Phase coexistence and strain gradients during metal-insulator transition in VO_2 epitaxial films: a structural study

José Santiso[1]
Laura Rodríguez[1], Felip Sandiumenge[2], Carlos Frontera[2], Gustau Catalán[1]
[1]*Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and the Barcelona Institute of Science and Technology (BIST). Building ICN2, Campus UAB, 08193, Bellaterra, Barcelona, Spain*
[2]*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), 08193, Bellaterra, Catalonia, Spain*
jose.santiso@icn2.cat

VO_2 material exhibiting Metal-Insulator transition (MIT) slightly above room temperature (T_{MIT}=68ºC) has received much attention for the fundamental characteristics of the transition [1], as well as for the potential applications derived from the large variation of the IR optical transmittance [2], and in electronic devices [3]. VO_2 presents a rutile (R) tetragonal structure in its high-temperature metallic phase, while subtle changes turn into the low-temperature insulating monoclinic M1 phase. Other polymorphs like the monoclinic M2, or the triclinic T phase, have been reported to appear during transition depending on the mechanical strain, generally under uniaxial tensile strain along c_R-axis [4] or uniaxial compression along $[110]_R$ direction [5]. In this work we show an accurate *in-situ* X-ray diffraction study of epitaxial films of $VO_2/TiO_2(001)$ during transition, based in 3D reciprocal space mapping (Fig. 1). The study reveals the coexistence of low-temperature and high-temperature phases in an extended range of temperature. While the R phase remains epitaxially clamped to the substrate, the M1 phase suffers from a progressive distortion during transformation, following a defined trajectory in the reciprocal space. These distortions consist of a continuous range of tilt angles and different degrees of uniaxial compression, which transform into T-phase, and ultimately into M2 domains, contrary to the expected behaviour under the tensile stress imposed by the substrate. The study reveals that the homogeneous nucleation of the M1 domains in a R matrix during cooling follows common $(111)_R$ // $(210)/(2\text{-}10)_{M1}$ planes, which correspond to the lowest interfacial energies due to structural matching. This particular microstructure is explained in terms of a competition between the epitaxial stress, induced by the substrate, and the internal stresses developed upon the confinement of M1 domains during transformation of neighbouring R domains. Remnant M2 domains generate persistent strain gradients even at room temperature when the film has transformed into M1.

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FIGURES

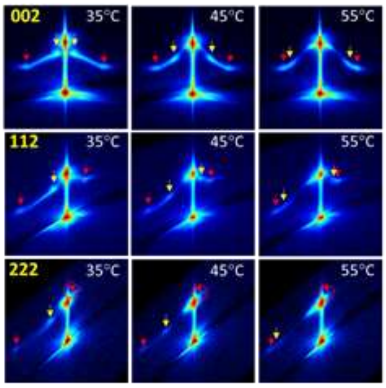

Figure 1: Projections of XRD 3D reciprocal space maps of 002, 112 and 222 reflections at varying temperatures upon transition. The arrows indicate coexisting components of M1, R, M2 and T phases.