Metal-insulator transition in vanadium dioxide inspected by analytical electron microscopy

Doc. Mgr. Vlastimil Křápek, Ph.D.

Brno University of Technology, Technická 2, 616 69 Brno, Czechia, E-mail: krapek@vutbr.cz

Vanadium dioxide (VO₂) stands out among materials exhibiting metal-insulator phase transition (MIT) due to the proximity of its transition temperature to the room temperature, which makes it a promising candidate for applications in photonics including fast optical switching or tunable optical metasurfaces [1]. The nature of the MIT in VO₂ is not yet fully understood, with the Mott transition and Peierls transition being the most considered scenarios. Here we probed the MIT in a scanning transmission electron microscope (STEM) with in-situ heating. A combination of imaging, diffraction, and spectroscopy with nanometer spatial resolution allowed us to locally correlate the optical properties of VO₂ with applied temperature and local variations in stoichiometry.

We fabricated a thin layer of VO2 by evaporation and verified the presence of MIT by ellipsometric characterization of the refractive index. Next, we fabricated lamellas for STEM by focused ion beam milling. Energy-dispersive X-ray spectroscopy revealed a rather homogeneous composition with only discernible contamination by Ga ions implanted during the milling. High-resolution and annular dark field STEM imaging revealed pronounced structural inhomogeneity – a porous and polycrystalline character of the examined lamella (Fig. 1a). A thinned part of the lamella has been examined by core-loss (Fig. 1b) and low-loss electron energy loss spectroscopy at temperatures well below and above the MIT, and the experimental loss spectra were interpreted by ab-initio simulations and by comparison with recent literature [2,3]. We observed that the thin part of the lamella is dominated by reduced vanadium oxides (VO, V_2O_3) and exhibit no switching. This fact presents an important limit for the fabrication of VO2 nanostructures. The thicker parts of the lamella are dominated by VO_2 and exhibit signatures of thermal-induced switching.



Fig. 1 (a) High-angle annular dark field image of VO lamella reveals its porous character. The EELS characterization was taken along the blue and red lines. (b) Core-loss EEL spectra obtained from the position represented by blue and red lines shown in panel b, averaged over ten positions labeled with corresponding numbers. The spectra from the thin part of the lamella are attributed to VO (11-20) and V₂O₃ (21-30) and do not change with the temperature. The spectra from the thicker part are attributed to VO₂, some of them exhibit changes with the temperature.

References

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