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Effects of randomness and inclination on the optical properties of multi-walled carbon nanotube arrays

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ABSTRACT

The optical properties of multi-walled carbon nanotube arrays are investigated using the finite-difference time-domain method, focusing on the effects of various structural randomness, including random position, diameter, length, and orientation. It is found that the arrays with random position, diameter, length, and small inclination angle have quite small absorption enhancement compared to the ordered arrays. The reflection spectra of the arrays with random position, diameter, and small inclination angles are almost identical to the ordered array, but large reflection suppression is seen in vertical arrays with random length. For oblique arrays, the absorptance increases with inclination angle for S-polarized light, but weakly depends on inclination angle for P-polarized light. By comparing the inclined arrays with different volume fractions, the reflectance is found to be largely determined by the local volume fraction of the top surface of carbon nanotube arrays.

1. Introduction

Carbon nanotubes (CNTs) have attracted considerable interest since their discovery in 1991 [1], because of their unusual electrical, mechanical, thermal, and optical properties. Optical properties of CNT arrays are especially interesting as CNTs are strongly anisotropic [2,3] and highly absorbing in the visible and infrared regions. Low density arrays of vertical CNTs have low effective index of refraction as well as numerous nanocavities that can effectively trap light, making them superior light absorbers [4–6]. For example, Yang et al. have reported multi-walled (MW) CNT arrays with reflectance as small as 0.045% in the visible range [4]. Wang et al. have shown that the optical reflectance of CNT array is about 0.5%, and can be either highly diffuse or specular, depending on the details of fabrication process [7]. Such high absorbing materials can be potentially useful in optical applications, such as radiometric temperature measurements [8] and fast-switching blackbody sources [9].

There have also been many theoretical investigations on the optical properties of CNT arrays. Calculations based on the density-functional theory have shown that the optical properties of small single-walled (SW) CNTs are strongly dependent on the chirality of each individual CNT [10]. However, when the CNTs are arranged in an array manner, the array usually contains CNTs with many different chiralities [11]. On the other hand, a single multi-walled (MW) CNT is composed of multiple concentric SWCNTs. Therefore, in most theoretical analysis, CNT arrays (regardless SW or MW) have been treated as an effective medium that contains many rolls of graphite. Some numerical electromagnetic calculations have also been carried out. For example, Lidorikis et al. used the finite-difference time-domain (FDTD) calculations for infinitely long MWCNT arrays with irradiation incident from one side of the array [12]. They predicted the CNT arrays to be good absorbers in the visible band and photonic crystals in ultraviolet band. In our earlier work, FDTD simulations are employed to calculate the optical properties of CNT arrays, with light incidence from the top [13]. We have demonstrated that the effective medium

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theory cannot fully capture the wave effect so that the prediction from such theory is not accurate enough [13]. It has also been shown that such arrays are nearly perfect absorbers, which is qualitatively consistent with the experimental results [13]. Quantitative comparison is still a challenge because structural randomness is inherent to any CNT array fabrication process, and the effects have not been included in previous theoretical studies. It becomes essential to uncover whether structural randomness will deteriorate the optical reflection or absorption. Recent investigations on silicon nanowire arrays show that small randomness such as random position, diameter, and length can greatly enhance the optical absorption of the arrays [14]. Compared to silicon, MWCNTs are anisotropic in their optical properties and are much more absorbing in nature, and the effects of structural randomness may show different features.

In this study, the effects of various structural randomness for CNT arrays, i.e., random position, diameter, length, and orientation, are investigated systematically using the FDTD method. The aligned CNTs with different inclination angles up to 60° are also studied. The results are compared with the optical properties of those ordered vertical arrays.

2. Simulation details

The different structures studied in this work are illustrated in Fig. 1(a). The ordered array contains 16 CNTs with 40 nm diameter and 1500 nm length. For all the simulations with structural randomness or CNT inclination, we choose a similar structure which contains 16 individual CNTs. No substrate is placed below the array, so we are actually investigating a film that contains an array of CNTs. The dielectric function of CNTs is obtained from an averaging of the dielectric function of graphite, as in our previous work [13]. The various random arrays are generated so that they have comparable volume fraction with the ordered array, and the details will be provided in subsequent discussions.

Fig. 1(b) is an illustration of the vertical cross section of the simulation domain. The 3D simulation domain is a tetragonal shape of 400×400 nm in the xy plane, and 3100 nm in the z direction. Periodic boundary condition is applied in the x and y directions and the perfectly matched layer (PML) is used to truncate the z direction. A Gaussian source is placed at 1500 nm above the upper surfaces of the arrays. Two power monitors placed above and below the array are used to measure the transmitted and reflected flux values which are then normalized by the source power to obtain the reflectance and transmittance spectra. The absorptance spectra are then calculated using $A = 1-T-R$, where T and R are the transmittance and reflectance from the simulation, respectively. Such a simulation setup is commonly used to model plane waves incident on a structure [13,15,16]. For all cases the reflectance and transmittance are calculated for 100 different wavelengths between 400 and 850 nm which cover the entire visible spectrum. All the simulations are performed with Lumerical FDTD solutions package. Automatic adaptive mesh with the highest possible accuracy is used for spatial discretization of the simulation domain.

Applying periodic boundary condition to this domain will generate pseudo-random configurations. These configurations are then used in the simulations to find out their optical properties. To ensure that our pseudo-random structures can capture the essential physics of real structural randomness, we have tested larger simulation domains with 800×800 nm or 1600×1600 nm size which contain 64 or 256 CNTs for the array with random position, and the results are similar. Also, ten randomly generated configurations are sampled for each types of randomness, and the transmittance and reflectance values are the average of the ten configurations.

3. Results and discussion

3.1. Effects of random position, diameter, and length

To generate CNT arrays with random position, the xy plane of the simulation domain is divided into sixteen 100×100 nm cells. One CNT with 40 nm diameter and 1500 nm length is

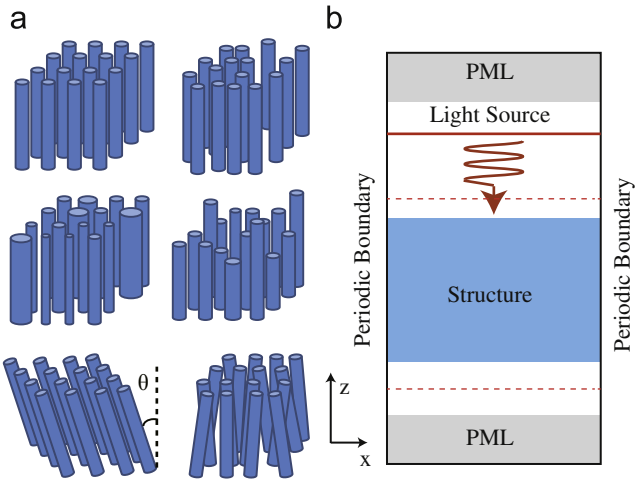

Fig. 1. (a) The various CNT arrays studied in this work, including ordered array, random position, random diameter, random length, oblique array, and random small inclination angle. (b) A cross section of the simulation setup. Periodic boundary conditions are applied at x and y directions. Two dashed curves indicate the flux monitors to calculate the reflected and transmitted flux.