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Probing and Mapping the Dynamics of Metal/Insulator Nanodomains Switching in V_2O_3 by Cryo-Spectromicroscopy Techniques

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V_2O_3 exhibits metal-to-insulator transitions (MITs) that can be activated under controlled external stimuli such as temperature (T), pressure, or chemical doping [1]. When cooled below 160 K, V2O3 undergoes a structural transition (from rhombohedral to monoclinic), associated with (i) a magnetic transition to an antiferromagnetic ordered state, (ii) a large volume change (+1.4%) and (iii) an IMT yielding a resistivity change of 7 orders of magnitude. This T-driven MIT has been extensively studied at the macroscopic scale (Fig.1a) [1], and remains still a perfect arena to probe *in situ* the V_2O_3 structural and electronic evolutions at the finest scale. More recently, the T-induced MIT has been mapped *in situ* by scanning photoemission spectroscopy [2], X-ray Linear Dichroism associated to PEEM [3] and nano-IR [4] revealing the microscopic electronic coexistence of insulator/metallic (I/M) domains, but with 25nm spatial resolution at best. However, this electronic phase separation may be due to local structural mechanisms, hence understanding the V_2O_3 electronic phase separation and its local mechanisms governing the I/M domains dynamics across the IMTs remains of key interests.

Here we performed *in situ* advanced monochromated STEM/EELS experiments on the NION CHROMATEM 200 MC for probing the relevant electronic excitations spectroscopic (from IR to soft X-ray) with an ultra-high EELS resolution at the sub-nm scale and below. We associate these instrumental capabilities with variable-T options under cryo-conditions using a double-tilt Henny Z cryo-holder (Figs.1a and 1b) using MEMS (Figs.1c and 1d) to vary continuously the temperature conditions from 120 K. Hence the T-activated V_2O_3 transition can be investigated to give a direct, simultaneous and local access to the structural and electronic evolution and correlate precisely the formation, nucleation, and dissolution of I/M nanodomains in V_2O_3. During low-temperature thermal cycling through the resistive transition, EELS spectra acquired in the low-loss regime present a characteristic plasmonic signature at 1.1eV only in the metallic phase (Fig.2a) [5]. At intermediate temperatures, the coexistence of I/M nanodomains was evidenced at the nanoscale (Fig. 2b and 2c). K-means clustering analyses confirm the electronic homogeneity of each domain (Fig. 2d). Upon heating, the electronic nanodomains switching can be observed dynamically over few degrees by following the propagation of the electronic domain wall which tends from insulating to metallic. In parallel, 4DSTEM nanodiffraction experiments also performed during the thermal cycles reveal the structural evolution of these I/M domains across the phase transition. These experiments aim at probing locally the mechanisms governing the transition and in particular the relation between the structural and electronic degrees of freedom.

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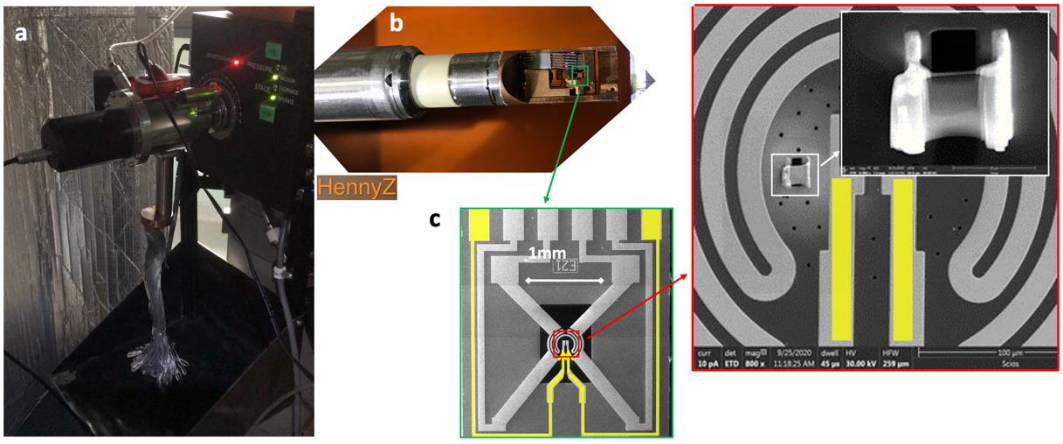

Figure 1. a. HennyZ double-tilt cryo-holder loaded in the NION CHROMATEM 200MC, b. tip of the double-tilt cryo-holder with a MEMS-based 6 contacts for heating and biasing options, c. and d. MEMS-based 6 contacts (biasing electrodes in yellow) with the V_2O_3 FIB lamella mounted in the heating area.

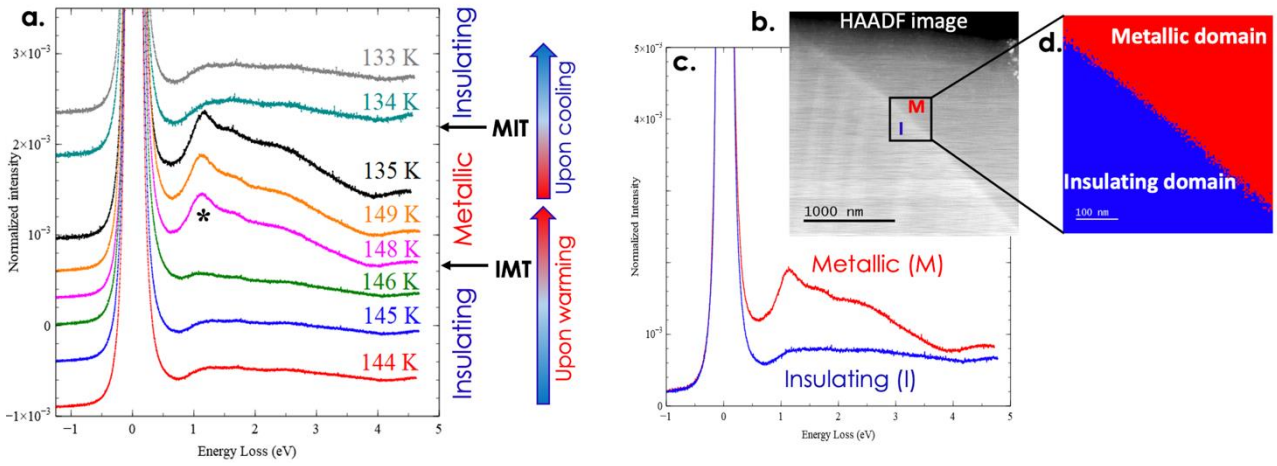

Figure 2. a. EELS spectra acquired every 1K across the resistive transition upon heating and cooling. * indicates the characteristic peak at 1.1eV only present in the metallic phase. b. STEM-HAADF image acquired at 147 K and c. the corresponding extracted EELS spectra from the region of interest (black square) at the I/M domain wall, d. electronic coexistence map of I/M domains at 147K; the reconstructed electronic map is obtained by applying the K-means clustering method

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