

Attosecond electron localization and screening dynamics in metals.

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A high degree of electron localization on the d-orbitals of transition metals and their compounds provides a lever to efficiently control their properties with light. For example, light absorption in VO₂ may result in an ultrafast electronic phase transition from a dielectric into metallic state [1], a topic that has gained a lot of attention. The essential timescale of electronic phase transitions is connected to the screening dynamics, which typically belongs to the attosecond domain, and it is followed by femtosecond electron-electron thermalization, which may blur the initial imprints of screening-induced charge re-distribution. Here we show that the properties of transition metals could in principle be manipulated much faster than the electron thermalization timescale and even faster than the optical cycle.

To study attosecond screening dynamics, we utilize an attosecond probe pulse with a photon energy tuned around the collective resonance in titanium. Absorption of the probe pulse is highly sensitive to the screening of the probe field and thus serves as a fine tool to study the electron dynamics induced by a few-cycle optical pump pulse with a time resolution well below the typical electron-electron thermalization timescale.

The experiments are performed on an attosecond beamline at ETH Zurich in a transient absorption configuration combined with attosecond photoelectron streaking from a noble gas, which provides a time reference [2,3]. We find that the pump pulse incident on the 50-nm Ti foil modifies the probe absorption much faster than the optical cycle, with a maximum response time estimated to 1.12 ± 0.29 fs. In the spectral domain, the absorption is increased above the M_{2,3} edge of Ti. It exhibits a peculiar spectral shape, which cannot be explained by the state-filling effect or electronic structure renormalization alone. With the help of *ab-initio* time-dependent density functional theory (TDDFT) calculations we find that the observed spectral shape is due to modified screening caused by the pump-induced electron localization on the Ti d-orbitals.

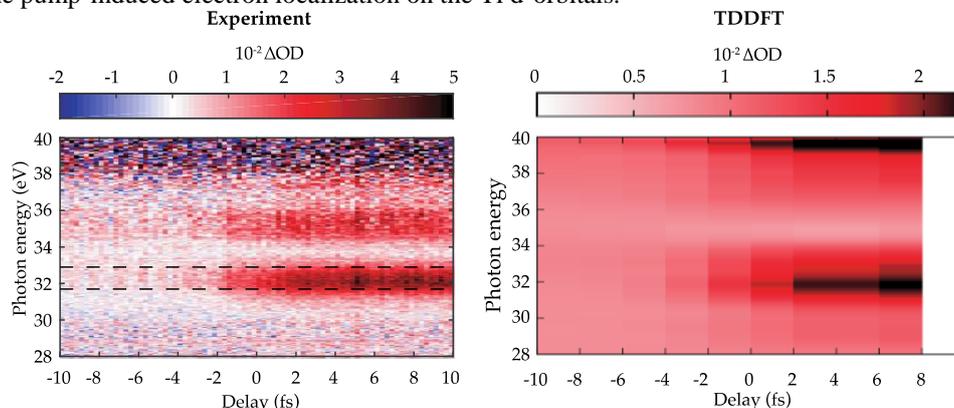


Fig. 1 Experimental and simulated (TDDFT) transient absorption spectrogram around the Ti M_{2,3} absorption edge, which is located at 32.6 eV.

Our findings highlight the light-induced screening dynamics unfolding prior to electron-electron thermalization and demonstrate the possibility to manipulate materials faster than the optical cycle via changing the relative population of localized and delocalized electronic orbitals. Similar dynamics is further observed in metallic Zr. We anticipate our findings to advance the understanding of ultrafast electronic phase transitions and to influence the interpretation of attosecond photoemission experiments.

References

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Abstract.

We experimentally investigate attosecond electron dynamics in transition metals with transient absorption spectroscopy and ab-initio calculations. We observe light-induced sub-cycle modification of electron-electron screening, suggesting that material properties can be manipulated much faster than electron-electron thermalization.