Migration and Dissociation of Excitons in Photoabsorbing Thin Films of Carbon Nanotubes Tailored for Photovoltaics

Meng-Yin Wu[1], Dominick J. Bindl[2], Randy D. Mehlenbacher[3], Maksim Grechko[3], Martin T. Zanni[3], Michael S. Arnold[2,*]

[1]Department of Electrical and Computer Engineering,
[2]Department of Materials Science and Engineering,
[3]Department of Chemistry,

University of Wisconsin-Madison,
Madison, WI 53706, U.S.A.

Semiconducting carbon nanotubes are attractive materials for the light absorbing components of solar cells and photodetectors because they offer strong, tunable optical absorptivity; ultrafast transport; and the potential for stability and solution-processability. Bilayered donor / acceptor heterojunction devices based on carbon nanotube photoabsorbers as the donors have already been realized with >1.0% AM1.5G solar power conversion efficiency, in which the photoresponse is primarily derived from the nanotubes. While these devices are exciting proofs-of-principal, there is still a need to improve their efficiency and to do to so across a broader spectrum. These improvements can be achieved in part by better understanding the photophysical responses and behaviors of mixtures of carbon nanotubes that are coupled to one-another in thin films.

Along these lines, here, we report on our recent investigations of the migration and dissociation of excitons in photoabsorbing thin films of coupled carbon nanotubes tailored for photovoltaics. We show that the performance of bilayered solar cells is limited by slow and relatively short-range inter-nanotube exciton energy transfer. In the bilayered cells, the internal quantum efficiency for photon to collected electron conversion efficiency falls off for nanotube film thicknesses beyond 5 nm, suggesting a short inter-nanotube exciton diffusion length ~ 5 nm. We have characterized inter-nanotube exciton migration in more detail using polarization dependent transient absorption spectroscopy. We observe two timescales for photoexcitation transfer. First, we observe that the selective photoexcitation of large bandgap nanotubes results in fast (< 500 fs) and long-range (> 5 nm) transfer of ~50% of the initial photoexcitation to surrounding small bandgap species, with a variety of orientations. However, the other ~50% of the photoexcitation evolves from larger to smaller bandgap nanotubes on a second, slower timescale more similar to the exciton non-radiative lifetime (10's of ps). Based on these data, we have implemented kinetic Monte Carlo models to evaluate possible bandgap distributions and nanotube morphologies for maximizing the transport of excitons to acceptors in bilayered and blended carbon nanotube / acceptor structures.