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Spectroscopy of Optical Excitations in Carbon Nanotubes

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Understanding the optical spectra and electronic excited state dynamics of carbon naotubes is important both for fundamental research and a wide variety of potential applications. In this presentation, we will report the results of a systematic study on semiconducting single-walled carbon nanotubes (SWNTs) obtained by utilizing complementary femtosecond spectroscopic techniques, including fluorescence up-conversion, frequency-resolved transient absorption, and three-pulse photon echo peakshift (3PEPS) spectroscopy. Our efforts have focused on optically selective detection of the spectra and dynamics associated with structurally distinct semiconducting SWNT species. Using individual nanotube enriched micelle-dispersed SWNT preparations, in combination with resonant excitation and detection, has enabled us to independently access selected species, such as the (8,3), (6,5), (7,5), (11,0), (7,6) and (9,5) nanotubes. We will discuss the following topics: (1) the excitonic nature of the elementary excitation and its unambiguous identification from direct determination of the exciton binding energy for a selected semiconducting nanotube, the (8,3) tube; (2) the spectroscopic and dynamical signatures of exciton-exciton annihilation and its predominant role in governing ultrafast excited state relaxation; (3) the annihilation-concomitant exciton dissociation and the spectroscopic and dynamic features of the resulting electron-hole continuum; (4) timescales characterizing the ultrafast thermalization processes. In addition, we will demonstrate the power of 3PEPS spectroscopy to elucidate the spectral properties and dynamics of SWNTs. This work was supported by the NSF.