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Two Dimensional White Light Spectroscopy Reveals Energy Transfer Pathways in Semiconducting Carbon Nanotube Thin Films

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UW-Madison — Carbon nanotubes are promising materials for the active layer in photovoltaic devices because of their tunable bandgaps and large exciton diffusion lengths. We are studying thin films of coupled semiconducting nanotubes to understand the dynamics of exciton transfer. Previous work used transient absorption to follow photoexcitation transfer from large to small bandgap nanotubes, but a comprehensive mechanism could not be obtained, largely due to the wide range of wavelengths over which these films absorb. We have developed two-dimensional white light spectroscopy (2D WL) as a novel probe of these films and many other systems in the solar energy sciences. By studying the evolution (as a function of waiting time) of crosspeaks between the E_{ii} states for different bandgap nanotubes, we are able to map out the energy transfer pathway. The advantage to using 2D WL over traditional 2D electronic spectroscopies is that the spectral bandwidth produced from supercontinuum generation is significantly larger than that accessible from an optical parametric amplifier. Thus, we are able to cover the entire film absorption simultaneously, thereby obtaining a map of the instantaneous exciton distribution. Several surprising results will be reported.

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