Spectroscopy in STEM/TEM

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Composition profiles across the metal-Nb_2O_5-metal stacks for resistive switching as studied by EDX and EELS

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Resistive switching in MIM (metal-insulator-metal) stacks is an effect that allows a promising technology which is able to overcome the size limitations of conventional non-volatile memories [1]. The resistive switching effect was already demonstrated for several transition metal oxides (TiO_2, NiO, HfO_2) [2]. The models of the switching mechanisms [3] suggest the important role of oxygen vacancies [4].

In this work Nb_2O_5 was used as switching insulator. An amorphous Nb_2O_5 layer was deposited on Pt bottom electrode by reactive dc magnetron sputtering from a metallic niobium target in an argon-oxygen atmosphere at room temperature [5]. In order to describe the effect of the oxygen vacancies on the switching mechanism, materials with different affinity to oxygen like Pt, Al and Ti were used for the top electrode. Cross sections for the transmission electron microscope (TEM) (figure 1) were prepared by focused ion beam (FIB).

Whereas a composition variation at the Nb_2O_5|Ti and Nb_2O_5|Al interfaces due to the oxidation of the electrodes (Ti, Al) could be determined both by energy dispersive X-ray spectroscopy (EDS) (figure 2) and by electron energy loss spectroscopy (EELS) (figure 3) in the scanning transmission electron microscope (STEM) the Nb_2O_5|Pt interface showed no changes in composition applying both methods. This behavior corresponds to electrical measurements indicating resistive switching for MIM stacks with Ti and Al top electrodes (figure 1).

Applying two different argon-oxygen ratios during the Nb_2O_5 deposition process [5] we are able to establish an intrinsic oxygen gradient in the niobium oxide layer between symmetric Pt electrodes. Using this approach we demonstrated resistive switching for Pt top electrodes, respectively. While the quantification of this intrinsic oxygen gradient by EDS failed, the complementary EELS solved this problem and shows a gradient in the oxygen K-edge intensity.

Using their different electron energy loss near edge structure (ELNES) different niobium oxides as well as titanium and aluminum oxides could be distinguished. Therefore a combination of experiment and FEFF simulation [6] is used to correlate switching mechanism and deviations in oxygen stoichiometry [7].

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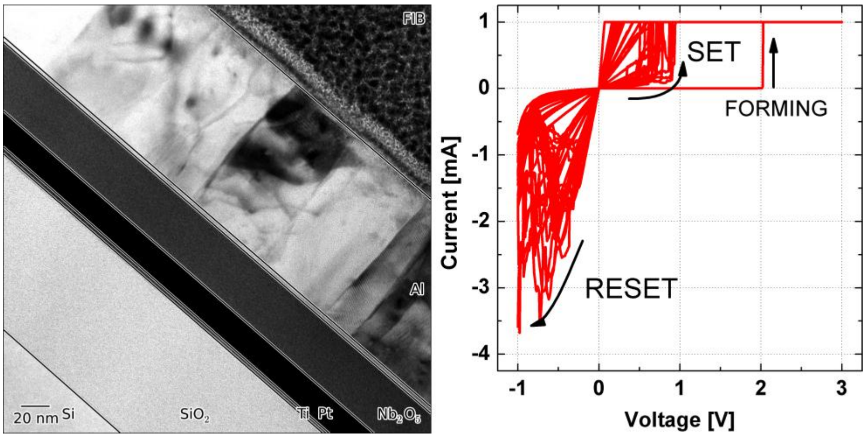

Figure 1. Bright field TEM micrograph of a Nb_2O_5 sample with Al top electrode (left). Current voltage characteristic of a similar stack design shows successful resistive switching (right).

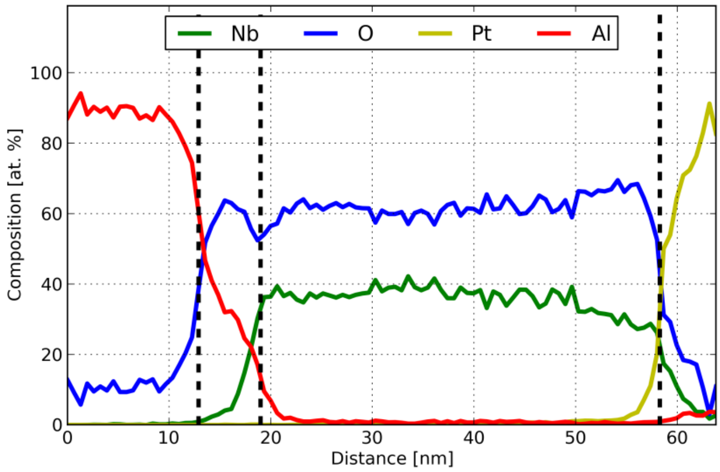

Figure 2. Development of the elementary composition across the MIM stack with Al top electrode obtained by EDS showing on one hand the aluminum oxide at the Nb_2O_5|Al interface and on the other hand the oxygen depletion in the Nb_2O_5 layer starting at this interface.

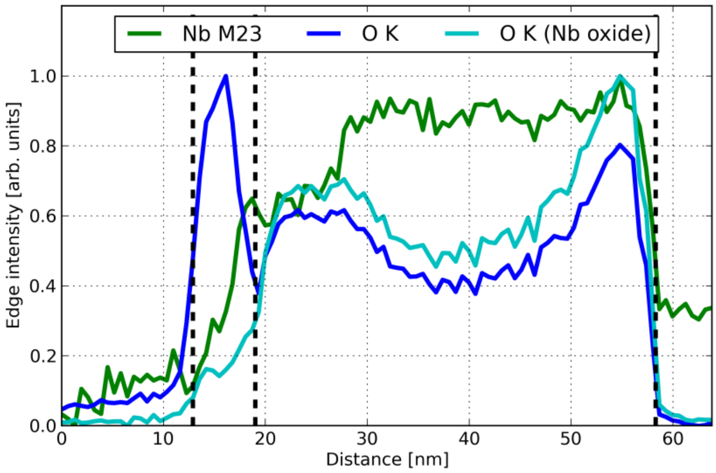

Figure 3. EELS line scan recorded parallel to EDS focusing on the Nb $M_{2,3}$ and the O K edges. Due to differences in edge shape as well as onset aluminum and niobium oxide are plotted separately. The total O K signal including both oxides shows a maximum value corresponding to the aluminum oxide whereas the O K intensity of niobium oxide drops within the Al oxide. Nb and O signal exhibit a similar value next to the Nb_2O_5|Al interface whereas away from this interface the Nb is constant while O content changes across the layer