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Ultrafast Valley-resolved Carrier Dynamics in Group IV Semiconductors

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Abstract: Attosecond transient absorption spectroscopy at the $M_{4,5}$-edge of Ge following ultrafast photoexcitation reveals valley-resolved hot electron and hole relaxation, carrier recombination and trapping in Ge and Si-Ge alloy in unprecedented clarity and simultaneously.

I. INTRODUCTION

Germanium and its alloys with Silicon have promise for creating multi-junction solar cells with higher efficiency and mid-infrared optoelectronics. However, measuring carrier dynamics is complicated by a multitude of energetically similar valleys and various relaxation and recombination pathways.

Here, attosecond transient absorption is employed to monitor carrier dynamics in Ge and SiGe of both electrons and holes simultaneously in the relevant valleys as well as induced band dynamics with few-femtosecond time resolution and high energy resolution.

II. EXPERIMENT AND RESULTS

In the experiment, a 5 fs VIS-NIR pump pulse excites carriers across the direct band gap of pure Ge and the largely indirect band gap in Si-Ge alloy. The dynamics are probed at the Ge $M_{4,5}$-edge (~30 eV) with a time-delayed broadband extreme ultraviolet pulse generated by high harmonic generation in xenon spanning ~20-45 eV.

A typical transient absorption signal at the Ge $M_{4,5}$-edge (Fig. 1a) contains the energetic distribution of both carriers, electrons and holes, due to state blocking as well as spectroscopic features induced by bandshifts (e.g. due to band gap renormalization) and excited state broadening (e.g. due to many body effects). These individual contributions are disentangled by an iterative procedure[1] yielding a representation of carriers over energy and time in unprecedented clarity (Fig. 1b) allowing the direct observation of hot carrier relaxation and recombination.

Specifically, hot carrier relaxation on a 100-fs time scale and carrier recombination on a 1-ps time scale are observed in pure Ge following excitation across the direct band gap. The fast recombination is attributed to the nanocrystalline nature of the sample. The high energy resolution further allows extracting valley-dependent carrier lifetimes (inset Fig. 1b) to obtain a full picture of the carrier dynamics.[1]

In contrast, in Si-Ge alloy phonon-assisted excitation across the indirect band gap is observed. Following hot carrier relaxation in the alloy, trapping of electrons in midgap states is experimentally observed and the asymmetry between electron and hole signal (Fig. 1c) indicates a significantly reduced recombination rate across the indirect gap.[2]

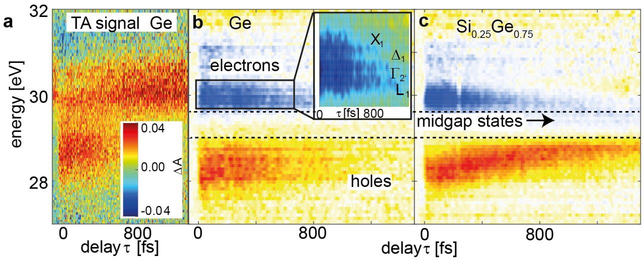

Fig. 1. (a) Typical transient absorption signal at the Ge $M_{4,5}$-edge. (b) Retrieved carrier dynamics from (a) featuring hot electron and hole relaxation and recombination (Inset: enlarged conduction band shows valley-specific carrier dynamics). (c) In Si-Ge alloy trapping of electrons in midgap states is observed.

III. CONCLUSIONS

In conclusion, attosecond transient absorption is successfully employed to track several electron volt wide spectroscopic features at the $M_{4,5}$-edge of Ge revealing valley-specific hot electron and hole dynamics. Applying the reporter atom concept as implemented for Si-Ge alloy holds great promise for studying carrier dynamics and localization in more complex systems such as ternary and quaternary semiconductors.

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