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Negative refraction and self-collimation in the far infrared with aligned carbon nanotube films

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ABSTRACT

This study demonstrates the far-infrared self-collimation and low-loss transmission of aligned carbon nanotube (CNT) films or arrays. The anisotropic dielectric functions of the CNT array is modeled using the effective medium theory considering the degree of alignment. The spectral regions where hyperbolic dispersion is satisfied are in the far-infrared. In the hyperbolic regime, energy propagates inside the CNT film along the optical axis for nearly all incidence angles. The self-collimation effect is also examined for tilted CNT thin films by tracing the Poynting vector trajectories. Low-loss transmission is explored to understand the impact of alignment on the penetration depth and transmission through the film. In conjunction with the surface radiative properties, the self-collimation and transmission characteristics are distinguished between the two hyperbolic bands of the CNT film. The insight obtained from this work may lead to the utilization of CNT arrays in polarization filtering and infrared imaging.

1. Introduction

A wide range of applications of engineered carbon nanotube (CNT) films have been realized in the last decade. Vertically aligned CNT (VACNT) films have demonstrated high absorption of radiation from the ultraviolet to the far-infrared [1–4]. CNT films have also shown promise as wavelength-selective transparent thin films [5–7]. Studies have also explored transversely oriented CNT arrays as visible light waveguides [8,9]. Using either vertically aligned or obliquely angled CNT arrays for electromagnetic wave manipulation in the infrared or terahertz regime certainly deserves attention as well. The characterization of the unique optical properties of arrayed CNT can contribute toward the development of better sub-diffraction thin film lens, and spectrally and/or polarization-selective thermal sensors [10,11].

Before the turn of the millennium, the scientific community has recognized the potential of nanomaterials for use in confining and manipulating light in unprecedented ways. Pendry and collaborators were early pioneers in designing electromagnetic metamaterials using patterned sub-wavelength standing cylindrical structures [12,13]. Since then, these studies have inspired numerous researchers to realize nanoscale structural analogs, such as silver nanowires [14], doped silicon nanowires [15], and other metallic nanorods [16,17]. The optical anisotropy in such nanowire arrays was identified as a mechanism for negative refraction [18,19]. As the permittivity for electric field along the nanowire axis approaches infinity, a "canalization" or collimation effect can be observed in aligned nanowire arrays [20,21]. This phenomenon is called "self-collimation," and Kosaka et al. [22] were the first to notice the self-collimation effect in photonic crystals. Depending on the geometry of the dispersion surface, these photonic crystals could achieve beam focusing or simple waveguide interconnects [23–25]. Sun et al. [26] observed negative refraction in a natural material, graphite, when the optical

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axis is perpendicular to the plane of incidence by utilizing the hyperbolic isofrequency contour or dispersion relation in the ultraviolet region. As it turns out, multi-walled aligned CNT arrays have similar optical properties as coordinate-transformed graphite [1,27]. The effective dielectric behavior is a uniaxial medium with hyperbolic dispersion in the mid- and far-infrared.

The present study investigates aligned CNT arrays or films with a focus on their hyperbolic nature, negative refraction, radiative properties, and the low-loss self-collimation in a broad infrared wavelength range. Here, the anisotropic dielectric functions of multi-walled CNT array are modeled based on those of graphite, with a coordinate transformation and effective homogenization. The effects of the packing density (filling ratio) and alignment factor on the spectral radiative properties are also examined. The CNT arrays with various tilting angles are studied using a transfer matrix formulation to assess the effectiveness and limitations of self-collimation in the CNT films.

2. Energy streamlines in an anisotropic film

Energy streamlines through a homogeneous thin film are formed by tracing the trajectories of Poynting vectors, which depend on the electric and magnetic fields (i.e., **E** and **H**) [28–30]. In the present study, the transfer matrix formulation is applied to a three-layer system shown in Fig. 1, where the top and bottom media (1 and 3) are semi-infinite free space or vacuum with relative permittivity $\varepsilon = 1$ and permeability $\mu = 1$. Medium 2 is made of aligned CNT film tilted in the plane of incidence (x–z plane) by an angle β with respect to the z axis. The CNT film acts as a uniaxial medium whose optical axis $\hat{\mathbf{c}}$ is parallel to the nanotubes. The CNT film is assumed to be nonmagnetic ($\mu = 1$) whose dielectric tensor in the (x,y,z) coordinates is given by [31,32]

$$\bar{\bar{\varepsilon}} \equiv \begin{pmatrix} \varepsilon_{xx} & 0 & \varepsilon_{xz} \\ 0 & \varepsilon_{yy} & 0 \\ \varepsilon_{zx} & 0 & \varepsilon_{zz} \end{pmatrix} = \begin{pmatrix} \varepsilon_O \cos^2\beta + \varepsilon_E \sin^2\beta & 0 & (\varepsilon_E - \varepsilon_O)\sin\beta\cos\beta \\ 0 & \varepsilon_O & 0 \\ (\varepsilon_E - \varepsilon_O)\sin\beta\cos\beta & 0 & \varepsilon_O \sin^2\beta + \varepsilon_E \cos^2\beta \end{pmatrix} \quad (1)$$

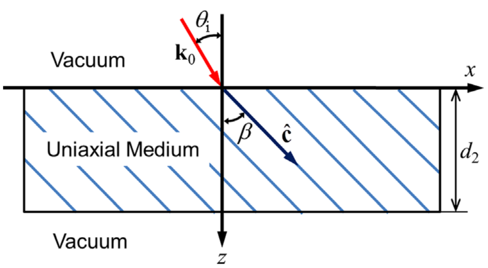

Fig. 1. Illustration of a uniaxial slab of thickness d_2 made of CNT arrays with a tilting angle β, for a plane wave incident from the top vacuum (medium 1). All media are assumed to be nonmagnetic and the uniaxial CNT array is modeled as an effective dielectric tensor whose optical axis $\hat{\mathbf{c}}$ is along the nanotube.

Here, ε_O and ε_E are the dielectric functions of the anisotropic vertically aligned CNT (VACNT) medium for the ordinary wave (electric field $\mathbf{E} \perp \hat{\mathbf{c}}$) and extraordinary wave ($\mathbf{E} \parallel \hat{\mathbf{c}}$), respectively [4]. The dielectric functions are modeled using the Maxwell–Garnett effective medium theory based on the ordinary ($\varepsilon_\perp$) and extraordinary ($\varepsilon_\parallel$) dielectric functions of graphite [1,33]. The effective dielectric functions can be expressed as functions of $\varepsilon_\perp$, $\varepsilon_\parallel$, the filling ratio f, and an alignment factor ζ, viz.

$$\varepsilon_O = \varepsilon_O(\varepsilon_\perp, \varepsilon_\parallel, f, \zeta) \quad (2a)$$

$$\varepsilon_E = \varepsilon_E(\varepsilon_\perp, \varepsilon_\parallel, f, \zeta) \quad (2b)$$

The dielectric functions of graphite for the ordinary wave and extraordinary wave can be found from [34–36]. The filling ratio (f) is defined as the volume occupied by a CNT filament per square unit volume [33]. Slight entanglement or random tilting is considered using the quantity ζ, which is not too far from unity (i.e., perfectly aligned case). In practice, the fabricated CNT arrays typically have ζ values between 0.950 and 0.995 [27]. The effective medium approach using the filling ratio with the misalignment weighting is valid in the infrared, since the electromagnetic wavelength is much greater than the CNT filament diameter and inter-CNT gap spacing [37–39]. Detailed expressions of ε_O and ε_E can be found from [1].

For a transverse electric (TE) wave incidence, since the electric field is parallel to the y axis, the CNT array behaves as an isotropic medium with a dielectric function ε_O, regardless of the tilting. The interest of this work is for transverse magnetic (TM) waves incident on the CNT film from free space. Since the optical axis is rotated in the plane of incidence, there exist no cross-polarizations. The magnetic field for a TM wave (with an angular frequency ω) is perpendicular to the plane of incidence as given by

$$H_{y,j}(x,z) = \left(A_j e^{ik_{z,j}^+ z} + B_j e^{ik_{z,j}^- z}\right) e^{ik_x z - i\omega t}, \quad j = 1, 2, 3 \quad (3)$$

Here, k_z^+ and k_z^- represent the z component of the wavevector for the forward and backward propagating waves, respectively, and A_j and B_j are the amplitude of the forward and backward waves in the jth medium. The x component of the wavevector is the same in all media due to phase matching; that is, $k_x = k_0 \sin\theta_i$, where θ_i is the angle of incidence and $k_0 = \omega/c_0$ with c_0 being the speed of light in vacuum.

The transfer matrix formulation has been used for calculation of radiative properties of thin-film multilayers, such as metal–dielectric multilayers and left-handed photonic crystals [28–30,40]. The transfer matrix for TM waves is given by

$$\mathbf{D}_j = \begin{pmatrix} 1 & 1 \\ Z_j & -Z_j \end{pmatrix}, \quad j = 1, 2, 3 \quad (4)$$

where the surface impedance is $Z_j = k_{z,j}^+/(\omega\varepsilon_0\varepsilon_j)$ for an isotropic medium, where ε_0 is the vacuum permittivity and ε_j is the relative permittivity of medium j. Since