

Tracking Ultrafast Carrier Dynamics in Single Semiconductor Nanowire Heterostructures

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Abstract. An understanding of non-equilibrium carrier dynamics in silicon (Si) nanowires (NWs) and NW heterostructures is very important due to their many nanophotonic and nanoelectronics applications. Here, we describe the first measurements of ultrafast carrier dynamics and diffusion in single heterostructured Si nanowires, obtained using ultrafast optical microscopy. By isolating individual nanowires, we avoid complications resulting from the broad size and alignment distribution in nanowire ensembles, allowing us to directly probe ultrafast carrier dynamics in these quasi-one-dimensional systems. Spatially-resolved pump-probe spectroscopy demonstrates the influence of surface-mediated mechanisms on carrier dynamics in a single NW, while polarization-resolved femtosecond pump-probe spectroscopy reveals a clear anisotropy in carrier lifetimes measured parallel and perpendicular to the NW axis, due to density-dependent Auger recombination. Furthermore, separating the pump and probe spots along the NW axis enabled us to track space and time dependent carrier diffusion in radial and axial NW heterostructures. These results enable us to reveal the influence of radial and axial interfaces on carrier dynamics and charge transport in these quasi-one-dimensional nanosystems, which can then be used to tailor carrier relaxation in a single nanowire heterostructure for a given application.

Introduction

There has been an explosion in research on semiconductor nanowires (NWs) in recent years, primarily due to their variety of potential electronic and optoelectronic applications in areas such as photodetection, electrically-driven lasers, nanoscale transistors, and solar cells [1]. Radial core/shell and axial nanowire heterostructures, composed of one or more layers with different properties, have enabled greater control of device operation for these applications [2]. The interfaces between different layers in these heterostructures strongly influence their properties and in turn device performance. However, a basic understanding of how these interfaces affect carrier dynamics and charge transport in individual NWs is lacking, as previous studies have provided little information due to their technical limitations (e.g., complications from studying NW ensembles, lack of spatial and temporal resolution, and the influence of contacts).

Here, we use ultrashort optical microscopy (UOM) to measure carrier dynamics and diffusion in single NW heterostructures with high temporal and spatial resolution, allowing us to track carrier

dynamics and charge transport in a non-contact, non-invasive manner. By performing ultrafast polarization-and-spatially-resolved optical pump-probe spectroscopy on different individual Si NWs, we are able to explain the relative influences of surface recombination and Auger recombination on carrier relaxation in a single Si NW [3]. The carrier lifetimes measured for NWs with different diameters are subsequently used to calculate the diffusion length and surface recombination velocity, which are essential parameters for electronic device applications. Finally, performing UOM with non-overlapping pump and probe beams enables us to directly measure carrier diffusion along these single SiNWs. These first ultrafast optical measurements on single Si NWs thus provide much insight into the basic mechanisms governing carrier relaxation in quasi-1D semiconductor NWs, while demonstrating our ability to extract device-relevant parameters in a non-contact manner.

Experiments and discussion

A femtosecond Ti:sapphire laser oscillator centered at 840 nm is divided into pump and probe beams, with the probe power < 10% of the pump power. The pump beam is then frequency-doubled in a BBO crystal to generate femtosecond pulses at 420 nm. Our initial experiments, focusing on polarization-and-diameter-dependent dynamics, imaged the sample position as well as the pump ($\sim 25 \mu\text{m}$ diameter) and probe ($\sim 20 \mu\text{m}$ diameter) spots onto a CCD camera through a 50X microscope objective lens at the back side of the sample (Fig. 1 (a)). These experiments used vapor-liquid-solid (VLS) grown SiNWs that were dry transferred onto a sapphire substrate with diameters of $d = 50\sim 240 \text{ nm}$, and lengths of $l \sim 80 \mu\text{m}$. Subsequently, to measure carrier diffusion, we used a 20X objective to focus the pump and probe beams to $5 \mu\text{m}$ and $2 \mu\text{m}$ spots, respectively, and slightly tilted the pump beam to spatially overlap or separate the two spots along the NW (Fig. 2(a)); this enabled us to measure carrier dynamics and track charge carriers as they propagate along the NW axis. The carrier diffusion experiments used Si and Si/SiO₂ core/shell NWs fabricated by a combination of e-beam lithography and Si deep reactive ion etching, followed by thermal oxidation and stripping steps to form pristine NW surfaces, that were then transferred onto a transparent sapphire substrate.

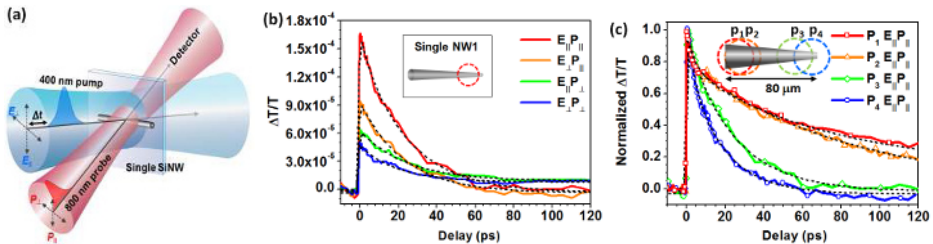


Fig. 1. (a) An illustration of a non-degenerate pump-probe measurement on a single Si NW in transmission. (b) Photoinduced transmission changes with different pump and probe polarizations at $12.0 \mu\text{J}/\text{cm}^2$ pump fluence. (c) Photoinduced transmission changes as a function of position along the NW (reprinted from [3] with permission).

Our initial experiments revealed a dependence of the photoinduced transmission change, $\Delta T/T$, in single Si NWs on the light polarization (Fig. 1(b)); here, we use $E_{||}$ and E_{\perp} to indicate pump polarizations parallel and perpendicular to the NW axis, and $P_{||}$ and P_{\perp} to indicate probe polarizations parallel and perpendicular to the NW axis, respectively. There is a clear anisotropy in the relaxation times measured for light polarized parallel and perpendicular to the NW axis, with faster relaxation for $E_{||}$ and $P_{||}$ than E_{\perp} and P_{\perp} . This is due to the polarization-dependent absorption of light in the NWs [3]; the dynamics for different polarizations were nearly identical when normalized to the initial photoexcited carrier density N for a given pump fluence and polarization. Furthermore, this

data fit well to a 1D Auger recombination model, indicating that this process dominates the dynamics at higher carrier densities. Finally, we performed position-dependent UOM experiments along the axis of a single tapered Si NWs (Fig. 1(c)), which showed that the carrier lifetime increased with the NW diameter. This is due to surface trapping and recombination, as observed in previous experiments on NW ensembles. Comparison of the polarization-dependent and diameter-dependent data enabled us to show that surface recombination dominates carrier relaxation at low densities, while Auger recombination governs the dynamics at high carrier densities [3].

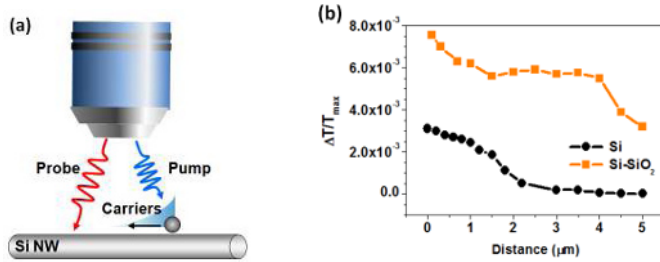


Fig. 2 (a) A conceptual illustration of UOM with spatially separated pump and probe beams. (b) Maximum $\Delta T/T$ values for both Si NWs as a function of separation.

Subsequently, UOM experiments with spatially separated pump and probe beams were performed on a single Si NW with and without a SiO₂ shell layer (Fig. 2(a)). With the probe fixed on one end of the NW, we varied the pump-probe separation from 0 to 5 μm. This revealed strongly position-dependent carrier dynamics, due to carrier diffusion (Fig. 2(b)). The magnitude of the $\Delta T/T$ signal from the core/shell Si NW is about twice that of the bare Si NW, and the decay time is slower in the core/shell NW (147 ps) than in the bare Si NW (~90 ps). This can be explained by the fact that surface passivation by SiO₂ reduces the surface trap density [4] as well as the surface recombination velocity [5]. These experiments thus reveal a significant influence of SiO₂ passivation on the carrier dynamics, while demonstrating our ability to track carrier diffusion through a single NW.

In conclusion, we have shown that ultrafast optical microscopy opens new pathways for directly studying carrier dynamics and charge transport in quasi-1D nanosystems. We directly measured polarization-dependent carrier dynamics and diffusion in Si and Si/SiO₂ NWs for the first time by tracking carriers through space and time after femtosecond photoexcitation. This research therefore has potential application to NW-based devices and optoelectronics by combining measurements at both micrometer distance and femtosecond time scales to reveal the intrinsic properties of these quasi-one-dimensional nanosystems.

References

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