Full-Field Structural Microscopy Reveals Dynamic Film–Substrate Interactions in VO_2 Neuromorphic Devices

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Understanding how structural transitions govern neuromorphic functionality in quantum materials requires characterization tools that can probe local transformations in operando and across multiple length scales. We combine dark-field X-ray microscopy (DFXM) with complementary X-ray and electron microscopies to reveal the structural evolution of voltage-driven filaments in VO_2 memristive devices and their unexpected coupling to the underlying substrate. DFXM provides high-resolution, full-field, structure-selective imaging, enabling us to visualize rutile filament formation without destructive specimen preparation or slow rastering. We find [1] that rutile channels contain residual monoclinic clusters, revealing internal nonuniformity, and that rutile nucleation beneath electrodes precedes the growth of conductive pathways. Additionally, repeated voltage cycling induces a medium-term (<30 min) memory effect: specific sites in the device gap switch at lower voltages even after a brief thermal reset. Strikingly, we show [2] that these electronic/structural transformations in the VO_2 film are not mechanically isolated: filament formation generates strong, highly asymmetric strain fields that imprint deep into the Al_2O_3 substrate. This strain feeds back into the film, guiding subsequent filament expansion and redefining local switching dynamics. The observed film–substrate feedback mechanism expands the conventional view of epitaxial strain from a static constraint to an active, reconfigurable parameter during device operation. These results position DFXM as a powerful platform for operando studies of correlated oxides and point toward substrate engineering as an emerging route to control and functionalize neuromorphic architectures.

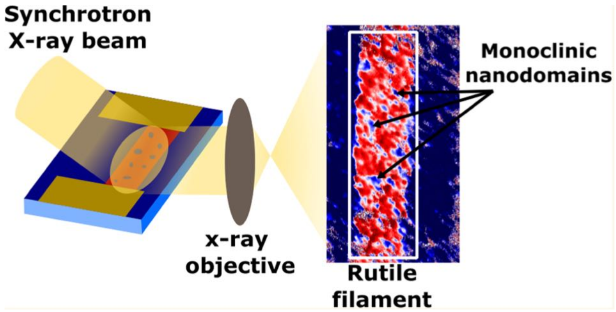

Figure 1: A schematic of DFXM imaging filamentary domains in a VO_2 neuromorphic device.

[1] E. Kisiel, et al. ACS Nano 19, 16, 15385–15394 (2025)
[2] E. Kisiel, et al., in review

Supplementary information

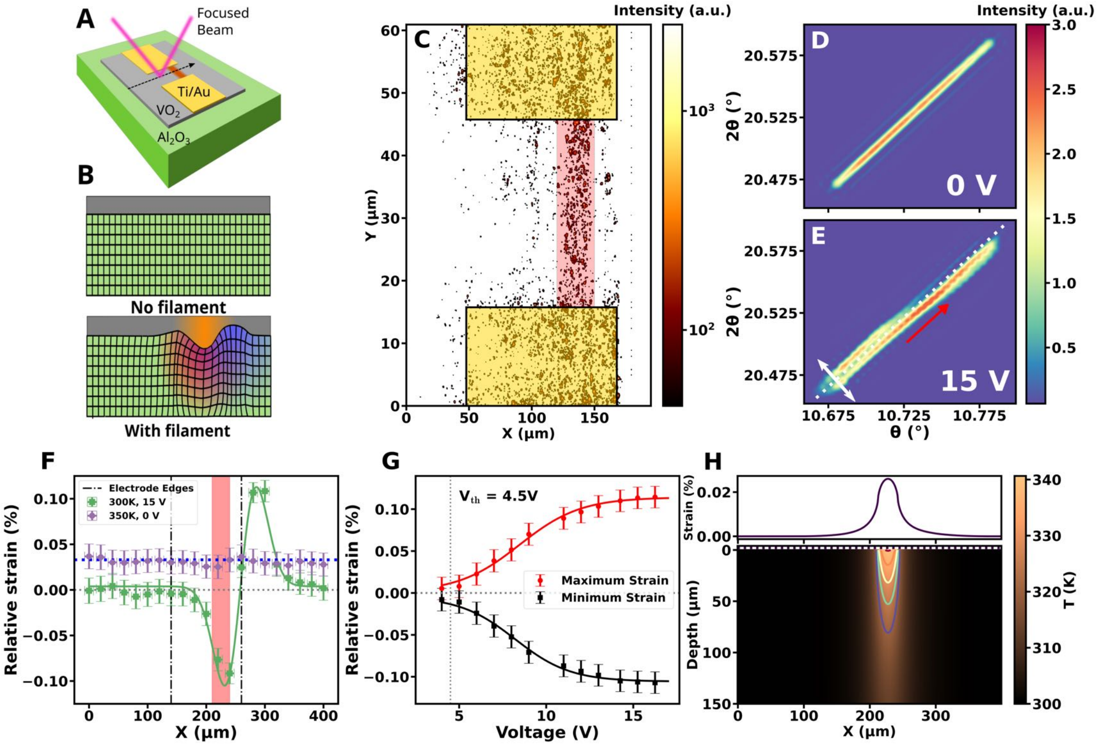

Fig. 2. Al_2O_3 substrate strain imprinted by the filament formation in the VO_2 film switching device. **A**, Schematic of the VO_2/Al_2O_3 device with microdiffraction scan direction (black arrow). **B**, Illustration of asymmetric substrate strain generated by the VO_2 filament. **C**, DFXM image of a voltage-induced rutile filament in VO_2; electrodes are gold outlines, filament in red. **D, E**, (012) Al_2O_3 substrate Bragg peak at 0 V (**D**) and 15 V (**E**) applied to the VO_2 film device. The peak at 15 V shows splitting and center of mass shift. The white line in the 15 V panel indicates the location of the 0 V peak. The white arrow indicates a splitting in the orientation while the red arrow indicates a shift in the center of mass. **F**, Measured strain profile in the Al_2O_3 substrate during the voltage-induced filament formation in the VO_2 film at 300 K (green line and symbols) and under equilibrium conditions at 350 K (*i.e.*, zero voltage) above the VO_2 film phase transition (purple line and symbols). The filament location is highlighted in red and the vertical dashed lines indicate the device electrode location. Unlike the spatially uniform phase transition in VO_2 at 350 K, voltage-induced localized phase transition in the film produces large asymmetric strain distribution in the underlying Al_2O_3 substrate. **G**, Voltage dependence of the asymmetric strain (peak and valley in red and black, respectively) in the Al_2O_3 substrate. The substrate strain development correlates with the threshold switching voltage, V_{th} = 4.5 V, of the VO_2 film device. **H**, Thermal simulations showing the substrate strain development induced by the VO_2 filament acting as a heat source. Contours in the bottom panel are in steps of 5 K from 320 K to 340 K. The simulated strain profile (top panel) has a drastically different magnitude and shape compared to the experimental observations in F, which indicates that the experimental results cannot be attributed to simple heating effects.