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Interplay between Electronic correlations and Coherent Structural Dynamics during the Insulator-to-Metal Phase Transition in VO_2 studied by means of fs-IR/EUV pump probe core-level photoemission

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Synopsis In an fs-IR/EUV pump-probe core-level photoemission experiment the Insulator to Metal Phase Transition has been identified as being electronically driven.

Materials exhibiting photoinduced transitions from an insulating to a metallic phase are attractive candidates for ultrafast electrooptic applications because of the significant changes in the resistivity and extremely fast optical switching upon the transition[1]. Vanadium dioxide (VO_2) is one example of these solids with strongly correlated electron systems: It undergoes a first-order transition from a high-temperature metallic to a low-temperature insulating phase at almost room temperature Tc = 340 K [2]. The Insulator Metal Transition is a structural phase transition from a monoclinic (M1) unit cell in the insulating phase to tetragonal (rutile) symmetry in the metallic phase. Recent theoretical and experimental work has shown that the VO_2 phase transition results from the interplay of electronic correlations and structural distortions and can thus be regarded as a correlation-assisted Peierls transition [3, 4].

We have directly probed the time evolution of electronic states in VO_2 across the insulator-metal transition in VO2 by using near-infrared / EUV pump probe technique [5]. The femtosecond EUV pulses with photon energy of about 95 eV have been generated by means of High Harmonic Generation (HHG) and the phase transition has been initiated by the 50 fs fundamental near-infrared pump pulses. Due to direct EUV photoemission the V3p core levels and the hybridized V3d O2p valence band states are accessible. While the valence band states directly reflect their excitation by the near infrared pulses, the core level states reflect local changes of the valence band states resulting from the primary near infrared excitation as well as from structural changes.

Different temporal behaviors of lattice and electronic degrees of freedom were observed across the near infrared initiated insulator-metal transition of VO_2. These changes establish that the monoclinic-rutile transition in VO2 involves both an electronic and a structural transition. Moderate excitation fluences result in a temporally decoupled electronic transition and lattice distortion, which indicates the existence of a monoclinic metallic state within the first-order insulator to metal transition and that transition is primarily driven not by structural transitions but by transitions inside the valence electron system. Moreover, we have identified two different types of transitional structures: one is connected with atomic motions on a fs time scale, the other with atomic motions on a ps time scale.

The present data answer a standing question in isolator metal transition in VO2 and demonstrate the potential of the time-resolved electron spectroscopy to study light induced processes.

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