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ISBN

9781943580118

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Publication Date

2016-12-16

Peer reviewed

Ultrafast Transient Absorption at the Germanium M_{4,5}-edge to Measure Electron and Hole Dynamics

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Abstract: Extreme ultraviolet (XUV) transient absorption at the germanium M_{4,5}-edge simultaneously measures electron and hole dynamics over 1.5 ps with few-femtosecond resolution. In the analysis, time-dependent density functional theory (TD-DFT) will be compared with experimental data.

OCIS Codes: (300.6470) Spectroscopy, semiconductors; (320.7130) Ultrafast processes in condensed matter, including semiconductors; (320.7150) Ultrafast spectroscopy.

XUV transient absorption is used to study electron and hole dynamics at the germanium M_{4,5}-edge. Previous transient absorption experiments in silicon dioxide [1] and silicon [2] performed at the silicon L_{2,3}-edge only reported electron dynamics in the conduction band. The valence band dynamics were not observed because the silicon L_{2,3}-edge is an excitation from a core 2*p* level. As such it has poor oscillator strength to the valence band, which is of 3*p* character. By instead investigating the M_{4,5}-edge state in germanium, which arises from a 3*d* core transition, sensitivity to the valence band and hole dynamics can be achieved.

Germanium is an industry-relevant [3,4] semiconductor whose band gap at room temperature is accessible with photons from a typical titanium sapphire source. For the experiment, 100 nm thin films of germanium were deposited using electron beam evaporation and then annealed at 450°C [5]. The annealing step produced multicrystalline samples with domain sizes of 5-10 nm, as confirmed by X-ray diffraction measurements. For the pump-probe experiment, 5 fs pulses with a central energy of 1.55 eV (Figure 1a.) are divided into two equal paths. The majority of the pulse is focused into a gas jet filled with xenon gas, where high harmonic generation (HHG) up-converts this pulse into the extreme ultraviolet (XUV). By employing appropriate gating optics in the high harmonic generation arm, the XUV spectrum is tailored to a continuum spanning 25 to 40 eV (Figure 1b.), with signatures of an attosecond continuum. The XUV continuum is ideal for studying the germanium M_{4,5}-edge, which corresponds to excitation from the 3*d*_{3/2} and 3*d*_{5/2} core electronic state to both the valence and conduction bands. The remainder of the pulse is delayed relative to the XUV pulse, and is used to pump electrons in from the valence to the conduction band in germanium (Figure 1c.).

Figure 1a. A spectrum of the 1.55 eV, 5 fs pump pulse used in the experiment and **b.** the XUV probe pulse spectrum used in this experiment. By the nature of the HHG process, it is necessarily temporally shorter than the 5 fs driving pulse. **c.** XUV transient absorption scheme shown here. The 5 fs pulse (red arrows) is used to excite electrons (red circles) to the conduction band leaving holes (white circles) in the valence band, both of which are probed with few-femtosecond resolution using the XUV pulse (blue arrows).

The differential optical density, ΔOD , as a function of time delay is shown in Figure 2. Two features are immediately visible at the $M_{4,5}$ -edge after excitation. The feature with positive ΔOD from 28-29 eV corresponds to the holes in the valence band, while the feature with negative ΔOD from 30-32 eV arises from the electrons in the conduction band. These features appear in less than 10 fs, within the instrumental response function measured in a XUV-NIR cross-correlation in argon [6]. After 1 ps, the two features have decayed completely, and the sample has completely relaxed. Analyzing the processes and decay mechanisms becomes feasible from the presented data set, as it is a complete picture of hole and electron dynamics measured in-situ at ultrafast timescales.

Figure 2. A measured ground state absorption spectrum of the $M_{4,5}$ -edge in germanium. This is shown next to the differential optical density, ΔOD , at different time delays (color bar on the right). Negative time delays correspond to the XUV pulse arriving first and positive time delays correspond to the 5 fs pump pulse arriving first. Two spectrally separated features can be seen. The Fermi level is indicated with a dashed black line in both plots.

In addition to further analysis of the dynamics, theoretical efforts are ongoing to reproduce the experimental data. To reproduce the experimental differential absorption traces, the XUV absorption of the ground state is first calculated and then compared to the XUV absorption of an excited state predicted by TD-DFT for a sub-10fs, 1.5 eV pulse with 10^{12} W/cm² intensity. This yields qualitative agreement, however, more realistic pulse parameters are expected to improve simulation results in the near future.

By observing electron and hole dynamics simultaneously, a complete understand of carrier dynamics after excitation can be achieved and XUV transient absorption proves to be a valuable tool in this goal. The wealth of dynamics visible in the time-dependent differential optical density traces (Figure 2) measured at ultrafast timescales is currently analyzed in depth. Further experimental details and analysis along with comparison to TD-DFT simulations will be reported at the conference.

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