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Visualization of Transport Dynamics in Nanostructures with Pump-Probe Microscopy.

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A detailed understanding of the factors that govern the transport of mobile charge carriers, acoustic phonons and thermal energy in nanostructures is critical to many emerging nanotechnologies in electronics, optoelectronics and solar energy conversion. We have combined ultrafast pump-probe spectroscopy with optical microscopy to directly image the transport phenomena in individual Si nanowires (NWs) with both spatial and temporal resolution. 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, exciting 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, providing the time evolution of the photoexcitation at a specific point within the structure. By correlating optical images with scanning electron microscopy images obtained from the same structures, we are able to correlate recombination behavior with specific structural features. Motion of the photogenerated carriers, propagation of acoustic modes and thermal transport are observed using a spatially-separated pump-probe configuration, in which carriers are created in one location and detected in another, allowing direct imaging of charge carriers and phonons as they move away from the excitation spot. In this 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, resulting in a spatial map of the photoinduced transparency at a specified pump-probe delay. Images collected at a series of delays shows the spatial-temporal evolution of the excitation, providing a direct visualization of carrier diffusion, acoustic mode propagation and thermal transport in semiconducting NWs.