Towards an in-situ TEM based correlation of structural modifications and switching characteristics in filamentary type HfO_2-RRAM

Zintler, A.[1], Sharath, S.U.[1], Petzold, S.[1], Pivak, Y.[2], Alff, L.[1] and Molina-Luna, L.[1]

[1] Technische Universität Darmstadt, Germany, [2] DENSsolutions, Netherlands

Hafina based resistive random access memory (RRAM) devices, also known as memristors, are promising candidates as next generation non-volatile memory due to their potential for high-density, high-speed, ultimate scalability, low power consumption and proven compatibility to complementary metal-oxide-semiconductor (CMOS) technology [1]. According to the current state of understanding, the resistive switching behavior of hafnia based devices relies on the electric field driven formation and dissolution of oxygen deficient nanoscale conducting paths often discussed as "filaments". [2]. The physical mechanisms responsible are mainly governed by the motion of oxygen ions or vacancies, Joule heating induced by strongly localized currents and interfacial oxygen exchange processes. All these processes are strongly related to the intrinsic material properties which are defined by the local atomic structure.

In our recent electric field dependent in-situ TEM studies [3], we demonstrated for the first time how to electrically contact and operate a lamella fabricated in a focused ion beam (FIB) system from a resistive random access memory (RRAM) device based on a Pt/HfO_2/TiN metal-insulator-metal (MIM) structure (see figure 1a). The electrical switching characteristics of the electron-transparent lamella were comparable to a conventional reference device. The lamella structure was initially found to be in a low resistance state and could be reset progressively to higher resistance states by increasing the positive bias applied to the Pt anode. Shown in figure 1b is a high resolution high-angle annular dark-field scanning transmission electron microscopy imaging revealed a nanocrystalline oxide thin film structure. In order to understand the influence of the local atomic structure in the electrically measured resistive switching mechanism we investigated all critical device components. The key properties of a memristive device are highly dependent by the choice of the electrode material. In this study, we used a highly crystalline TiN bottom electrode grown on c-cut Al_2O_3. Complementary Electron backscatter diffraction (EBSD), TEM imaging and electron diffraction investigations of the bottom electrode revealed a highly textured microstructure consisting of quasi-symmetric (111) oriented grains with a 30° in-plane rotation to each other (see figures 3 and 4). Nucleation of a potential filament-like structures in the Hafnia thin film layer will be strongly dependent on oxygen exchange with the reactive electrode. The rupture point in the reset process is expected to be found at this interface. The capability to incorporate oxygen into the bottom electrode strongly depends on the local atomic structure. Recent studies suggest that the local defect ordering of the filament rupture point is the underlying mechanism for the quantum conduction model proposed for this material class [1]. Structural understanding of the bottom electrode, the hafnia layer and the corresponding interface provides an insight into the conditions necessary for possible filament formation and rupture.

References:

[1] S. U. Sharath *et al.*, "Control of Switching Modes and Conductance Quantization in Oxygen Engineered HfO_x based Memristive Devices," *Adv. Funct. Mater.*, vol. 27, no. 31, Jul. 2017.

[2] R. Waser, R. Dittmann, G. Staikov, and K. Szot, "Redox-Based Resistive Switching Memories - Nanoionic Mechanisms, Prospects, and Challenges," *Adv. Mater.*, vol. 21, no. 25 - 26, pp. 2632 - 2663, Jul. 2009.

[3] A. Zintler *et al.*, "FIB based fabrication of an operative Pt/HfO_2/TiN device for resistive switching inside a transmission electron microscope," *Ultramicroscopy*, vol. 181, pp. 144 - 149, Oct. 2017.

[4] The authors acknowledge financial support from the Deutsche Forschungsgemeinschaft (DFG) under research grant MO 3010/3-1.

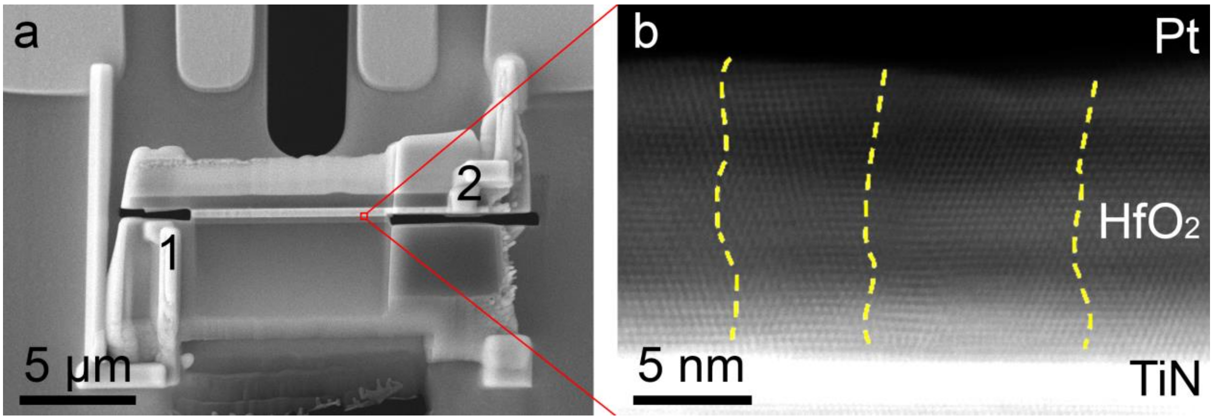

Figure 1: a. SEM image of a FIB prepared HfO_2 device on a MEMS based chip used for in-situ biasing TEM experiments, contacts for electric field application are denoted as 1 (bottom TiN electrode) and 2 (top Pt electrode), b. corresponding high resolution HAADF-STEM image of the HfO_2 layer and the corresponding electrode interfaces, boundaries between different orientation HfO_2 grains are marked.

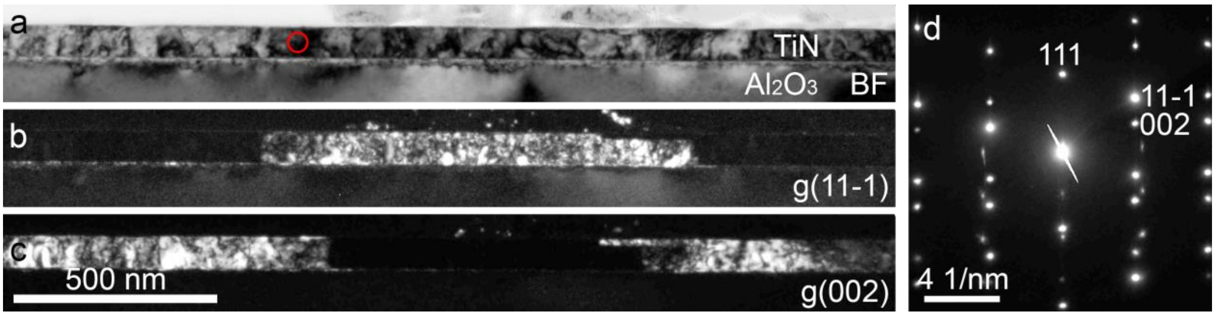

Figure 2: a. Bright-field cross sectional image of the TiN electrode/Al_2O_3 interface region, b. g(11"1) dark-field image of the same region, c. g(002) dark-field image of the same region, d. corresponding NBD pattern, origin depicted in a.

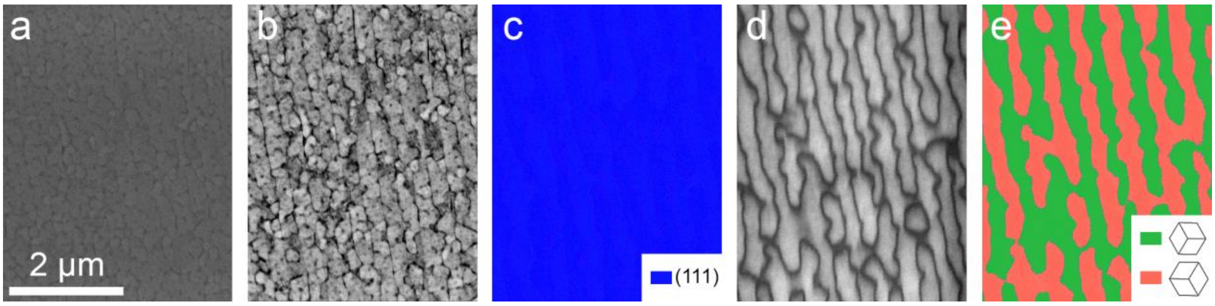

Figure 3: TiN electrode top view in a. secondary electron contrast, b. back scattered electron contrast, c. inverse pole figure (IPF) map of the (111) oriented TiN layer, d. image quality (IQ) map of (note boundary regions of quasi-symmetric grains with decreased quality factor), e. color coded map of the two possible orientations.