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Review

Ultrafast dynamics during the photoinduced phase transition in VO_2

Daniel Wegkamp, Julia Stähler *

Department of Physical Chemistry, Fritz-Haber-Institute of the Max Planck Society, Faradayweg 4-6, 14195 Berlin, Germany

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ABSTRACT

The phase transition of VO_2 from a monoclinic insulator to a rutile metal, which occurs thermally at T_C = 340 K, can also be driven by strong photoexcitation. The ultrafast dynamics during this photoinduced phase transition (PIPT) have attracted great scientific attention for decades, as this approach promises to answer the question of whether the insulator-to-metal (IMT) transition is caused by electronic or crystallographic processes through disentanglement of the different contributions in the time domain. We review our recent results achieved by femtosecond time-resolved photoelectron, optical, and coherent phonon spectroscopy and discuss them within the framework of a selection of latest, complementary studies of the ultrafast PIPT in VO_2. We show that the population change of electrons and holes caused by photoexcitation launches a highly non-equilibrium plasma phase characterized by enhanced screening due to quasi-free carriers and followed by two branches of non-equilibrium dynamics: (i) an instantaneous (within the time resolution) collapse of the insulating gap that precedes charge carrier relaxation and significant ionic motion and (ii) an instantaneous lattice potential symmetry change that represents the onset of the crystallographic phase transition through ionic motion on longer timescales. We discuss the interconnection between these two non-thermal pathways with

Abbreviations: CB, conduction band; CIA, Coulomb interaction; COHSEX, Coulomb hole and statically screened exchange; CPT, crystallographic phase transition; DFT, density functional theory; DMFT, dynamical mean field theory; DOS, density of states; FWHM, full width at half maximum; FT, Fourier transform; HSE, Heyd, Scuseria and Ernzerhof (functional); IMT, insulator-to-metal transition; LDA, local-density approximation; LPC, lattice potential change; LSDA, spin-polarised local-density approximation; MD, molecular dynamics; mM, monoclinic metallic; mTHz, multitherahertz; PES, photoelectron spectroscopy; PIPT, photoinduced phase transition; PLD, pulsed laser deposition; R, rutile; UED, ultrafast electron diffraction; UHV, ultrahigh vacuum; VB, valence band; VO_2, vanadium dioxide; WLC, whitelight continuum.

* Corresponding author.
E-mail address: staehler@fhi-berlin.mpg.de (J. Stähler).

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particular focus on the meaning of the critical fluence of the PIPT in different types of experiments. Based on this, we conclude that the PIPT threshold identified in optical experiments is most probably determined by the excitation density required to drive *the lattice potential change rather than the IMT*. These considerations suggest that the IMT can be driven by weaker excitation, predicting a transiently metallic, monoclinic state of VO_2 that is not stabilized by the non-thermal structural transition and, thus, decays on ultrafast timescales.

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1. Introduction

Vanadium dioxide (VO_2) exhibits a thermally induced, first-order, reversible phase transition in crystallographic and electronic structure from an insulating-monoclinic to a metallic-rutile phase, both discovered more than 50 years ago by Westman and Morin [1,2]. Among other reasons, VO_2 attracted significant scientific interest for many decades due to its transition temperature, which is only slightly above room temperature ($T_C \simeq 340$ K). The transitions are, thus, experimentally easily accessible and qualify VO_2 for a manifold of applications. Another cause is the *coincidence* of the crystallographic phase transition (CPT) and the insulator-to-metal transition (IMT), which suggested a parental connection between the two. Not least the inability of both, theory and experiment, to unambiguously prove any suggested explanation of the underlying physical mechanisms provoked many scientists' curiosity.

The establishment of ultrafast lasers in the second half of the last century as powerful tools to perform time-resolved spectroscopy opened up a new path to shed light into the controversy of the phase transition in VO_2 when it was shown by Roach and Balberg [3] that the material exhibits a metal-like