Non-Radiative Exciton Decay in Single-Walled Carbon Nanotubes

MARK HARRAH, JUDE SCHNECK, Boston University, ALEXANDER GREEN, Wyss Institute, Harvard University, MARK HERSAM, Northwestern University, LAWRENCE ZIEGLER, ANNA SWAN, Boston University — We report on the exciton dynamics for an ensemble of individual, suspended (6,5) single-walled carbon nanotubes via single color $E_{22}$ pump-probe spectroscopy for a wide range of pump fluences. The calculated initial exciton population ranges from $\sim 5$ to $120$ excitons per $\sim 725$ nm long nanotube, putting the high fluence experiment well into the nonlinear regime. The pump-probe data is not well described by multi-exponential decay or by power law behavior for all fluences. We have developed a single model that describes all data, ranging over two decades of pump fluence and three decades of delay times. The signal decay at low fluence is dominated by a stretched exponential that is consistent with the distribution of relaxation rates resulting from diffusion-limited contact quenching for a nanotube ensemble. The change in dynamics as a function of increasing pump intensity is attributed to exciton-exciton Auger de-excitation in the $E_{11}$ subband and, to a lesser extent, in the $E_{22}$ subband. The initial sub-picosecond decay of the observed response is attributed to $E_{22}$ excitons rapidly acquiring non-zero momentum while remaining in the $E_{22}$ subband.