

## STEM Video of Electronically-Driven Metal-Insulator Transitions in Nanoscale NbO<sub>2</sub> Devices

William A. Hubbard<sup>1</sup>, Toyannath Joshi<sup>2</sup>, Pavel Borisov<sup>2</sup>, David Lederman<sup>2,3</sup>, and B. C. Regan<sup>1</sup>

<sup>1</sup> Department of Physics & Astronomy and California NanoSystems Institute, University of California, Los Angeles, CA 90095, USA

<sup>2</sup> Department of Physics and Astronomy, West Virginia University, Morgantown, WV 26506, USA

<sup>3</sup> Department of Physics, University of California, Santa Cruz, CA 95064, USA

Above a characteristic temperature, some insulating materials, typically metal oxides, undergo a reversible transition into a low resistivity phase. Such metal-insulator transition (MIT) materials are being studied for possible applications in electronic devices, as this transition can be triggered locally via Joule-heating. Among the known MIT materials, niobium dioxide (NbO<sub>2</sub>) is of particular interest because it has a transition temperature (1081 K) that is above typical operating temperatures for semiconductor electronics applications.

In bulk NbO<sub>2</sub> the MIT occurs due to a crystallographic transformation between rutile and distorted rutile structure above and below the transition temperature, respectively [1]. In an NbO<sub>2</sub> thin film device under voltage bias, this transition produces a volatile and reversible low resistance state above a threshold voltage. Because of this property, NbO<sub>2</sub> has been suggested as a possible selector material in crossbar arrays of resistive memory elements as a method for preventing “sneak current” switching. Though several bulk heating and biasing studies have been performed, the MIT in nanoscale devices under local Joule heating is not well understood. Here we report on imaging the MIT *in situ* in nanoscale, horizontally-aligned Pt/NbO<sub>2</sub>/Pt devices with multi-detector scanning transmission electron microscope (STEM) imaging.

Our horizontal devices (Figure 1, left inset) allow TEM imaging of an MIT material between biasing electrodes. Ti/Pt (5/25 nm) electrodes (50 nm wide with 30 nm between electrodes) were fabricated on an electron transparent Si<sub>3</sub>N<sub>4</sub> membrane using electron beam lithography. 50 nm of NbO<sub>2</sub> was deposited over the electrodes via pulsed laser deposition at 700°C substrate temperature in 2 mTorr Ar+O<sub>2</sub> growth atmosphere with 1% O<sub>2</sub> content using an NbO<sub>2</sub> ceramic target (AJA International.). Film quality and thickness were assessed with x-ray photoelectron spectroscopy, x-ray diffraction, and x-ray reflectometry, confirming the major NbO<sub>2</sub> phase, which forms with an additional top Nb<sub>2</sub>O<sub>5</sub> layer of ~2 nm due to air exposure [2]. STEM video was obtained simultaneously from 3 different detectors, collecting electrons at low (bright field or BF), intermediate (dark field or DF), and higher scattering angles (high-angle dark field or HDF). Each detector is sensitive to different contrast mechanisms; diffraction contrast is more apparent at low and intermediate scattering angles while thickness and atomic number *Z* contrast dominate at higher angles.

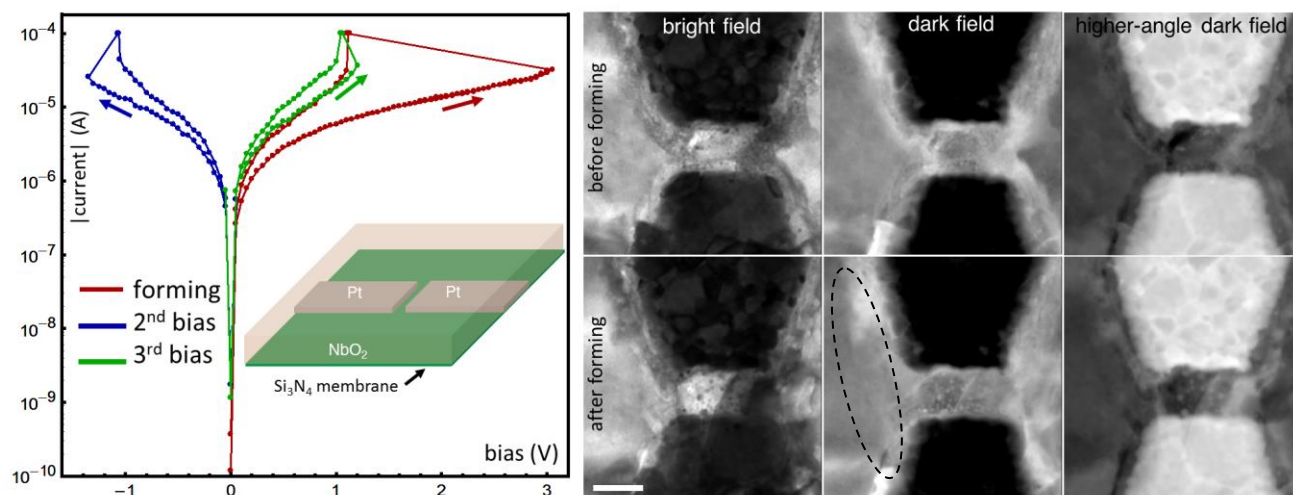
Current-voltage (IV) data acquired *in situ* show an electroforming step and two subsequent transition cycles (Figure 1, left). The bias voltage was ramped slowly (~50 mV/s) and the current was limited to 40 μA. After an initial electroforming step, the device exhibited reversible, polarity-independent threshold switching at ~1V. Such switching repeated over multiple cycles. We observed similar behavior in several devices biased *in situ*. Despite our non-standard geometry, the devices switched with an electric field of ~3x10<sup>7</sup> V/m and ~1 nW/nm<sup>3</sup> power density, values comparable to those of vertical

Pt/NbO<sub>2</sub>/Pt devices [4]. The hysteresis in the IV curves is evidence of the expected MIT switching behavior.

The structure of the Pt/NbO<sub>2</sub>/Pt device changed noticeably during the forming step (Figure 1, right). In each detector the electrode geometry appears unchanged by the transition, ruling out the formation of a conducting channel through mass transport of electrode material [3]. In the BF and DF channels, significant NbO<sub>2</sub> grain reconfiguration can be seen between the electrodes. In the BF image a ~50 nm wide dark grain appears overlapping with the lower contact, and is presumably situated in a plane above the electrodes. The DF channel shows that, while regions of NbO<sub>2</sub> far from the electrodes change very little during forming, several grains appear in the region running along the length of, and ~20 nm to the left of, the electrodes (encircled in Figure 1, right). This reconfiguration suggests that electroforming occurs in a region surrounding the electrodes, and not just between them, thus the forming transition is likely triggered thermally, rather than via field-driven processes. Subsequent MITs drive more subtle reconfigurations (STEM images not shown) of the NbO<sub>2</sub> grains in the regions affected by forming.

#### References:

- [1] A. Bolzan *et al*, Journal of Solid State Chemistry **113** (2105), p. 9-14
- [2] T. Joshi *et al*, Journal of Physics D: Applied Physics **48** (2015), p. 335308
- [3] W. A. Hubbard *et al*, Nano Letters **15** (2015), p. 3983-3987
- [4] M.D. Pickett, *et al*, Nanotechnology **23** (2012), p. 215202
- [5] This work has been supported by FAME, one of six centers of STARnet, a Semiconductor Research Corporation program sponsored by MARCO and DARPA. The authors acknowledge the use of instruments at the Electron Imaging Center for NanoMachines supported by NIH 1S10RR23057 and the CNSI at UCLA, as well as the Shared Research Facilities at WVU.



**Figure 1.** (left) Current vs bias voltage plot for a forming and two subsequent transition cycles for a device biased *in situ*. The colored arrows indicate the point ordering in each cycle. Inset: device schematic. (right) STEM images from 3 different detectors showing the device before (upper row) and after (lower row) forming. The dashed oval encircles features outside the expected switching region that changed significantly during electroforming. Scale bar is 25 nm.