Polarization anisotropy of transient carrier dynamics in single Si nanowires

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Abstract: We present the first ultrafast time-resolved, polarization-dependent experiments on both single- and ensemble-silicon nanowires using non-degenerate spectroscopy. Anisotropy was observed for polarizations perpendicular and parallel to the nanowire.

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In recent years, much interest has been shown in the optical properties of silicon (Si) nanowires (NWs) for their wide range of potential applications, particularly in enabling optoelectronic circuitry and functional devices on the nanoscale [1]. It is especially worth investigating the polarization dependence of light interactions with individual nanowires to utilize the unique directional optical properties available from these quasi-one-dimensional (1D) nanostructures. Recently, photoluminescence (PL) measurements on individual NWs have reported a strong dependence of the PL polarization on NW alignment [2]. Polarization anisotropy in the PL spectrum is produced by the 1D nature of the NWs, as well as their large dielectric contrast with the surrounding medium. However, there remains a lack of basic understanding on how light interacts with individual NWs on an ultrashort time scale, as the vast majority of ultrafast optical experiments on semiconductor NWs have been done on NW ensembles [3, 4]. Here, we have measured polarization-dependent ultrafast carrier dynamics on single Si NWs and Si NW ensembles using non-degenerate pump-probe spectroscopy to excite and probe carriers above the indirect band gap (E_g = 1.12 eV). In particular, the first measured polarization dependence of transient carrier dynamics in *single* Si NWs can give deep insight into the influence of light polarization on light-matter interactions in Si NWs.

The Si NWs used for this measurement were grown through vapor-liquid-solid (VLS) synthesis using chemical vapor deposition. The NWs were dry transferred onto a sapphire substrate with an areal density of ~5.5×10^6 NWs/cm^2 and diameters of d~50 nm (Fig. 1(a)). A nanomanipulator was then used to transfer an isolated Si NW with d=50 nm and length l=80 μm onto a patterned sapphire substrate (Fig. 1(b)). To measure the polarization-dependent transient carrier dynamics, the output of a 30 femtosecond (fs) Ti:sapphire laser oscillator is divided into pump and probe beams, with the probe power < 10% of the pump power (Fig. 1(c)). The pump beam is then frequency-doubled in a BBO crystal to generate 420 nm pulses. The magnitude of the photoinduced changes in probe transmission with 420 nm pump and 840 nm probe pulses were ~$\Delta T/T \sim 10^{-5}$.

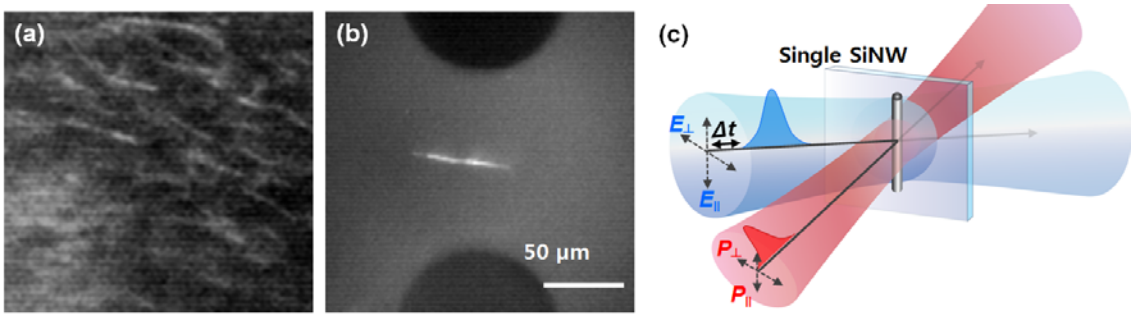

Fig. 1. Optical microscope images of d=50 nm Si (a) ensemble NWs and (b) single NWs (at the center). (c) A conceptual illustration of a polarization-dependent non-degenerate pump-probe measurement on a single Si NW in transmission.

Polarization-sensitive pump-probe experiments on Si NW ensembles (not shown here) reveal that the magnitude of the ΔT/T signal is maximum for both beams polarized parallel to the NW axis and minimum for both beams polarized perpendicular to the NW axis, as expected since light absorption is maximum for light polarized parallel to the NW axis [3,4]; however, there was no significant anisotropy in the measured relaxation times. The photoinduced change in transmission was then measured on the single NW sample (and compared to measurements on the NW

ensemble and bulk Si) to explore polarization-dependent carrier dynamics without the complications resulting from the broad NW size and alignment distribution in the NW ensemble (Fig. 2(a)). Curve fits (dashed lines in Fig. 2(a)) are used to extract a rise time of ~500 fs and a fast initial decay time of ~2 ps, followed by a ~30 ps slow decay. The faster decay process in the NW ensemble as compared to bulk silicon can be explained by the increased influence of surface traps and recombination centers as the degree of spatial confinement increases [5]. Carrier relaxation is even faster in the single Si NW measured here (Fig. 2(a)), although this will depend on the specific parameters (diameter, length, alignment) of a given NW.

Further polarization-dependent experiments on the single NW sample show the strong influence of the laser polarization on carrier relaxation (Fig. 2(b)). The transient transmission is maximum for both polarizations parallel to the NW axis, as in the NW ensemble. However, in stark contrast to our measurements on the NW ensemble, there is a clear anisotropy in the ultrafast dynamics measured for light polarized parallel and perpendicular to the long axis of the NW, with an overall faster decay for parallel polarizations, as shown in the inset. As mentioned above, more of the pump is absorbed for the parallel polarization, leading to a higher initial carrier density in this configuration. Density-dependent effects such as Auger recombination, which typically cause carrier lifetimes to decrease with increasing carrier density [5], may thus lead to the observed polarization dependence of the carrier dynamics.

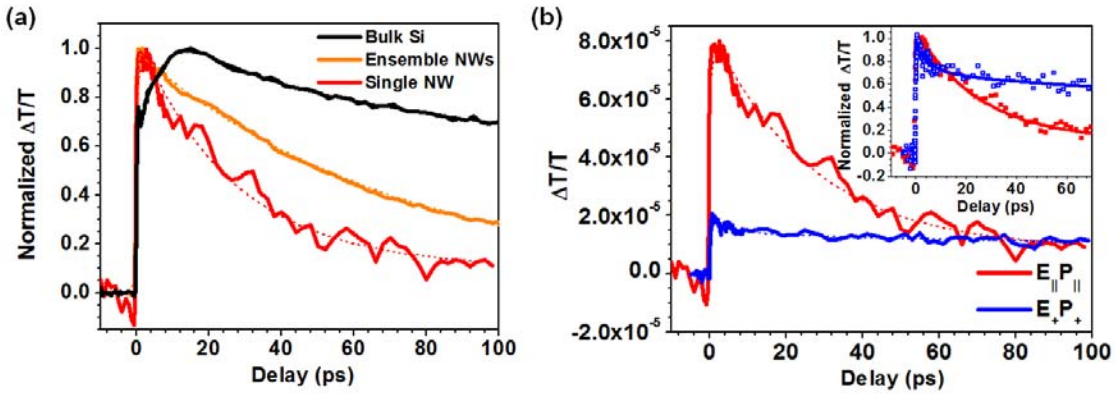

Fig. 2. (a) Comparison of the normalized ultrafast transmission measured in bulk Si, NW ensembles, and single Si NWs with pump and probe polarization parallel to the NW axis. (b) Pump-induced change in transmission for different pump and probe polarizations in a single SiNW. The normalized ΔT/T signals are shown in the inset.

In conclusion, the polarization dependence of the transient carrier dynamics was measured in Si NW ensembles and single Si NWs. The measured ultrafast changes in transmission on a single Si NW show that the carrier lifetimes and magnitude of the ΔT/T signal strongly depend on the pump and probe polarizations, unlike in NW ensembles. This underscores the importance of performing optical measurements on individual NWs to avoid ensemble broadening effects that can obscure the underlying physics. Finally, the observed anisotropy in single NWs could enable advanced applications, such as optical switching and polarization-sensitive photodetection, on the nanoscale, where directional control and high spatial resolution are much desired.

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