Dynamics of correlated electrons in ultrashort electric fields: From simple to complex systems

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Abstract

The experimental investigation of correlated electron dynamics in ultrashort laser pulses is discussed. Bound quantum states are accessed by using attosecond transient absorption spectroscopy which enables a high temporal resolution with an associated broadband spectral coverage, while at the same time state selectivity at high spectral resolution is achieved with a grating-based spectrometer. The presented ultrafast measurement and control of two-electron dynamics in the helium atom serves as a most basic model system to test fundamental theories. Furthermore, first measurements are presented on vanadium dioxide (VO_2), a thin-film solid state target which undergoes an ultrafast insulator-to-metal phase transition.

The dynamical interplay between multiple electrons in any natural system is a key ingredient to determine its macroscopic properties. Observing and controlling these dynamics by using ultrashort laser pulses is a promising approach to understand and manipulate the system under study at a fundamental level.

The most basic natural system that allows an investigation of such dynamics is the helium atom, with two electrons bound to the nucleus. Using the technique of attosecond transient absorption spectroscopy, a correlated two-electron wave packet could be experimentally reconstructed and its 1.2-fs quantum beat was directly resolved in real time [1]. The wave packet is constituted of two low-lying doubly excited states with principal quantum numbers $N = 2,3$. The correlated nature of both active electrons is depicted in Fig. 1, where a one-dimensional cut along the laser polarization serves as a simplified illustration of the quantum mechanical probability distribution of the two electrons. By tuning the laser-pulse intensity, this approach also enabled experimental controllability of the shape of the two-electron wave packet at a selected time after the interaction. Here, the identification of laser-controlled absorption lines and relating their profile to intrinsic phases of the associated quantum states [2], served as a key tool for this investigation.

Regarding more complex systems, transition metal oxides represent a class of materials with many interesting effects and applications, such as ultrafast insulator-to-metal phase transitions [3] which can be photoinduced. The interaction of multiple electrons in the open d-shell of the transition metal here plays a key role. A prominent example is vanadium dioxide, where the thermally induced phase transition has been discovered in 1959 [4], and the exact interplay between competing electronic and structural effects is still subject of intense research. Thus the unprecedented temporal and spectral capabilities of attosecond transient absorption spectroscopy provide a unique tool to shed new light on this system.

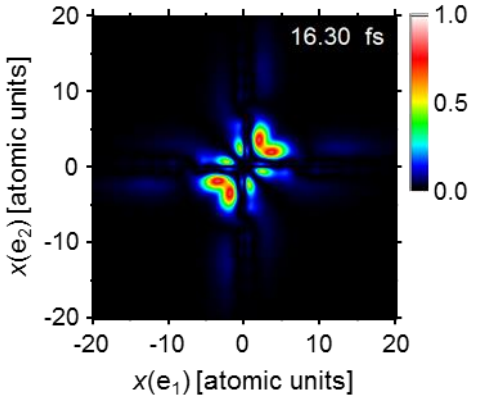

Fig. 1. Two-electron probability distribution along the laser polarization direction, plotted as a function of the position x of each electron, at a snapshot $t = 16.3$ fs after excitation. The involved states 2s2p and sp_{23+} form a correlated two-electron wave packet in helium.

In the presented experiment, a few-cycle visible pulse is used to excite carriers from the valence into the conduction band of VO_2. The subsequent temporal evolution of this non-equilibrium state is traced in real time, with spectroscopic access to the vanadium M-edge at 40 eV, where an inner 3p electron is excited into an empty 3d state of the conduction band. Spectrally broad absorption changes are observed over a range of 20 eV, as well as an ultrafast redistribution of spectral weight within few 10s of femtoseconds. A comparison to thermally induced spectral changes, which differ from the photoinduced results, hints on the importance of transient intermediate states that are accessed by the ultrashort laser pulse, where electron correlation effects should play a role.

References

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