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Attosecond transient reflectivity of electron dynamics in germanium

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Abstract: Attosecond transient reflectivity in the extreme ultraviolet is developed for monitoring band-gap excitation dynamics in germanium. The investigations unravel the fastest carrier-scattering processes on the few-femtosecond time scale, and carrier thermalization on longer time scales.

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1. Introduction

Valence-to conduction band excitation in semiconductors is the centerpiece of modern optoelectronics. The excitation process and subsequent electron dynamics determine the electron and hole mobilities of semiconductors and thus the performance of semiconductor-based devices.

The electron relaxation dynamics upon band-gap excitation in germanium have been investigated by several femtosecond pump-probe studies in the visible [1-3]. However, none of those studies addressed the fastest time scales of the excitation process itself or the carrier-carrier scattering dynamics in the first few fs after excitation.

Attosecond science holds the promise to unravel the fastest electronic processes in solid-state materials [4,5]. Attosecond transient absorption of nanometer-thick films in particular combines the highest temporal resolution with a good energy resolution. Furthermore, it takes advantage of the element-specificity of the core-to-valence and core-to-conduction band transitions in the extreme ultraviolet (XUV) and soft x-ray region of the spectrum.

Here attosecond transient reflectivity in the extreme ultraviolet is developed. Transient reflectivity allows the study of thick and robust single-crystalline samples while simultaneously exploiting the advantages inherent to transient absorption. By analyzing the reflection from the sample in terms of a complex-valued index of refraction, both the real and imaginary part of the refractive index are extracted. Signatures of the transient excitation of electrons into the conduction band, and the creation of holes in the valence band of Germanium can be identified in the data, complementary with observations in XUV transient absorption experiments [6]. An initial broadening of the transient features on a sub-10 fs time scale is observed, which is indicative of carrier-carrier scattering times. Subsequently, carrier thermalization sets in and leads to a relaxation of the transient features on a femtosecond to picosecond time scale.

2. Experimental details

Sub-5 fs carrier-envelope phase stabilized laser pulses are generated through hollow-core fiber compression. The pulse energy after compression is about 600 μJ , and the spectrum of the pulses is centered around 700 nm and spans from 500-950 nm. A fraction of the pulses is used as pump pulses for injecting electrons from the valence into the

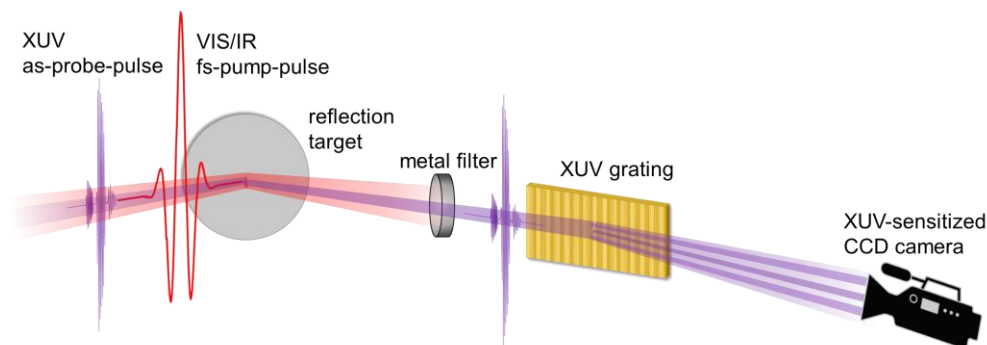


Fig. 1: Schematic illustration of the experimental setup.

conduction band across the 0.8 eV direct band gap. The other fraction is used for generating attosecond pulses in the extreme ultraviolet part of the spectrum (27 eV – 40 eV) through high-harmonic generation from xenon. Both pulses are recombined by an annular mirror and focused on the single-crystalline germanium target (see Fig. 1). The target reflects the pulses into an XUV spectrometer consisting of a flat-field grating and an XUV-sensitized CCD camera. This configuration allows to track the reflection spectrum of Ge as a function of the pump-probe delay on attosecond to picosecond time scales. Previous XUV femtosecond studies of magnetic materials employing reflection geometries used samples which were directly deposited on the grating [7].

The attosecond pulses in the XUV are used for monitoring the carrier dynamics at the $M_{4,5}$ edges of germanium through promoting electrons from the $3d_{3/2}$ and $3d_{5/2}$ core orbitals into the valence and conduction band after band-gap excitation through the pump pulses. The spin-orbit splitting of the core state can be treated through a Fourier reconstruction method and does not affect the conclusions presented in the present abstract. The reflection from the germanium target can be referenced to a reflection from a separate gold mirror, which enables determining the absolute reflectivity of the sample.

3. Results and Discussion

Measured absolute reflectivities for germanium in the absence of optical excitation are shown in Fig. 2(a). Independent measurements for s-polarized (blue dash-dotted line) and p-polarized (red solid line) XUV pulses can be used to reconstruct the complex-valued index of refraction $n = (1-\delta) + i\beta$ (Fig. 2(b)) of germanium through a numerical inversion of the underlying Fresnel equations. Both the real part δ (solid green line) and the imaginary part β (dash-dotted black line) of the refractive index have a minimum around 29.5 eV, which corresponds to a transition from the 3d core orbital to the conduction band, and further local maxima and minima at higher energies.

Transient reflectivity traces are shown in Fig. 3. The relative change in reflectivity $\Delta R/R$ is plotted as a function of the XUV photon energy as well as the pump-probe delay. Figure 3(a) shows the slow dynamics in germanium stretching from -200 fs to 2200 fs, whereas Fig. 3(b) resolves the fastest dynamics right after valence-to-conduction band excitation with attosecond precision.

The two strongest features in both traces are a broadband decrease in reflectivity around 28 eV, and an increase around 29.5 eV. Our reconstruction of the complex-valued refractive index (Fig. 2) enabled the identification of the feature at 29.5 eV as a core-to-conduction band transition. The excitation of electrons into the conduction band leads to Pauli state blocking and therefore to a lowered absorption, which explains the observed increase in reflectivity. Consequently, at lower energies holes in the valence band open up, which facilitate a stronger absorption of the XUV and a decreased reflectivity as observed experimentally around 28 eV. In particular, the spectral feature corresponding to the valence-band edge around 28.5 eV in Fig. 3(a) shifts towards higher energies for longer pump-probe delays, which is caused by hole thermalization towards the top of the valence band.

Multiple weaker features are visible in Fig. 3(a) at higher energies around 31.2 eV, 33 eV and 34.5 eV. These features occur at the same photon energies where we could identify local extrema in the real and imaginary parts of the refractive index. We attribute those transient features to an overall shift and broadening of the reflection spectrum upon photoexcitation. The spectral shifts are attributed to a band-gap renormalization following photoexcitation, whereas the overall broadening of all spectral features reflects a change in the core-hole life-time due to a modified screening after valence-to-conduction band excitation.

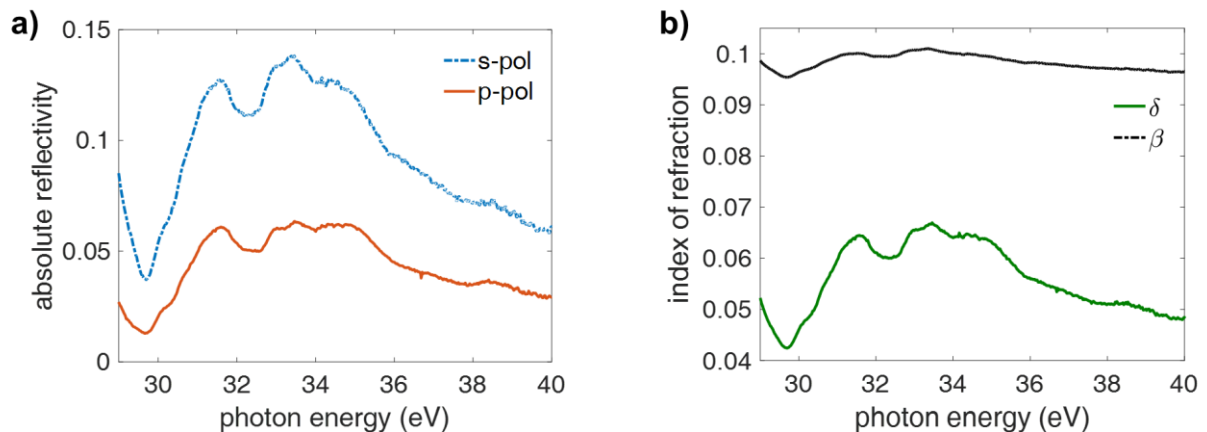


Fig 2: (a) Static measurements of absolute reflectivities of unexcited germanium for s-polarization and p-polarization. (b) Reconstructed real (δ) and imaginary (β) part of the index of refraction.

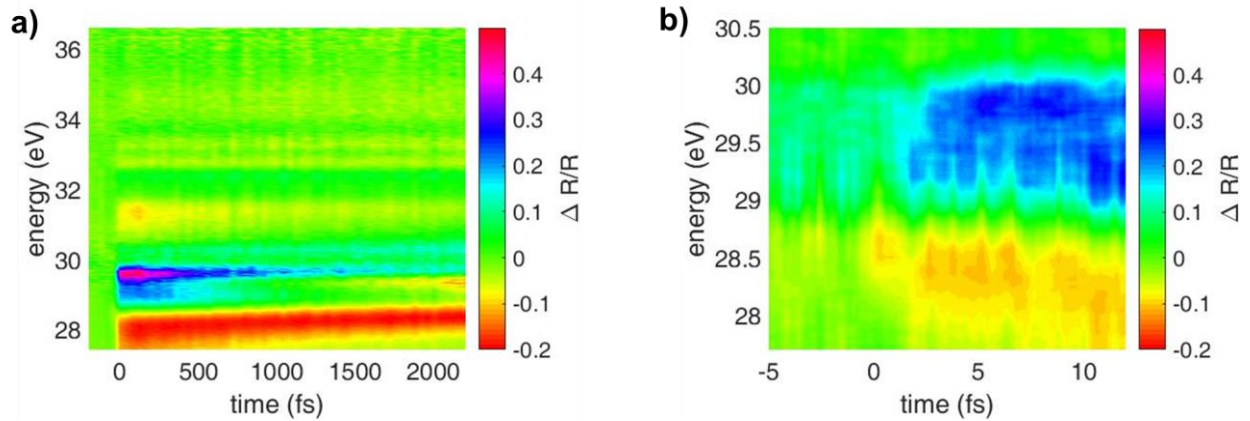


Fig. 3: (a) Transient reflectivity changes $\Delta R/R$ in the XUV as a function of the pump-probe delay and photon energy.
(b) Same as (a) for shorter pump-probe delays.

Most intriguingly, the short time dynamics of germanium (Fig. 3(b)) reveal a time structure of the electron and hole distributions within the duration of the sub-5 fs laser pulse used for carrier excitation. Electron holes at 28.5 eV with a narrow energy width of about 0.4 eV are created at 0 fs time delay and subsequently broaden to a width of about 1 eV within the duration of the excitation pulse. This finding is attributed to carrier-carrier scattering, which leads to the above mentioned changes in screening of the core-hole, that shorten the core-hole life-time and thus increase the width of all spectral features.

4. Conclusions and Outlook

We introduced attosecond transient reflectivity as a new technique to investigate electron dynamics in solids. Transient reflectivity can combine the highest temporal resolution with a high frequency resolution as well as element specificity. The reconstruction of the full complex-valued index of refraction in the XUV was demonstrated. This will be used to track the time-dynamics of the real and imaginary part separately, thus yielding more information than typical transient absorption experiments.

Our results on germanium revealed the separate observation of the time dynamics of holes in the valence band and electrons in the conduction band, complementary with previous XUV absorption studies [6]. Besides thermalization rates on the femtosecond to picosecond time scale, few-femtosecond dynamics of the initial hole distribution could be observed. These dynamics are indicative of the fastest carrier-carrier scattering processes.

5. Acknowledgments

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