

Title: The influence of structural disorder and phonon on the metal-to-insulator transition of VO<sub>2</sub>

Sang-Wook Han, In-Hui Hwang, Zhenlan Jin, and Chang-In Park

Department of Physics Education and Institute of Fusion Science, Jeonbuk National University, Jeonju 54896, Korea

[shan@jbnu.ac.kr](mailto:shan@jbnu.ac.kr)

We used temperature-dependent XAFS measurements to examine the local structural properties around vanadium atoms at the V K edge from VO<sub>2</sub> films. A direct comparison of the simultaneously-measured resistance and XAFS regarding the VO<sub>2</sub> films showed that the thermally-driven structural transition occurred prior to the resistance transition during a heating, while this change simultaneously occurred during a cooling. EXAFS measurements revealed significant increases of the Debye-Waller factors of the V-O and V-V pairs in the {111} direction of the R-phase VO<sub>2</sub> that are due to the phonons of the V-V arrays along the same direction in a metallic phase. FEFF calculations showed nearly no changes in local density of states in the V 3d orbitals. The thermally-induced phonons in the {111} direction assist the delocalization of the V 3d<sup>1</sup> electrons in the R phase VO<sub>2</sub> and the electrons likely migrate via the V-V array in the {111} direction as well as the V-V dimerization along the c-axis. This study clarifies that a tetragonal symmetry is essentially important for the metallic phase in VO<sub>2</sub>. The existence of a substantial amount of structural disorder on the V-V pairs along the c-axis in both the M<sub>1</sub> and R phases indicates the structural instability of the V-V arrays in the axis. The anomalous structural disorder that was observed on all of the atomic sites at the structural phase transition prevented the migration of the V 3d<sup>1</sup> electrons, resulting in a Mott insulator in the M<sub>2</sub>-phase VO<sub>2</sub>.

The work was conducted under the auspices of the Basic Science Research Program through the National Research Foundation of Korea government grant funded by the Ministry of Science and ICT (No. 2016R1D1A3B03931824 and No. 2017K1A3A7A09016390). The XAFS data were collected at the 8C beamline of Pohang Light Source in Korea.