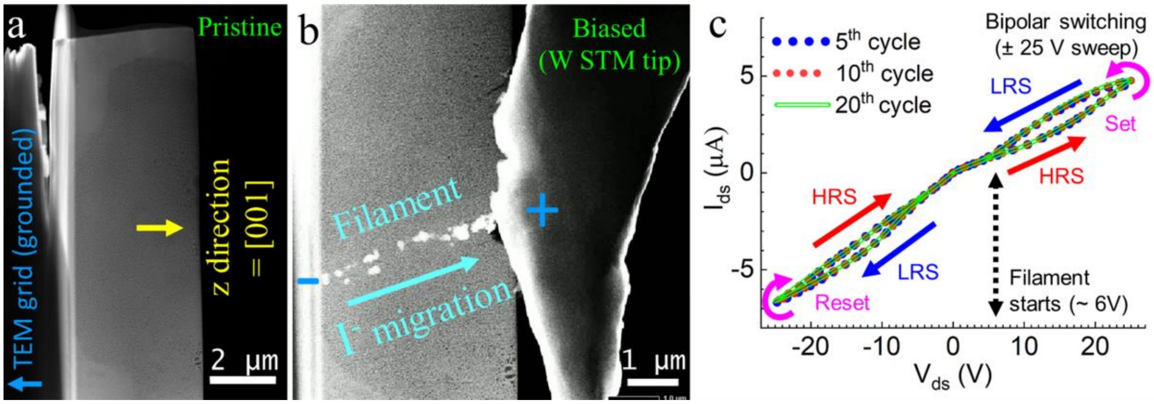

Figure 2. a and b. in-situ observation of filament formation through z-direction of (PbI2)0.5(BiI3)0.5 using an electrical biasing holder (Nanofactory). b. Preserved I-V hysteresis under ± 25 V cyclic sweeping showing bipolar memristive switching.

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