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In-situ TEM Observations of Resistance Switching in Strontium Titanate Devices

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Strontium Titanate ($SrTiO_3$) is a prototype perovskite oxide, with applications in photonics, neuromorphic computing and as a dielectric. A particular important property of $SrTiO_3$, is room temperature ferroelectricity, which can be driven by the incorporation of lattice strain and an overall distortion of the lattice, that generate dipole moments polarizing the local nanostructure [1]. These materials have also received a great deal of scientific attention due to the resistive switching (RS) phenomena [2], which is used in resistive random-access memory (RRAM) [3]. The resistive switching mechanisms have been widely investigated in oxides [4, 5] and two main physical theories involving atomic defects have been proposed to explain the switching behavior [6]. The first proposed electronic mechanism is based on charge-transfer processes, including field and impact ionization of traps and refilling by electron injection. The second theory is based on diffusion of atoms/ions assuming formation and breaking of conductive filaments through the insulator storage medium in mechanism that is comparable to [4], which is hardly reproducible as the breakdown of a dielectric, is stochastic by nature.

In this study, we address atomic scale investigations of $SrTiO_3$ by combining high-resolution and analytical (S)TEM with *in-situ* TEM to obtain a comprehensive view of structure, electronical properties and compositions. Dynamic measurements using *in-situ* TEM based on electrical biasing and heating, allow the configuration of specific defects and related switching dynamics to be correlated with the presence of polar domains, the charge sate of defects and the defect level presence in the bandgap. This will give the unique opportunity to study ferroelectric switching phenomena, such as, domain wall nucleation, defects and ferroelectric boundaries.

Recent experimental work of our group showed that $SrTiO_3$ with a Sr deficiency of up to 20% could be grown as a single crystalline by metal organic vapor phase epitaxy (MOVPE). Samples with high off-stoichiometry showed forming-less resistive switching with on/off ratios as high as 10^3 without a forming step [7]. Findings by EELS and HRTEM show evidence of the presence of Ti antisite defects and suggest these antisite defects as a source for the resistive switching. This can be explained by the fact that the antisite defects of Ti on Sr sites (Ti_{Sr}) results in an off-centre defect that causes a ferroelectric polarization that can be switched between two different states. Figure 1 shows the structural changes of the device recorded in the dark-field (DF) imaging mode, using the (001) forbidden reflection. It clearly shows the presence of nanopolar domains in the off-stoichiometric film caused by sweeping the voltage from -3 to 3V using *in-situ* TEM. The changes of the contrast in the off-stoichiometric $SrTiO_3$ film grown on the $SrTiO_3$ substrate were visible above a voltage of (-/+) 2V. We assign these contrast inversions to a switching of the domain polarization by the applied electric field. When reducing the voltage to 0V, the system stays in a stable state, but the polarization can be erased, with the local film reversing back to a high-resistance state.

Our *in-situ* I-V measurements are in excellent agreement with *ex-situ* I-V curves and confirm our recent findings that resistive switching in Sr deficient $SrTiO_3$ thin films is observed at substantially lower voltages than those required to form filaments. Our hypothesis is that the resistive switching is based on trap assisted tunneling and ferroelectric switching. To confirm this hypothesis and to also see how the increase in the defect concentration will affect individual dipoles by possible interactions between them to form stable nanopolar domains, more advanced atomic investigations are ongoing and will be presented.

References:

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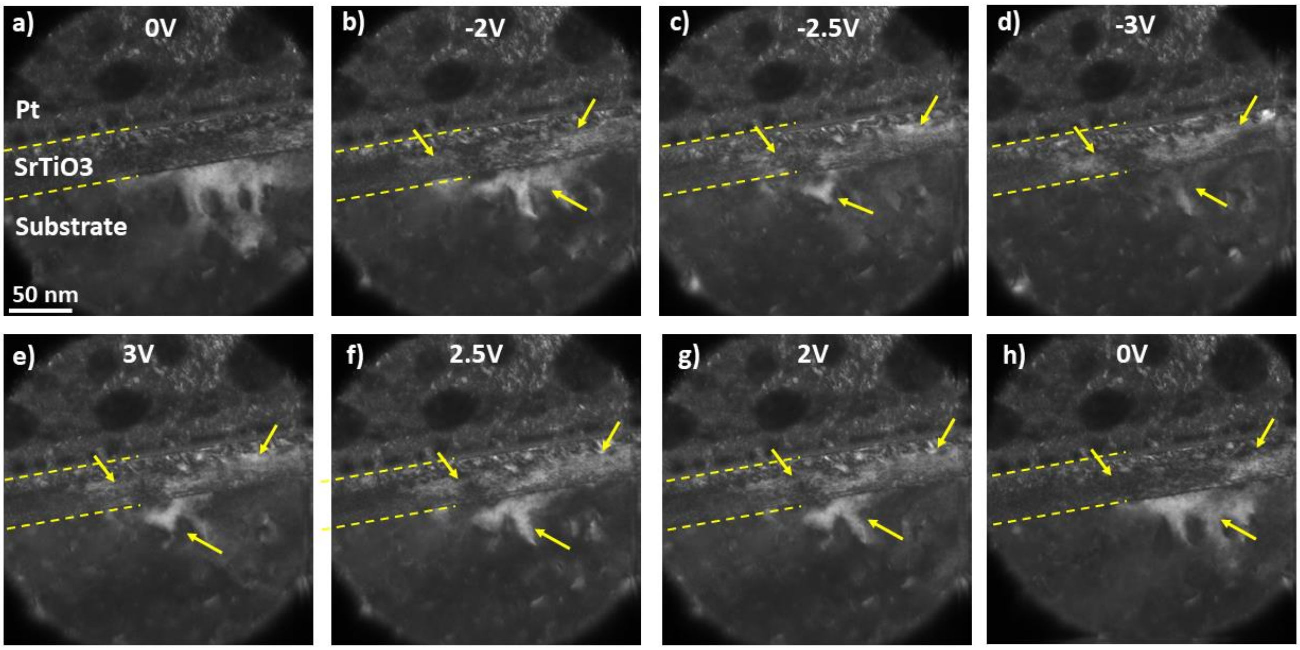

Figure 1. (a–h) Snapshots of I-V sweep on the device from 0 to -3V and from 3V to 0V, recorded in the same dark-field conditions. (a) and (h) pristine state at 0V, (b-d) applying negative bias involving changes in structure, due to coupling between the current and the internal domain polarizations. The yellow arrows indicating the area where the changes happened. (e-g) applying positive bias, showing relatively same switching behaviors.