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Attosecond kinetics of photoexcited Germanium

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Abstract: Attosecond transient reflectivity is developed to observe the photoexcitation dynamics in germanium. Attosecond time-resolved measurements of the dielectric function reveal a few-femtosecond collective electronic response time, which renormalizes the Coulomb interaction between the excited carriers.

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1. Introduction

The electronic motion in a semiconductor after light absorption is a central aspect of modern opto-electronics. However, a real-time observation of the initial electronic response following single-photon excitation has so far defied experimental approaches due to its extreme time scales.

Here, attosecond transient reflectivity in the extreme ultraviolet is developed. The technique is applied to measure the attosecond time-resolved dielectric function at the germanium core-level transition following carrier promotion by visible-to-infrared sub-5 femtosecond (fs) pulses from the valence into the conduction band in single-crystalline germanium. The buildup of holes and electrons in the valance and conduction band is monitored on attosecond timescales by the change in the reflectivity at the germanium $M_{4,5}$ edge (30 eV).

Earlier time-resolved experiments explored the advantages of reflection geometries in the visible or extreme ultraviolet spectral range for monitoring ultrafast demagnetization dynamics [1] and coherent phonons [2]. In the present study, attosecond-time resolved reflection spectroscopy is used to monitor the full transient dielectric function and to unravel the earliest electron kinetics after photoexcitation.

2. Results

The reflectivity changes in germanium are shown in Fig. 1a over a time scale of 50 fs. The Fermi level is associated with energies around 29.3 eV. The overall reflectivity changes can mainly be attributed to the newly created electrons and holes, which cause a reflectivity decrease and increase due to the opening and blocking of new absorption channels for the germanium core-level transition around the Fermi level, respectively. Furthermore, the photoexcited carrier distribution causes a shrinkage of the band gap due to band-gap renormalization following photodoping, as well as a core-level shift due to the modified core-level screening, causing a total red-shift of the excited state spectrum compared to the ground state spectrum.

The electron and hole features, which correspond to the largest reflectivity changes around the Fermi level, are analyzed in more detail. The electron and hole features are found to exhibit a 1.4 fs oscillation as shown in Fig. 1b. The associated oscillation frequency is indicative of a 2ω oscillation, where ω is the fundamental frequency of the photoexcitation pump pulse. A 2ω oscillation is an indication of a field-induced polarization and transient population of the bands, which follow the electric field rather than the envelope of the laser

pulse. Not all modulations follow exactly a 2ω periodicity. The lineouts from 29.8 to 30.2 eV are not in phase with the oscillations from 28.9 to 29.2 eV, and the signals possess higher frequency components. Such higher frequency modulations can be caused by higher order response of the semiconductor to the electric field of the excitation pulse. A time-dependent semiconductor Bloch equations will be developed to describe the inter- and intra-band couplings during photoexcitation of germanium and unravel the exact origin of the observed oscillations.

The measurement of the attosecond dielectric function further enables tracking of the buildup of screening of the core-hole potential due to the collective electronic motion in the valence and conduction. In the raw data shown in Fig 1a, we observe an initial broadening and shifting of the bleach in reflectivity around 27-29 eV. This response of the system, which is too fast to involve nuclear motion, can be further analyzed through a measurement of the full dielectric function. This is achieved by measuring the transient reflectivity changes upon photoexcitation for pump and probe pulses, which are polarized parallel or perpendicular with respect to the plane of incidence, and subsequently inverting data with the underlying Fresnel equations.

It is experimentally observed through the real part of the dielectric function that a bare, unscreened Coulomb potential is formed instantaneously after photoexcitation (not shown). A subsequent broadening of the real part of the dielectric function over a few fs is attributed to screening of the Coulomb potential due to the collective electronic response of electrons and holes in the valence and conduction bands, respectively. Simultaneously, the imaginary part of the dielectric function tracks the buildup of new populations of carriers in the valence and conduction band. The experiment shows that two sharp features appear instantaneously due to the change in state-filling in the valence and conduction band, with an additional broadening occurring on a few fs time scale. This broadening is attributed to a response time of the collective electronic motion, which screens the Coulomb potential and changes the absorption due to carrier redistribution. The time scale of the screening agrees with the inverse plasma frequency of the electron-hole plasma created by photoexcitation at the estimated densities. Comparable time scales have been deduced from electro-optical sampling of Terahertz pulses resonant with the electron-hole plasma frequency, which were transmitted through photoexcited semiconductors [3]. The present study directly resolves how the photoexcited carriers in their respective bands respond following carrier injection from the valence into the conduction band.

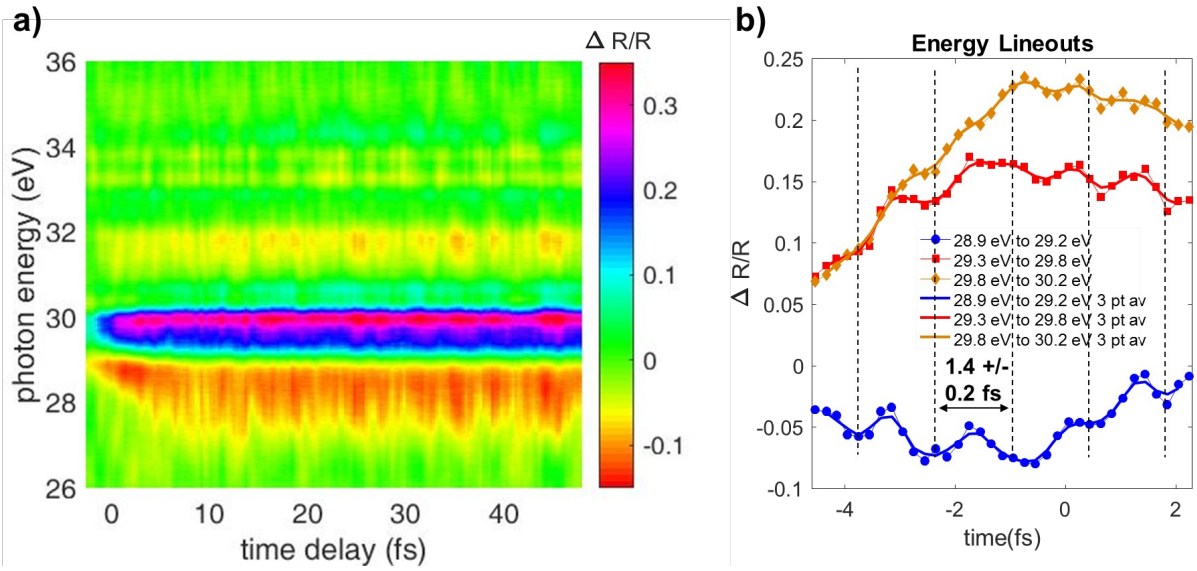

Figure 1: (a) Transient reflectivity changes of photoexcited germanium. (b) Lineouts of the reflectivity changes integrated over three different energy ranges as indicated in the figure. The dashed vertical

lines are separated by 1.4 fs, which corresponds to half of the optical cycle duration of the excitation pulse. The error bars of 0.2 fs correspond to the smallest time delay increment, which was sampled in the experiment.

3. Conclusion

Attosecond transient reflectivity was developed to monitor the electronic response of germanium during and directly after photoexcitation. A field-driven transient population buildup as well as a finite response time following photoexcitation were identified experimentally.

Similar electronic responses after photoexcitation are anticipated to occur in all materials. The time scales involved will limit the ultimate speed of charge transfer in metal complexes, charge separations in photovoltaic cells and phase transitions in strongly correlated materials. The results presented here thus provide an experimental measurement of the fundamental timescales of charged particle interactions in solids.

4. References

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