Tracking Carrier Dynamics in Semiconductor Nanostructures Through Space and Time

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Scientific Thrust Area: Nanophotonics and Optical Nanomaterials

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Research Achievement: Semiconductor nanostructures have attracted much recent attention due to the unique size-dependent scaling of their physical properties, providing researchers an opportunity to tune these parameters for applications in areas ranging from medicine to solar energy. Many of these applications depend critically on a detailed knowledge of the response of these nanosystems to photoexcitation on a femtosecond time scale. Ultrafast optical spectroscopy (UOS) is the only technique capable of resolving dynamics in conventional metals and semiconductors at the fundamental time scales of electron and lattice motion.[1] Therefore, the application of UOS to probing the dynamic response of semiconductor nanostructures after femtosecond excitation will increase our understanding of their physics and enable their optimization for applications. In this work, we present ultrafast wavelength-and-time-resolved spectroscopic experiments on one-dimensional semiconductor nanowires (NWs) and quantum dots-in-a-well (DWELL) heterostructures, as well as the first demonstration of a new technique, ultrafast optical wide field microscopy, for performing space-and-time-resolved measurements.

Our density-dependent differential transmission (DT) experiments on a DWELL heterostructure, which consists of InAs quantum dot (QD) layers embedded in InGaAs quantum wells (QW) that are subsequently embedded in bulk GaAs, are particularly relevant to lasing applications.[2] The most unique feature of our data is the negative DT signal ($\Delta T/T<0$) observed at the QD excited state (λ~1047 nm) at high densities (Fig. 1); the low density dynamics have been previously described.[3] Comparison of the positive DT signal from the QW (not shown) with the negative part of the DT signal at the QD excited state reveals that the QW population strongly influences light absorption at the QD excited state through Coulomb interactions between the photoexcited carriers, which has not previously been observed.

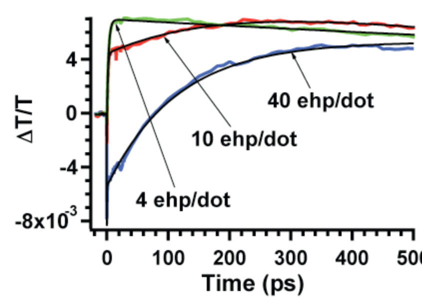

Fig. 1. Density-dependent 800 nm pump, 1047 probe measurements on a DWELL heterostructure.

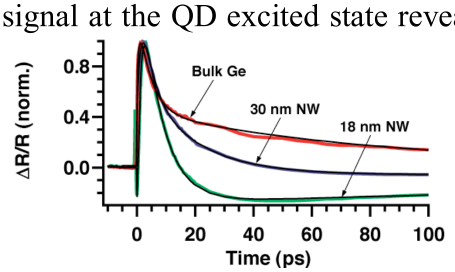

Fig. 2. Optical pump-probe measurements on Ge NWs as a function of NW diameter.[5]

Optical pump-probe experiments on semiconductor nanowires reveal novel effects resulting from the enhanced influence of surface and defect states. An example is shown in Figure 2, revealing that carrier lifetimes increase with diameter in Ge NWs due to trapping at surface states.[5]

Similar experiments performed on GaN NWs demonstrated the ability to tailor the influence of defect states on carrier relaxation through control of the growth temperature. Ultrafast optical spectroscopy thus enables us to shed light on carrier relaxation in these one-dimensional systems.

Finally, we have recently demonstrated an ultrafast optical microscope that combines the spatial resolution of wide field optical microscopy with the temporal resolution of ultrafast optical spectroscopy to rapidly and sensitively acquire images of nearly any sample with high sensitivity, temporal, and spatial resolution. This unique tool is based on a two-dimensional smart pixel array detector that separately demodulates the signal on each pixel, allowing us to detect transmission changes as small as $\Delta T/T \sim 10^{-5}$. We have acquired time-resolved images of a gold patterned Si film (Figure 3) to demonstrate this powerful concept for the first time.

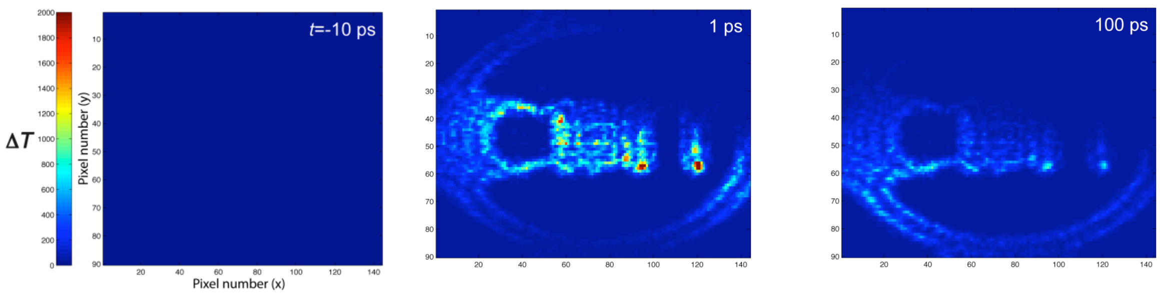

Fig. 3. Ultrafast optical wide field microscope images at different time delays between the pump and probe.

Future Work: We plan to extend our measurements of carrier dynamics in DWELL heterostructures to pump different quantized energy levels over a wide density and temperature range. In addition, we have recently developed a mid-infrared pump, terahertz (THz) probe system that will enable us to simulate photodetector operation with femtosecond time resolution. We will also perform optical-pump THz-probe experiments to measure the photoinduced conductivity in semiconductor NWs. Measurements on radially heterostructured NWs will also enable us to control the influence of surface states on the measured dynamics. Finally, we will apply ultrafast optical wide field microscopy to a number of materials, ranging from nanowires to neurons, to unravel coupled space-time dynamics in these complex systems.

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Publications:

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