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Deterministic Modelling of Carbon Nanotube Near-Infrared Solar Cells

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With solution-process-ability, scale-able fabrication and purification, and cheap input materials, semiconducting single-walled carbon nanotube (SWNT) networks represent promising materials for near-IR solar cell (SC) applications. This promise has motivated a body of work not only developing solar cells but also exploring alignment/deposition methods and SWNT photovoltaic (PV) physics. Despite this interest, there is to date no quantitative model of SWNT solar cell operation analogous to bulk semiconductor p-n junction PVs, allowing a rigorous understanding of the physical tradeoffs driving experimental observations and informing what research will enable technological progress. In this work we have derived the steady state operation of planar heterojunction SWNT PVs from the fundamental light absorption, exciton transport, and free carrier transport behaviors of single nanotubes. Our method can treat arbitrary distributions of nanotube chiralities, lengths, orientations, defect types and concentration, bundle fraction and size, density, dielectric environment, electrode combinations, etc. We achieve this by treating individual SWNT properties as random variables, and describing the network by the dependent distributions of those properties, yielding coupled stochastic differential equations for light absorption, exciton transport, and free carrier transport. Applying the model to monochiral (6,5) films in aligned and isotropic configurations, we find that there is a strongly optimal film thickness at a given nanotube network density and orientation, reflecting an inherent tradeoff between light absorption (i.e. exciton generation) and diffusion to the electrodes. This optimal shifts lower with increasing density, and is ultra-thin ($<$10 nm) for horizontally-aligned films but 50-200 nm for vertically aligned films. We show that due to weak inter-SWNT exciton transport relative to exceptional intra-SWNT diffusion, vertically-aligned films are unambiguously favored at densities above 3% of close-packed; at lower densities however an optimum emerges at an intermediate angle to compensate for weaker light absorption of vertical nanotubes. Films with in-plane aligned nanotubes are unambiguously the worst design.