

Uncovering switching and failure mechanism in memristive devices by *in-situ* spectromicroscopy

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It is generally assumed that voltage-driven oxygen-ion migration and the resulting nanoscale redox processes drive the resistance change in transition metal oxide based memristive devices. Direct observation of the switching and failure mechanism, however, remain challenging because the net changes of structure, stoichiometry, and valence state during switching are very small and occur primarily at electrode interfaces or within nanoscale filaments.

Here we will present local changes in the chemical and electronic structure of SrTiO₃-based memristive devices utilizing high-resolution *operando* characterization tools like transmission electron microscopy (TEM) and photoemission electron microscopy (PEEM). We chose SrTiO₃ as a single crystalline model material, which offers a well-understood platform and well-characterized spectroscopic signatures.

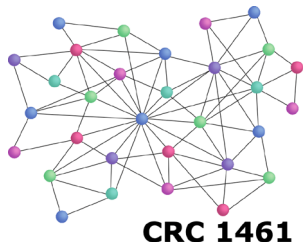
To overcome the surface sensitivity typically limiting PEEM investigations of memristive devices, we either delaminated the top electrode in-situ [1] or employed photoelectron-transparent graphene top electrodes were used to attain spectroscopic information from the buried SrTiO₃ layer [2]. We could thereby determine the position of in-gap states within the SrTiO₃ filaments and use it as input for our calculations of the electronic transport [3]. Quantitative maps of the oxygen vacancy concentration obtained during *in situ* switching confirm that localized oxygen evolution and reincorporation reactions rather than purely internal movement of oxygen vacancies cause the resistance change. A remarkable agreement between experimental quantification of the redox state and device simulation reveals that changes in oxygen vacancy concentration by a factor of 2 at electrode-oxide interfaces cause a modulation of the effective Schottky barrier and lead to >2 orders of magnitude change in device resistance [2].

Moreover, in-situ PEEM analysis enabled us to identify the microscopic origin of retention failure in SrTiO₃ devices [1] and to reveal two different mechanisms for the cycle-to-cycle variability of our devices, namely the change of the shape of the filaments and the movement of filaments during cycling [4].

(References on page 2)

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