In-situ electron microscopy study of non-volatile resistive switching in Mott insulator VO₂

Shaobo Cheng¹, Min-Han Lee², Xing Li³, Lorenzo Fratino⁴, Marcelo Rozenberg⁴, Ivan Schuller⁵ and Yimei Zhu⁶

¹Department of Condensed Matter Physics and Materials Science, Brookhaven National Laboratory, Upton, NY, USA, ²University of California-San Diego, United States, ³Key Laboratory of Material Physics, School of Physics and Microelectronics, Zhengzhou University, Zhengzhou, Henan, P. R. China, United States, ⁴Laboratoire de Physique des Solides, CNRS, Université Paris-Sud, Université Paris-Saclay, Orsay, France, United States, ⁵Materials Science and Engineering Program, University of California-San Diego, La Jolla, CA, USA, United States, ⁶Department of Condensed Matter Physics and Materials Science, Brookhaven National Laboratory, Upton, NY, USA, United States

In-situ transmission electron microscopy (TEM) has been widely used for resolving the nanoscale changes (both structurally and electronically) on the functional devices. [1-3] VO₂, a typical Mott insulator, has near room temperature metal-insulator-transition (MIT), which has great potential in the applications for electronic devices. [4,5] The MIT is the result of the rutile-monoclinic structure transition and features volatile resistive switching. It is intuitive to wonder whether the nonvolatile resistive switching behavior can be triggered within the same system as well.

In this work, both ex-situ device measurements and in-situ biasing TEM studies have been combined to show the nonvolatile switching and recovery behaviors in VO₂ based device.[6] As shown in Fig.1, a 150 nm VO₂ thin film is sandwiched by Au top electrodes and a Ga-doped Ge conductive bottom electrode, whose configuration will benefit the future 3D crossbar-shape circuits. By using the ex-situ device measurements, different resistive switching behaviors can be realized under different temperatures. The volatile switching occurs near the phase transition temperature, while the non-volatile one is realized at lower temperature.

In-situ biasing TEM experimental results reveal the fact that the applied bias can not only induces the phase transition via Joule heating, but also triggers the electroforming process. The competition between Joule heating and electroforming process varies with the temperature. As shown in Fig. 2, a new metallic Magnéli phase V₅O₉ has been created by electroforming process and forms new conductive filament. The reversibility of this conductive filament was also verified by annealing the sample in oxygen environment, which helps to (partially) relax the V₅O₉ conductive filament. The change of transportation property was simultaneously measured and results are shown in Fig. 2(f). With partially relaxed conductive filament, a smaller voltage is needed to recreate the V₅O₉ conductive pathway in the second measurement. It is worthwhile to mention that, unlike the TiO₂ system, [3] the reset process of non-volatile resistive switching cannot be realized in VO₂ system due to the fact monoclinic-to-rutile phase transition will be triggered under large electric current.
In conclusion, we identify, for the first time, the structure (Magnéli phase $V_5O_9$) of the conductive filament during the electroforming process using in-situ microscopy and the mechanism of the non-volatile switching in $VO_2$. We have demonstrated that two different resistive switching behaviors can be realized using the $VO_2$/Ge resistive switching device, which is of great interest to emulate both neuronal and synaptic behaviors for the future neuromorphic computing on the same system.

Figure 1. Ex-situ device measurement for the Au/VO$_2$/Ge system. (a) The out-of-plane device configuration for transportation measurement. Au patterns are deposited and used as top electrode. (b) Resistance vs temperature curve showing near four order change in resistance across the metal-insulator transition on pristine VO$_2$ sample. (c) The typical volatile resistive switching behavior measured at 330 K. (d) Measured nonvolatile resistive switching behavior for the same device under 300 K.

Figure 2. In situ biasing TEM studies on Au/VO$_2$/Ge out-of-plane device at room temperature. (a) Low magnification HAADF-STEM image showing the experimental setup. (b) Selected area electron diffraction pattern acquired after applying bias. Besides the diffraction spots from VO$_2$, the V$_5$O$_9$ phase can be identified. (c) The simulated electron diffraction pattern of combined VO$_2$ & V$_5$O$_9$ phases. (d) Dark field TEM image showing the formation of V$_5$O$_9$ phase conductive filament after applying biasing using the V$_5$O$_9$ (001) diffraction spot. (e) Schematic model of the V$_5$O$_9$ filament. (f) The simultaneously acquired I-V curves in biasing TEM experiments. The second measurement was conducted after partially...
removing the Magnéli phase V5O9 in Oxygen. The embedded schematic diagrams showing the evolution of the conductive filament.

References
[7] This work is supported by the Quantum Materials for Energy Efficient Neuromorphic Computing, an Energy Frontier Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under Grant No. DE-SC0019273. Electron microscopy work at BNL was supported by the U.S. Department of Energy, Office of Basic Energy Science, Division of Materials Science and Engineering, under Contract No. DE-SC0012704.