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FEM MODELING OF PERIODIC ARRAYS OF MULTI-WALLED CARBON NANOTUBES

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Abstract—Multiwalled carbon nanotubes display dielectric properties similar to those of graphite, which can be calculated using the well known Drude-Lorentz model. However, most computational softwares lack the capacity to directly incorporate this model into the simulations. We present the finite element modeling of optical propagation through periodic arrays of multiwalled carbon nanotubes. The dielectric function of nanotubes was incorporated into the model by using polynomial curve fitting technique. The computational analysis revealed interesting metamaterial filtering effects displayed by the highly dense square lattice arrays of carbon nanotubes, having lattice constants of the order few hundred nanometers. The curve fitting results for the dielectric function can also be used for simulating other interesting optical applications based on nanotube arrays.

1. INTRODUCTION

Multiwalled carbon nanotubes (MWCNTs) were first discovered by S. Iijima in 1991 [1] and have since then been the focus of enormous research. MWCNTs are structurally similar to a concentric array of single graphite sheets wrapped into cylindrical tubes [2]. They are mostly metallic and are able to carry high current densities. Due to their high electrical conductivity and aspect ratios they produce very strong electric fields, which makes them an efficient source of electrons, as has been demonstrated in field emission displays [3].

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Arrays of MWCNTs have been used as electrodes in rectifiers [4], X-ray sources [5], gas sensors [6], as nano-electrodes for liquid crystal based electro-optical devices [7], and other electrical applications. Carbon nanotubes are very promising materials for device fabrication because of their excellent electrical, thermal and mechanical properties in addition to high aspect ratio and resistance to chemical and physical attack.

Multiwalled carbon nanotubes also have nanoscale dimensions comparable to the wavelength of light. Due to this they present a strong optical interaction which has been exploited by numerous photonic applications. Interesting optical properties of carbon nanotubes have been utilized in solar cells and other photovoltaic devices for increasing the efficiency [8]. Nanotube arrays have also been characterized as optical antennas [9], metamaterials [10] and photonic crystals [11]. These applications require precise positioning of the nanotubes having well defined dimensions. As the CNT fabrication requires an expensive and complex process [12] therefore modeling of these devices becomes an essential task for their further development. We present in this paper the finite element modeling (in a widely used commercial software COMSOL Multiphysics®) of wave propagation through the two dimensional (2D) periodic arrays of multiwalled carbon nanotubes. The incorporation of the frequency dependent and anisotropic optical properties of CNTs into the commercial softwares is a difficult task. The dielectric properties of CNTs, defined by the Drude-Lorentz model, were incorporated into the simulations by using the polynomial curve fitting technique. A high density periodic array was modeled (with lattice constant of 400 nm and nanotube radius of 50 nm) and an artificial metamaterial filtering effect was observed. Highly dense periodic arrays of CNTs displayed an artificial plasma like behavior with a lower plasmon frequency (which dictates the lowest transmission frequency). The reported results can also be utilized for analyzing other optical applications based on the MWCNT arrays.

2. OPTICAL PROPERTIES OF MWCNTS

The optical properties of multiwalled carbon nanotubes have been widely researched and their study is imperative before exploring their photonic device applications. MWCNTs are strongly anisotropic in nature [13, 14] and highly nonlinear [15] because they are structurally similar to rolled up graphite sheets. In graphite the carbon atoms are arranged in almost parallel layers. In each layer, carbon atoms form a network of regular hexagons. Since the distance of the neighboring layers is almost 2.7 times greater than that between the two nearest neighboring carbon atoms in one layer, a large anisotropy in structural, electronic and optical properties exists. It has been reported that the general optical features observed for MWCNTs are very similar to those of bulk graphite [15, 16]. Optical properties of graphite are highly anisotropic, with different dielectric response functions $\varepsilon_{//}(\omega)$ and $\varepsilon_{\perp}(\omega)$ depending on the electric field being polarized along or perpendicular to its *c*-axis. *C*-axis is a symmetry axis perpendicular to the basal plane of the graphite layers. Graphene, i.e., a single graphite layer, has also been shown to have a similar optical response to bulk graphite for light incident normal to its plane and polarized along it [17]. If we assume the curvature effects in the CNTs to be small and the local dielectric function to be well described by that of graphite, for electric field polarized parallel to the CNT axis we have only the $\varepsilon_{\perp}(\omega)$ contribution to consider, while for perpendicular we need to consider both $\varepsilon_{\perp}(\omega)$ and $\varepsilon_{//}(\omega)$.

The in-plane dielectric function $\varepsilon_{\perp}(\omega)$ for the MWCNTs or graphite can be obtained through the widely used Drude-Lorentz model. According to the model the dielectric function $\varepsilon_{\perp}(\omega)$ can be expressed as

$$\varepsilon_{\perp}(\omega) = \varepsilon_{\perp}^{f}(\omega) + \varepsilon_{\perp}^{b}(\omega) \tag{1}$$

where $\varepsilon_{\perp}^{f}(\omega)$ represents the intra-band effects (also referred as free electron effects) and $\varepsilon_{\perp}^{b}(\omega)$ represent the inter-band effects (usually referred as bound-electron effects). The intra-band part $\varepsilon_{\perp}^{f}(\omega)$ of the dielectric function is described by the free-electron or Drude model

$$\varepsilon_{\perp}^{f}(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega/\tau} \tag{2}$$

where ω_p and τ are free electron plasma frequency and relaxation time. The inter-band contribution $\varepsilon_{\perp}^{b}(\omega)$ to the dielectric function is described by the modified Lorentz model. This model assigns oscillators to major critical points in the joint density of states corresponding to inter-band transition energies $\hbar\omega_m$ with some additional oscillators modeling the absorption between the critical points. Each oscillator is characterized by its oscillator strength σ_m, the damping constant γ_m and frequency ω_m. The contribution of the inter-band transitions is given by

$$\varepsilon_{\perp}^{b}(\omega) = \sum_{m=1}^{M} \frac{\sigma_m^2}{\omega_m^2 - \omega^2 - i\omega\gamma_m} \tag{3}$$

where m is the number of damping oscillators employed in the model. The effective dielectric expression for the Drude-Lorentz model [16] can be written as

$$\varepsilon_{\perp}(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega/\tau} + \sum_{m=1}^{M} \frac{\sigma_m^2}{\omega_m^2 - \omega^2 - i\omega\gamma_m} \qquad (4)$$

Several groups have proposed data for the effective optical properties of MWCNTs which can be fitted with a Drude-Lorentz model. The data presented in reference [16] displays an excellent fit between the experimentally measured graphite dielectric function and the calculated model while using $M = 7$. Using these model parameters, the Durde–Lorentz model was established in MATLAB and the CNT dielectric constant was obtained for the optical and near infrared region, as shown in Figure 1. The complex dielectric function comprised of both a real and an imaginary part.

3. POLYNOMIAL CURVE FITTING

For the CNT simulations performed at a single frequency, the corresponding value for $\varepsilon_{\perp}(\omega)$ can be calculated from the model and incorporated into the simulation as the materials properties of CNTs. However, for the simulations designed for studying the optical transmission, band gaps and other photonic properties of carbon nanotubes, $\varepsilon_{\perp}(\omega)$ (the function of frequency) has to be completely incorporated into the simulation, so that its value can be calculated over the whole frequency spectrum. From Equation (4) for $\varepsilon_{\perp}(\omega)$, it can be observed that for calculating the second Lorentz term a loop of M iterations is required. Such calculation is trivial for computational softwares like MATLAB. However, most commercially available electromagnetic computational softwares lack the capacity of performing the iterative loop calculation for obtaining $\varepsilon_{\perp}(\omega)$.

Therefore, to obtain a simpler expression for the dielectric constant of nanotubes, the curve fitting was performed using Origin data analysis and graphing software. Polynomial curve fitting (with an order of nine) was performed on the graphs of $\varepsilon_{\perp}(\omega) = \mathrm{Re}\left[\varepsilon_{\perp}(\omega)\right] + j \times \mathrm{Im}\left[\varepsilon_{\perp}(\omega)\right]$, calculated using Equation (4). Wavelength dependent polynomial functions were obtained for both the real and imaginary components of the CNT dielectric constant over the optical and IR regime, as shown in Figure 1. The residual errors generated while performing the curve fitting are shown in Figure 1(b). These errors can be decreased by increasing the order of polynomial fitting. Table 1 shows the calculated polynomial coefficients for the fitted curves. By using these coefficients as *global variables* in COMSOL, the real and imaginary parts of the CNT dielectric constant were effectively

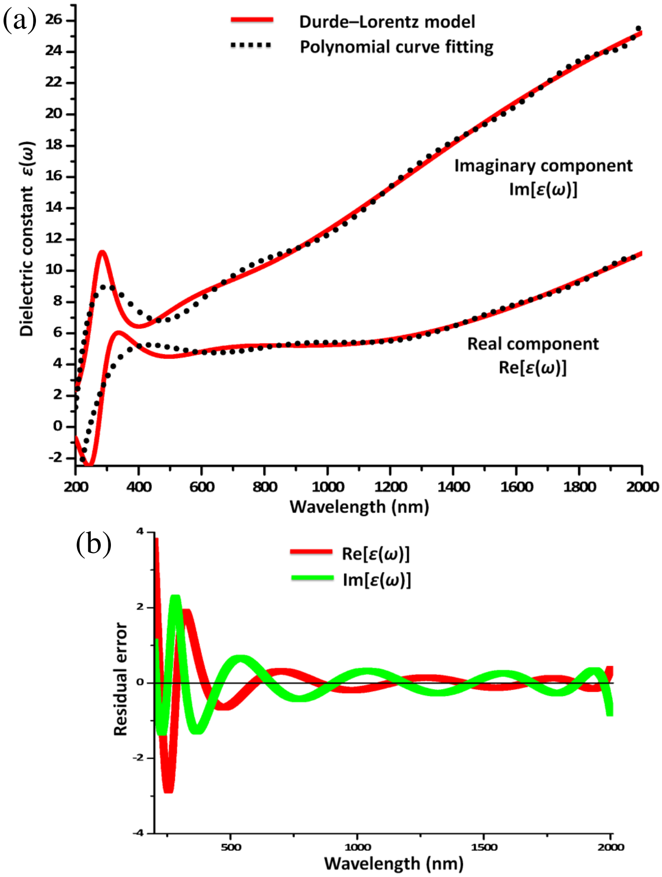

Figure 1. (a) Dielectric function $\varepsilon_{\perp}(\omega)$ of carbon nanotubes for light polarized parallel to the nanotube axis. Solid lines represent the Drude-Lorentz model used in reference [16]. The dashed lines represent the polynomial fitted curves. (b) The residual error generated by the curve fitting.

generated and incorporated as the medium properties of CNTs in the simulations.

This polynomial expression for the nanotube dielectric constant was used for the computational modeling of the MWCNTs based metamaterials. The model was incorporated into the simulations as a function of frequency, allowing the effective calculations of optical transmission spectrum of light propagating through the nanotube arrays based devices.

Table 1. Displays the polynomial curve fitting data for the real and imaginary components of the dielectric function of multiwalled carbon nanotubes.

y=B0+B1*λ^1 +B2*λ^2+B3*λ^3+ B4*λ^4+B5*λ^5+B6*λ^6 +B7*λ^7 +B8*λ^8 +B9*λ^9	**Re**$[\varepsilon_\perp(\lambda)]$	**Im**$[\varepsilon_\perp(\lambda)]$
B0	−59.6353	−243.431
B1	0.47024	3.12728
B2	−0.00114	−0.01586
B3	4.68E-07	4.34E-05
B4	2.70E-09	−7.12E-08
B5	−5.72E-12	7.34E-11
B6	5.35E-15	−4.79E-14
B7	−2.69E-18	1.92E-17
B8	7.12E-22	−4.32E-21
B9	−7.75E-26	4.17E-25

4. MODELING OF MWCNTS BASED METAMATERIALS

It has been reported that periodic arrays of thin metal wire structures act as metamaterials and display a cutoff filtering response in the frequency domains depending on the array geometry [18, 19]. These structures demonstrate artificial plasma frequencies which are much lower than in the metal structures and can be utilized for filtering in microwave and terahertz frequency domains [20, 21]. Such plasmonic metamaterials which operate in the optical regime were established using highly dense two-dimensional periodic arrays of multiwalled carbon nanotubes, as metallic nano-wire structures.

According to reference [18] when the arrays of thin metallic wires are excited by electric fields (parallel to the wire structures), they display artificial low-density plasma like behavior, with a red-shifted plasmon wavelength given by:

$$\lambda_p = a\sqrt{2\pi \ln{(a/r)}} \tag{5}$$

where a is the lattice constant of the 2D metallic wire array and r is the radius, as shown in Figure 2. Electromagnetic wave propagation only occurs through the array below λ_p, due to which the structure acts as a nanophotonic high-pass filter. From Equation (5), an array of metallic wires, having a lattice constant of 400 nm and radius of

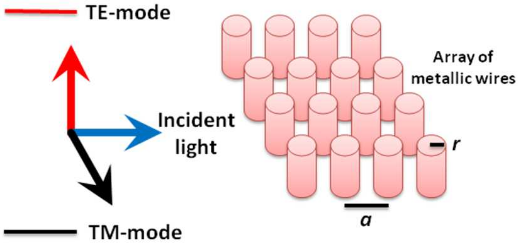

Figure 2. An array of infinitely long thin metal wires of radius r and a lattice constant of a behaves like a low frequency plasma for the electric field oriented along the wires.

50 nm, displays a plasma wavelength of $\lambda_p = 1.44\,\mu\text{m}$. Here, we realize the structure with a square lattice array of MWCNTs.

Computational modeling of the periodic array of MWCNTs was performed to confirm that the high pass filtering effect could be obtained at λ_p, as predicted by the plasmonic theory of metamaterials. We utilized the RF Waves application mode of COMSOL to model the TE (transverse electric (parallel to the nanotubes)) and TM (transverse magnetic (perpendicular to the nanotubes)) propagation of optical/near terahertz waves through the nanotube array. Figure 3(a) shows the geometry of the model established for a 2D square lattice of multiwalled carbon nanotubes, with a lattice constant a of 400 nm and a tube radius of 50 nm.

The model represents the bisection of the infinite vertical rods (MWCNTs) in the x-y plane. The medium surrounding the tubes was set to free space by setting the dielectric constant to 1. The dielectric constant for the MWCNTs was obtained using the curve fitted polynomial expression. The dielectric constant was incorporated into the model as a function of frequency allowing one to calculate accurately the transmission spectrum offered by the nanotube arrays. The propagating waves of various wavelengths were incident on the 2D array from the left of the model. After establishing the shown model geometry a mesh was generated (with maximum element size of $\lambda/5$) over it to define a computational domain and the results were computed by using COMSOL's inbuilt solver.

The modeled wave propagation results are presented in Figures 3(b)–(e) which show that the transmission of light through the nanotube arrays was gradually reduced with the increase in wavelengths. While the 800 nm wave is observed to propagate across the array, the propagation of the 900 nm and 1000 nm waves is highly at-

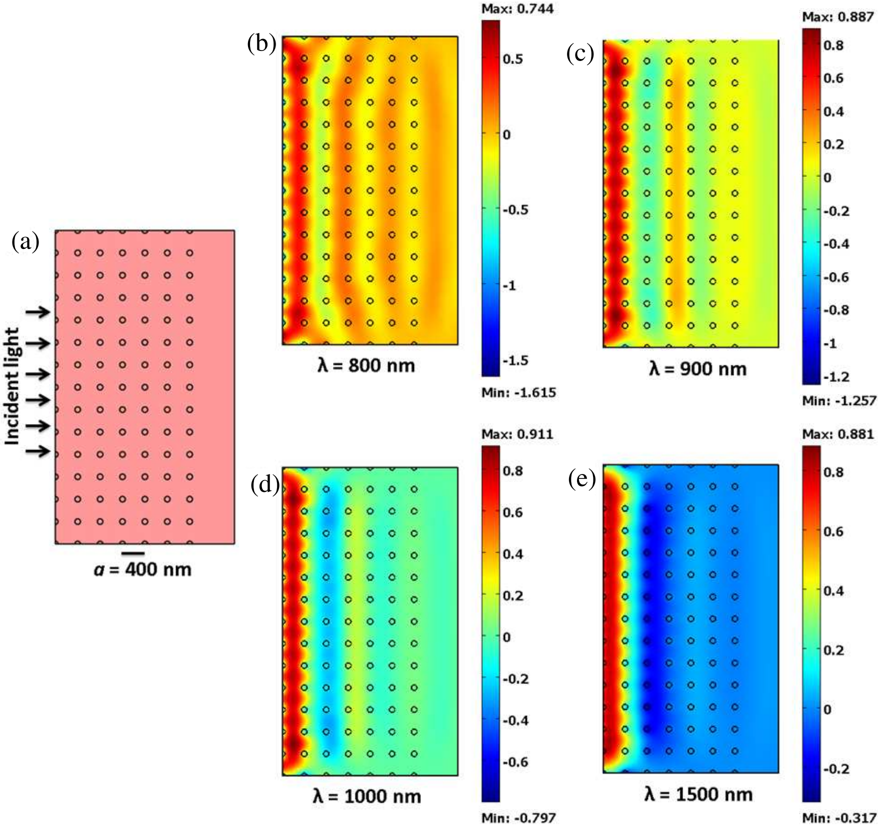

Figure 3. Model geometry of a 2D lattice array of MWCNTs, with a lattice constant 400 nm and tube radius of 50 nm. Modeled results for the TE wave propagation (electric field strength) through the carbon nanotube array, for wavelengths of (b) 800 nm, (c) 900 nm, (d) 1.0 μm and (e) 1.5 μm. A complete transmission is observed for the wavelength of 800 nm and total reflection for the wavelength of 1.5 μm, displaying the high pass filtering effect towards TE polarization of light.

tenuated and very less intensity is transmitted. The nanotube array displays a plasma like behavior with a decreasing skin depth (with the increase in wavelength) through which the incident light penetrates. In Figure 3(e), the 1500 nm wave is observed to be completely reflected from the nanotube array.

This illustrates the filtering response displayed by the nanotube arrays towards incident light. Furthermore, to observe the filtering response more clearly the transmission spectra were also computed for both TE and TM polarizations of light, as shown in Figure 4. The transmission spectrum for the light polarized parallel to the nanotubes (TE) displayed a series of band gaps and transmission bands up till the wavelength of 800 nm. This behavior is similar to that of photonic crystal materials based on periodic arrays of dielectric rods. However, after the wavelength of 830 nm the transmission through the arrays starts to drastically decrease until no more propagation was allowed through the arrays for the wavelength of 1.4 μm and above.

The arrays clearly demonstrate a plasmonic filtering response toward TE polarized light, with a half power cut-off plasmonic wavelength λ_p of the order 830 nm, as shown in Figure 4. Beyond λ_p the transmission through the MWCNT array decreases with increasing magnitude and an infinitely extending band gap (region of no propagation) is observed.

The simulated cut-off filtering response not being as sharp as predicted by the metamaterial theory and the mismatch between the plasmon wavelengths λ_p can be explained by the one dimensional conductivity and lossy nature of multiwalled carbon nanotubes (in the optical regime). It is one of the assumptions of the metamaterial theory (Equation (5)) that the periodic wires structures are perfect metals composed of an electric cloud in which electrons are free to move within the wire structures [18]. However, the multiwalled carbon nanotubes are composed of concentrically wrapped sheets of graphite

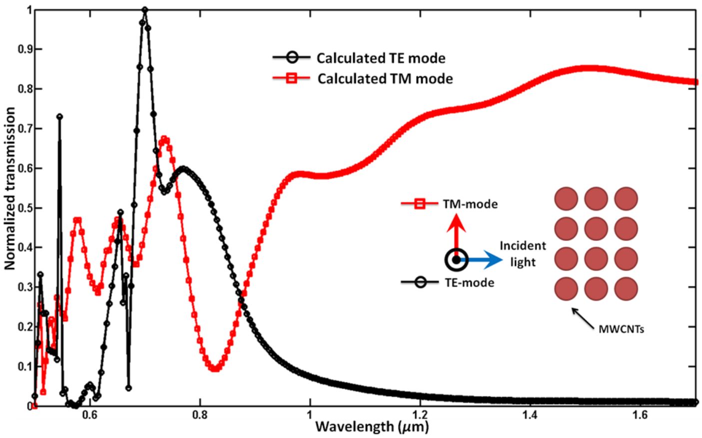

Figure 4. The simulated TE and TM transmission spectra for the square lattice array of MWCNTs. The TE transmission spectrum illustrates the plasmonic filtering response. No such cut-off effect is observed in the TM spectrum.

which allow fast electronic motion only within the graphite sheets. This explains the different effective plasmon frequency displayed by the arrays of MWCNTs.

The transmission spectrum displayed by the array towards light polarized perpendicular (TM) to the nanotubes was also simulated, as shown in Figure 4. The TM spectrum displayed no cut-off filtering effect as predicted by the metamaterial theory. TM light produces transverse electronic motion on the nanotubes but due to their small radiuses compared to the array lattice constant no significant mutual coupling effects take place. Hence most of the TM light is transmitted through the array. There is a strong dip observed is the TM spectrum, near the wavelength of 820 nm corresponding to the diffraction of light satisfying the Bragg condition $\lambda/2 \approx a$, where $a = 400\,\text{nm}$ for the modeled CNT array [22].

5. CONCLUSION

In conclusion, we demonstrate the finite element modeling of periodic arrays of multiwalled carbon nanotubes. The dielectric properties multiwalled carbon nanotubes are similar to graphite and can be calculated by using the Drude-Lorentz model. Most computational softwares lack the capacity to directly incorporate this model into the simulations. We incorporate the dielectric function of nanotubes by using the curve fitting technique and modeled the optical transmission through the periodic arrays. Highly dense periodic arrays of carbon nanotubes act as plasmonic metamaterials displaying artificial filtering effects. We present the computational analysis of these filtering effects. The results presented pave way towards the effective simulations of interesting nanotube arrays based applications like photonic crystals waveguides, switches and diffraction gratings.

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