Visualization of Charge Carrier Motion in Semiconductor Nanowires with Ultrafast Pump-Probe Microscopy

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Abstract: Femtosecond pump-probe microscopy is used to directly visualize the diffusion of photogenerated charge carriers in undoped silicon nanowires, as well as charge separation in a nanowire encoded with an axial p-type/intrinsic/n-type (p-i-n) junction.
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1. Introduction

A detailed understanding of the factors that govern the motion of mobile charge carriers through nanostructures is critical to many emerging nanotechnologies in electronics, optoelectronics and solar energy conversion. While the motion of charge carriers at low carrier densities is uncorrelated and easy to understand, many active electronic components operate at high carrier concentrations resulting from heavy doping or high injection. In this regime, carrier-carrier interactions and other many body effects (e.g. dopant/carrier interactions, electron screening, and electron-hole scattering) must be considered. We have combined ultrafast pump-probe spectroscopy with optical microscopy [1, 2] to directly image the charge carrier dynamics in individual Si nanowires (NWs) with both spatial and temporal resolution.

2. Pump-Probe Microscopy

The femtosecond pump-probe microscope is illustrated in Fig. 1. In these experiments, an individual NW is excited by a 425 nm femtosecond pump pulse that has been focused to a diffraction limited spot (350 nm) by a microscope objective, producing photogenerated carriers (electrons and holes) in a localized region of the structure. After a well-defined delay, pump-induced changes to the transmission of an 850 nm probe pulse are detected.

Figure 1 shows pump-probe decay kinetics when the probe beam is spatially overlapped with the pump pulse. Transients for three NWs are shown with the location of the excitation depicted by red circles in their corresponding SEM images. The photoinduced transparency observed at early times arises from band filling by the photogenerated free carriers. It decays over several hundred picoseconds, and eventually changes sign to yield a low-amplitude absorptive component that persists beyond 1 ns and is attributed to a combination of trapped carriers and thermal effects. In this spatially-overlapped pump-probe (SOPP) configuration, the decay of the bleach reflects both

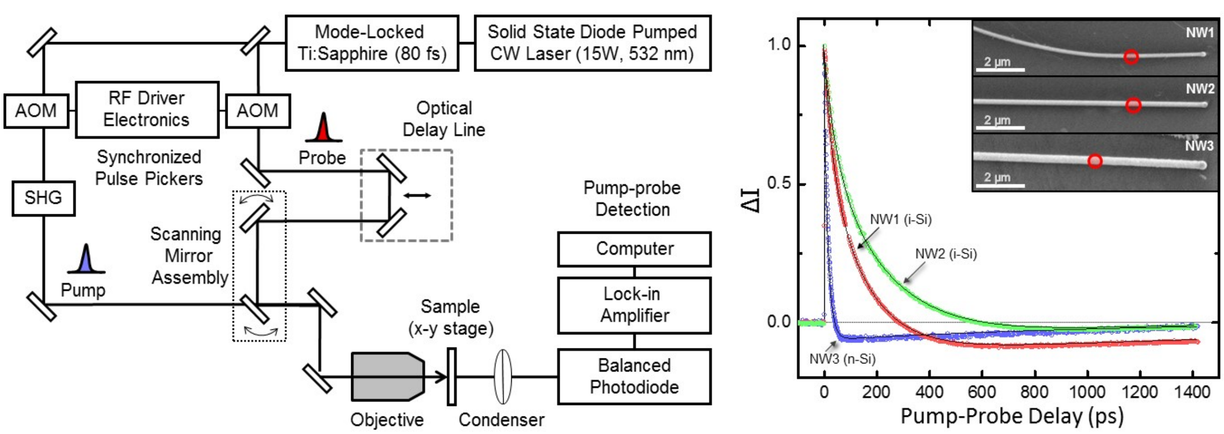

Figure 1. (Left) Schematic diagram of the pump-probe microscope. Pump (425 nm) and probe (850 nm) pulses are focused to diffraction limited spots by a microscope objective. A sample stage allows the two pulses to be overlap with a specific structure. A scanning mirror assembly scans the angle of the probe beam as it enters the objective, enabling it to be spatially separated relative to the pump pulse. (Right) Pump-probe decay kinetics obtained with spatially overlapped pump and probe beams following for three different Si nanowires; NW1 (red, 160 nm) and NW2 (green, 210 nm) are i-type, NW3 (blue, 330 nm) is n-type. All three were fit to a triexponential decay (solid lines). Inset: SEM images showing the location of pump excitation as a red circle; scale bars, 2 μm.

electron-hole recombination and migration of carriers away from the excitation spot. The recombination time (~200 ps) extracted from the bleach decay scales with NW diameter, consistent with surface mediated recombination.

3. Imaging Charge Carrier Motion

Motion of the photogenerated carriers is observed using a spatially-separated pump-probe (SSPP) configuration, in which carriers are created in one location and detected in another, allowing direct imaging of charge carriers as move away from the excitation spot. In this SSPP configuration the pump beam is held fixed and the position of the probe beam is scanned by varying the angle of the probe beam as it enters the objective (Fig 2A), resulting in a spatial map of the photoinduced transparency (and the free carriers) at a specified pump-probe delay. Images collected at a series of delays shows the spatial-temporal evolution of the charge cloud following excitation, providing a direct visualization of carrier diffusion in undoped NWs and charge separation in Si p-i-n junctions.

Spatially-separated pump-probe images obtained on undoped NWs are shown Fig. 2B [1]. The location of the pump pulse excitation is indicated by the red circle in the scanning electron microscopy (SEM) image at the left. The spatial extent of the photoinduced transparency created by the focused pump pulse is depicted by the red colors in the 0 ps frame. At later times, this transparency elongates, reflecting free carrier migration along the NW axis. The growth of the charge cloud is consistent with ambipolar diffusion in bulk Si. The blue spot that appears at longer times reflects the long-lived transient absorption (Fig. 1), which remains localized at the excitation spot.

Charge separation is apparent in SSPP images obtained from Si NWs encoded with axial p-type/intrinsic/n-type junctions (Fig. 2C). Here photoexcited carriers are produced at the center of the intrinsic region. Initially the carrier motion is dominated by carrier-carrier interactions, which opposes charge separation and causes the charge cloud to spreads in both directions. As it spreads, the carrier density decreases due to electon-hole recombination. By 300-400 ps it reaches the boundaries of the intrinsic segment, at which point its evolution becomes asymmetric. Over the next 200 ps, a bleach appears in the n-type region that then persists well beyond 1 ns, indicative of the formation of a long-lived charge separated state.

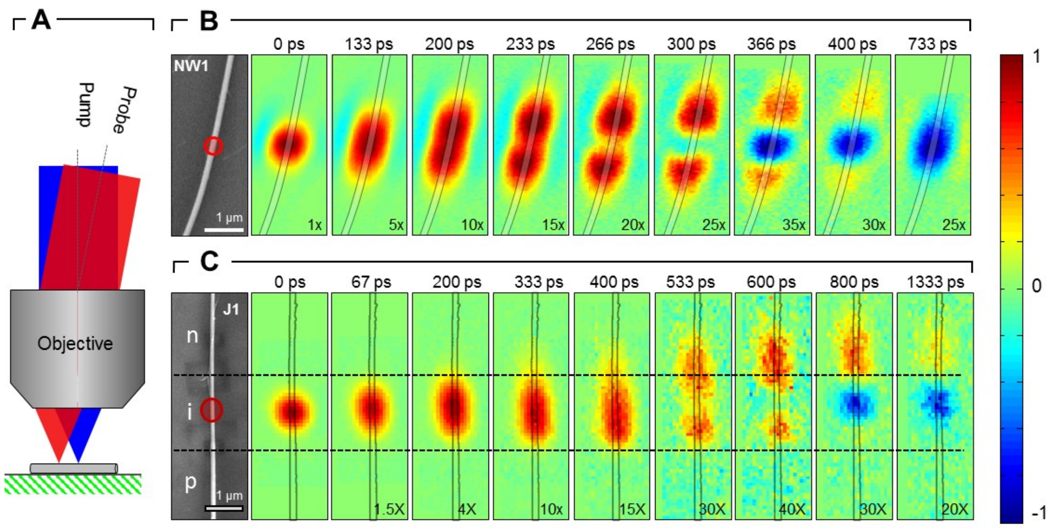

Figure 2. Imaging of charge carrier motion in individual Si nanowires. (A) Schematic illustration of spatially separated scanning. (B) SSPP microscopy images depicting carrier diffusion in an undoped Si nanowire. Left, SEM images of the NW with the location of the excitation spot depicted by the red circle. Right, series of SSPP images acquired at the delay times denoted above each image. Location of the nanowire is depicted by the faint lines. Each image is depicted using a normalized color scale with relative amplitudes indicated by scaling factors in the bottom-right corner. (C) SSPP images showing charge separation in a Si NW encoded with an axial p-i-n junction. The n-type region is in the upper portion; the p-type is in the lower. Dashed lines denote the boundaries of the doped regions. Excitation is in the middle of the intrinsic segment, at the location of the red circle.

4. References

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