Quantitative Atomic Scale Imaging of Conducting Filaments of Resistive Switching Memories Using Monochromated STEM-EELS Technique

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Redox-based resistive switching phenomena are found in many metal oxides and hold great promise for applications in next-generation memories and neuromorphic computing systems [1]. Such kind of resistive switching involves the formation and disruption of electrically conducting filaments through ion migration accompanied by local electrochemical redox reactions. Hence, the conducting filaments are the functional basis of the resistive switching. The underlying mechanism in the conducting filaments are often explained by point defects, but so far clear experimental evidence of such defects is missing. In this work [2], we apply STEM, EELS and EDX atomic-scale imaging techniques to quantitatively characterize nanosized conducting filaments in electroformed Au/FeSrTiO$_3$/Nb:SrTiO$_3$ thin film devices, providing direct experimental evidence for the point defect nature of the conducting filaments.

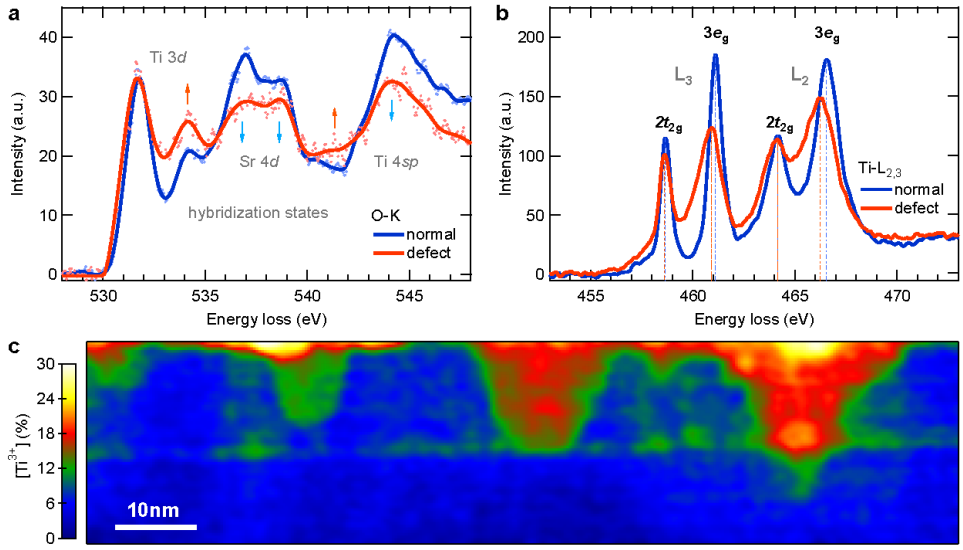

Fig. 1: a) and b) EELS fine structures for O-K and Ti $L_{2,3}$ edges, respectively. Blue line: normal $SrTiO_3$ dominated by Ti^{4+}, red line: defect $SrTiO_3$ with the highest amount of Ti^{3+} for the filaments. (c) Ti^{3+} distribution map.

$SrTi_{0.98}Fe_{0.02}O_3$ thin films about 17 nm in thickness were expitaxially grown on a 0.5-wt% Nb doped $SrTiO_3$ substrate by PLD. After electric forming by a sweep to +5 V to set the devices into the ON state, cross-sectional TEM specimens were cut by FIB milling. In both ABF and HAADF STEM cross-sectional images, we observed V-shaped areas showing darker contrast compared with their surroundings, indicating that these areas are conducting filaments viewed in cross-section. We obtained a Ti^{3+} distribution map by monochromated EELS spectrum imaging (Fig. 1), which clearly reveals four different V-shaped defect areas in the film developed to different stages. The shape of the filaments can be explained by the electric field and chemical gradients driving ion drift and chemical diffusion, respectively.

Quantitative EDX analysis indicates also considerable Sr- and O-vacancies in the filaments. By taking both Sr-deficiency and static lattice distortion into account, multislice STEM image simulations can qualitatively reproduce the decrease of mean intensity in both ABF and HAADF

images of the filaments. Intriguingly, atomic resolution STEM imaging reveals that the perovskite lattice is maintained intact at such high defect concentrations and considerable lattice distortions (Fig. 2).

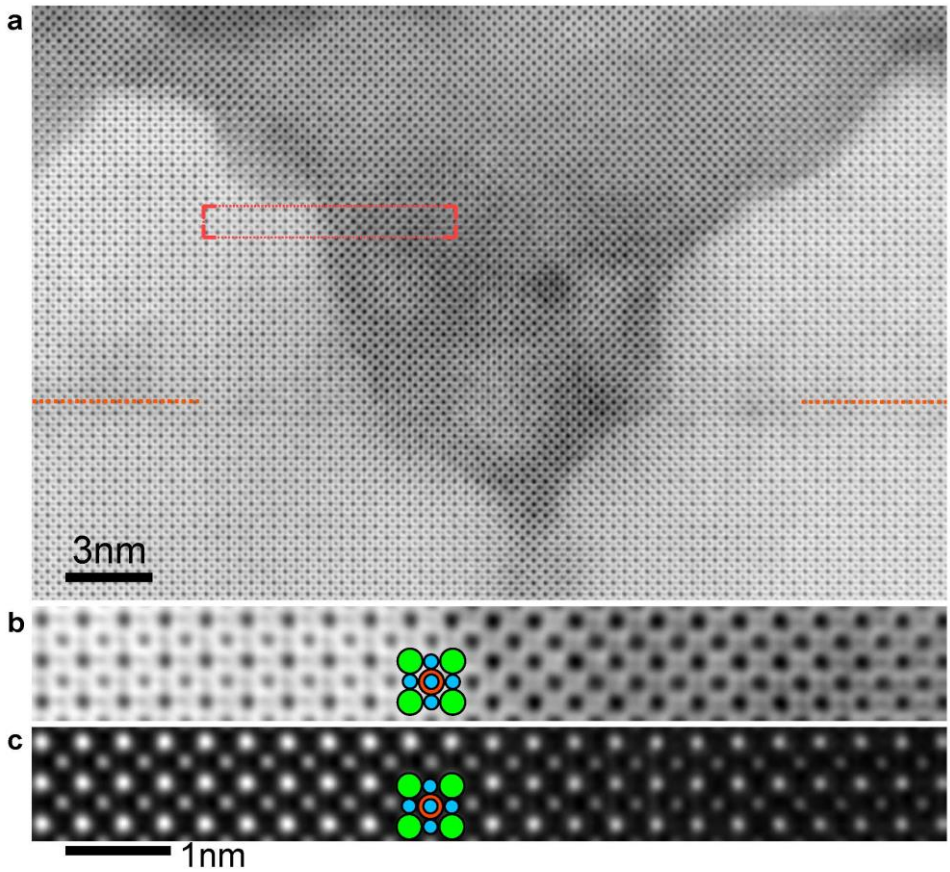

Fig. 2: a) ABF image of the region marked in Fig 1d. Lines denote the film/substrate interface. b) Magnified part marked in a). c) Corresponding HAADF image to b). Atomic symbol Sr: green, Ti: red, O: blue.

The present work [2], for the first time, provides clear experimental evidence for the existence of multiple nanosized conducting filaments formed solely by atomic-scale point defects in a $SrTiO_3$ based thin film device. First insights into the switching mechanism are deduced from a snapshot simultaneously showing multiple nanosized filaments in different evolutionary stages. The coexistence of a high Ti^{3+} concentration along with Sr- and O-vacancies in the conducting filaments provides atomic scale explanations for the resistive switching mechanisms. The results shed light on the complexity of the conducting filaments formation e.g. that cation and anion defects need to be considered jointly. The presence of multiple nanosized conducting filaments in a single device may provide a mechanism for multi-level resistive switching.

References and Acknowledgements

[1] R. Waser, R. Dittmann, G. Staikov, K. Szot, Redox-Based Resistive Switching Memories - Nanoionic Mechanisms, Prospects, and Challenges, Adv. Mater. 2009, 21, 2632 - 2663.

[2] H. Du, C.-L. Jia, A. Koehl, J. Barthel, R. Dittmann, R. Waser, and J. Mayer, Nanosized Conducting Filaments Formed by Atomic-Scale Defects in Redox-Based Resistive Switching Memories, Chem. Mater., 2017, 29, 3164 - 3173.

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