Tracking charge carriers through space and time in single silicon core-shell nanowires

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Abstract: We map space-and-time-dependent carrier dynamics in single silicon nanowires firstly, using ultrafast optical microscopy. This enables us to directly measure acoustic phonon oscillations and carrier velocities in Si and Si/SiO_2 core-shell nanowires.

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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, including photodetectors, electrically-driven lasers, nanoscale transistors, and solar cells [1]. Recent success in the fabrication of axial and radial core/shell heterostructures on NWs, composed of one or more layers with different properties, has enabled greater control of device operation for optoelectronics and solar cells [2]. Since interfaces between different layers in heterostructured NWs strongly influence their properties and in turn device performance, it is especially important to understand carrier dynamics in these quasi-one dimensional (1D) systems.

Here, we use ultrafast optical microscopy (UOM) to directly examine carrier dynamics and carrier velocities in both single silicon (Si) core and Si/SiO_2 core/shell NWs with high temporal and spatial resolution in a non-contact manner. By measuring the time-resolved photoinduced change in transmission ($\Delta T/T$) at a specific position and varying the relative pump and probe positions, we can track carrier relaxation and determine carrier velocities along the NW. A striking difference was observed in carrier dynamics for Si NWs with or without SiO_2, due to trapping in unpassivated surface states. Finally, we also observed strong acoustic phonon oscillations in both Si and Si/SiO_2 NWs for the first time, independent of the separation distance.

We employed ultrafast optical microscopy to track carriers through space and time in a single NW. 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. By using a 20X (0.4 NA) microscopic objective lens, the pump (5 μm spot size) and probe (2 μm spot size) beams are collinearly focused at the sample position, with polarization parallel to the NW axis. By slightly tilting the pump beam, we can spatially separate the two spots along the NW, which enables us to directly track charge carriers (Fig. 1 (a)). The transmitted probe light was then collected through a 50X objective lens and its intensity was measured by a photodiode, while the NW and light positions are simultaneously monitored using a CCD camera. The Si NWs were fabricated by a combination of e-beam lithography and Si deep reactive ion etching with a diameter of 255 nm and length of 9 μm. A subsequent SiO_2 wet etching process makes Si/SiO_2 core/shell NWs, after which both NWs are dry transferred to a sapphire substrate (Fig. 1(b)).

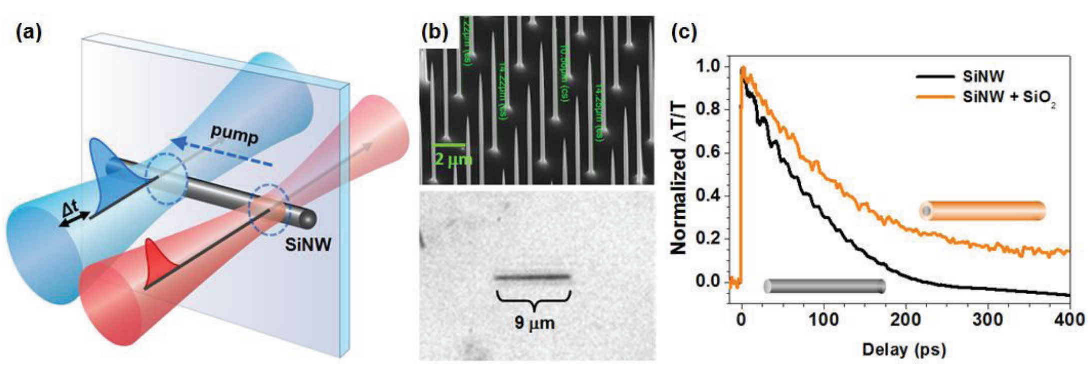

Fig. 1. An SEM image of pillar-type Si NWs on the silicon substrate ((a), top) and an optical microscope image of a single Si NW on a sapphire substrate, taken in our experimental system ((a), bottom) are shown. (b) A conceptual illustration of UOM with spatially separated pump and probe beams. (c) Photoinduced transmission change ($\Delta T/T$) for overlapping pump and probe spots on a single SiNW and a single SiO_2 encapsulated SiNW.

Photoinduced transmission changes were measured for a single core–only Si NW and a single Si/SiO_2 core/shell NW with overlapping pump and probe spots (Fig. 1(c)). The magnitude of the ΔT/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 (147 ps) than in the bare Si NW (~90 ps). This can be explained by the fact that surface passivation by SiO_2 reduces the surface trap density [3] and potentially also the surface recombination velocity [4]. The interface trap density at the surface in semiconductor core/shell NWs may generally be higher than in the bulk, but still significantly lower than in bare core-only NWs.

We then performed UOM experiments on the single Si NW and the single Si/SiO_2 NW while varying the spatial separation between the pump and probe beams, revealing a significant influence of SiO_2 passivation on the carrier dynamics (Fig. 2) along with strong oscillations due to acoustic phonons (also known as Brillouin oscillations [5]). When the pump pulse irradiates the NWs, it generates a longitudinal sound wave directed toward the substrate, which then diffracts the time-delayed probe pulse when it enters the sample, causing strong oscillations in the intensity of the transmitted probe. The Brillouin oscillation period τ can be expressed by $\tau = \lambda/2n\nu$, where the longitudinal sound velocity for Si is $\nu = 9.32\times10^3$ m/s and the refractive index is $n = 3.65$, giving an oscillation period τ =12.5 ps, which agrees well with our measured value of 12.9 ps. Because this acoustic oscillation is initiated by the photoexcited charge carriers, the oscillation periods are the same for both Si NWs, with or without the SiO_2 layer. In contrast, the carrier transport velocities are significantly modified by the oxidized SiO_2 layer. The carrier velocity for the bare Si NW, extracted by measuring the pump-probe separation and the rise time of the ΔT/T signal (Fig. 2(a)), is ~5×10^6 cm/s, while the carrier velocity for the Si/SiO_2 core/shell NW is ~2×10^6 cm/s (Fig. 2(b)). The difference in carrier transport velocity between the two NWs can be dramatically visualized in the insets to Fig. 2, which represent plots of the ΔT/T signal as a function of pump-probe separation and delay time. The slope of the maximum ΔT/T signal, with accompanying acoustic phonon oscillations, is much larger in the bare core-only sample.

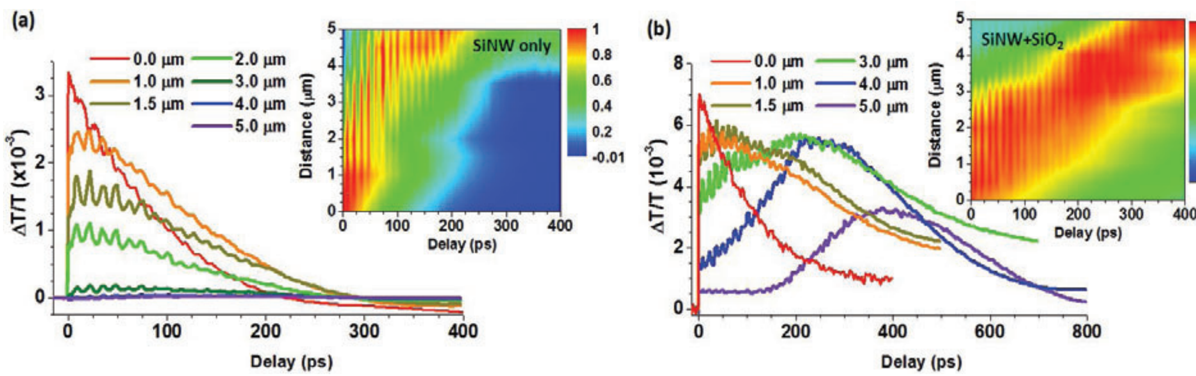

Fig. 2. Photoinduced transmission changes for various pump-probe separations for (a) the bare Si NW and (b) the SiO_2 encapsulated SiNW. The insets present two-dimensional maps of the normalized ΔT/T signals as functions of separation and time delay.

In conclusion, ultrafast optical microscopy opens new pathways to directly study carrier dynamics and charge transport in quasi-1D nanosystems. Here, we directly measured the carrier velocity and observed acoustic phonon oscillations in Si and Si/SiO_2 NWs for the first time by tracking carriers through space and time after femtosecond photoexcitation. This research has potential application to NW-based devices, optoelectronics, and sensitive photodetection by combining measurements at both nanometer distance and femtosecond time scales to reveal the intrinsic properties of these quasi-one-dimensional nanosystems.

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