DECaNT Numerical Tool for Exciton Dynamics in Carbon Nanotube Films

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ABSTRACT

Photons incident on carbon nanotube (CNT) films tend to generate tightly bound electron-hole pairs known as excitons. We present a simulation tool for studying exciton diffusion through CNT arrays, based on a microscopic theory of exciton transfer between pairs of CNTs. Utilizing a Monte Carlo algorithm, we can study the diffusion behavior of exciton ensembles based on array properties such as density, composition, and morphology.

INTRODUCTION

Carbon nanotubes are a promising material for several applications, notably including solar-energy harvesting. The process by which solar photons can be converted into useful energy involves absorption by the carbon nanotube, which creates bound electron-hole pairs known as excitons. Excitons must diffuse through the array of CNTs to a harvesting layer, where the electron and hole dissociate and the charge can be captured. The exciton diffusion process is therefore of the utmost importance for understanding the efficiency of energy harvesting in CNT devices. Experimental investigations into exciton diffusion have been fruitful, but here we showcase our unique simulation tool that allows tremendous control over the properties of the CNT array, which can help us to isolate and study the effects of different parameters on exciton diffusion through realistic films [1].

SIMULATION TOOL

We begin the simulation process by generating a realistic 3D array (or “mesh” or “film”) of hundreds to thousands of CNTs. We generate each CNT in several short segments, which are connected with constraints that allow rotation but keep the segments together, like a ball and socket joint. A group of CNTs is generated and then released from the air into a container of specified size, where they dynamically settle according to Newton’s laws. This dynamic generation process is achieved using the Bullet Physics software library [2]. A snapshot of the mesh generation process is shown in Fig. 1.

We use a Python script to take the coordinates of each tube segment from the mesh generation process and interpolate along the length of each CNT to increase the density of points.

The new set of points, along with the CNT properties (chirality, which determines bandstructure, and relative orientation) are input to a Monte Carlo algorithm. The Monte Carlo code utilizes scattering rates calculated from a detailed microscopic theory [3] of transfer between pairs of CNTs to simulate the movement of thousands of excitons through the array. By studying the aggregate behavior, we discover the diffusion properties of excitons throughout the array. In Fig. 2, we show the diffusion-tensor elements for an array of hundreds of CNTs with a single chirality (this particular chirality is commonly denoted as [4,2]). We note that there is about one order of magnitude difference between in-plane and out-of-plane diffusion constants.

Our mesh generation tool allows us to control several variables—such as film thickness, intertube spacing (Fig 3.) dielectric environment, and array morphology—and study their effect on exciton diffusion. The simulation tool is open source and free to use.

ACKNOWLEDGMENT

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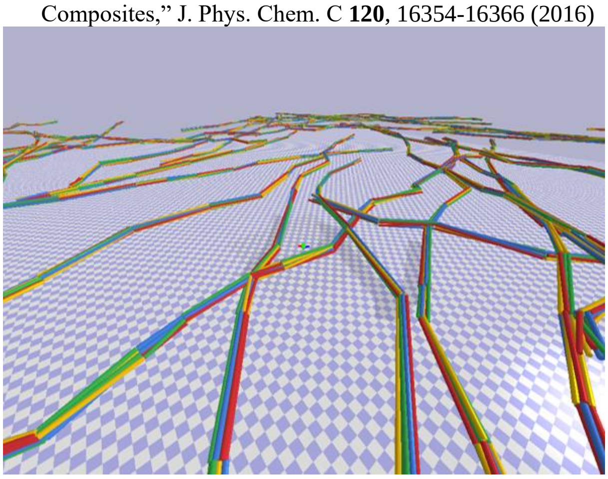

Fig. **1**. Snapshot of the CNT mesh generation process using Bullet Physics [1-2]. Each CNT is composed of several segments that are connected together via cone constraints.

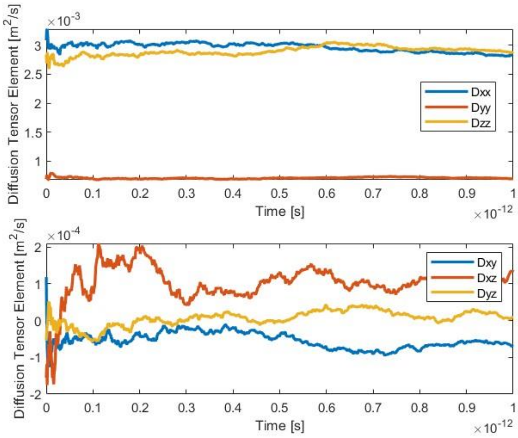

Fig. **2**. Mean squared displacements vs. time for each of the components of the diffusion tensor. The diffusion tensor is the long time limit of these plots.

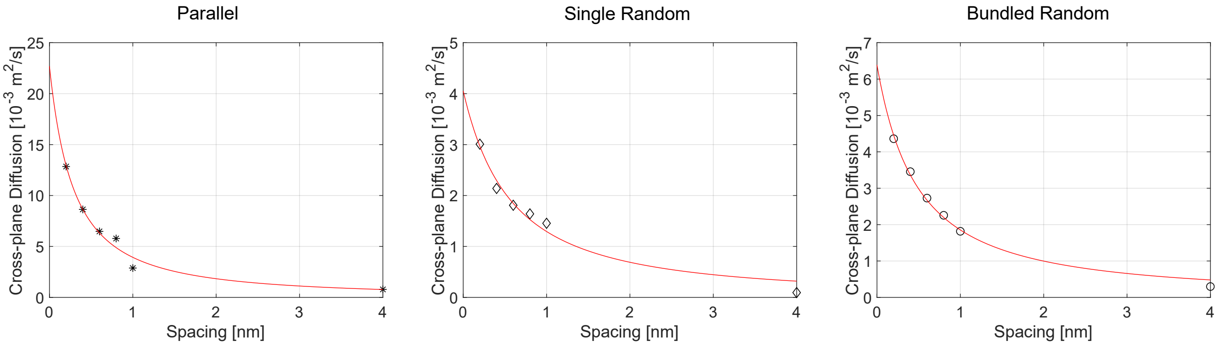

Fig. 3. Effect of additional intertube spacing on cross-plane diffusion constants for three different morphologies of CNT array. Parallel refers to aligned CNTs, "Single Random" refers to randomly oriented unbundled CNTs, and "Bundled Random" refers to randomly oriented bundles of seven CNTs (hexagonally packed) [1].