An ultra-broadband electromagnetically indefinite medium formed by aligned carbon nanotubes

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Anisotropic materials with different signs of components of the permittivity tensor are called indefinite materials. Known realizations of indefinite media suffer of high absorption losses. We show that periodic arrays of parallel carbon nanotubes (CNTs) can behave as a low-loss indefinite medium in the infrared range. We show that a finite-thickness slab of CNTs supports the propagation of backward waves with small attenuation in an ultra-broad frequency band. In prospective, CNT arrays can be used for subwavelength focusing and detection, enhancing the radiation efficiency of small sources.

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Metal-dielectric nanostructures [1, 2] have already brought applications to microscopy and sensing [3] and have the potential for realization of future nanosized optical devices and circuits [4]. In 2003 the authors of [5] noticed the potential of so-called indefinite metamaterials for subwavelength imaging of objects at electrically large distances from them (the concept of such imaging was first introduced in [6]). Indefinite metamaterials are artificial uniaxial materials in which the axial and tangential components of the permittivity and permeability tensors have different signs. In these materials the isofrequency surfaces have hyperbolic shape. This results in a possibility to design a “hyperlens”, where evanescent near fields are transformed into propagating modes and can be transported at electrically long distances [7].

The main challenge on the way to realize this and other effects is in the realization of low-loss indefinite materials. For waves of only one linear polarization (TM-polarized waves with respect to the axis of positive permittivity) it is enough to realize a layer of an indefinite dielectric metamaterial [8], whose permeability is unity and only components of the permittivity tensor have different signs. For the visible range such a metamaterial was designed in [8] as an array of parallel plasmonic (metal) nanowires. Since all plasmonic phenomena are related to strong dissipation, the axial component of the permittivity has a significant imaginary part which strongly restricts the subwavelength imaging property of the hyperlens [8]. In the microwave range, materials with negative permittivity can be realized as arrays of thin metal wires [9, 10]. However, if the field varies along the wires, the properties of the structure are more complicated that those of a continuous medium due to strong spatial dispersion [11]. The spatial dispersion can be suppressed partially [12] or even totally [13–15]. However, manufacturing of structures [12–15] becomes a challenge already at millimeter waves as dimensions need to be quite small for high-frequency applications. For electromagnetic waves in the THz and mid infrared (MIR) range there are no known structures with an indefinite permittivity tensor. The imaginary and real parts of complex permittivity of metals in this range are of the same order and rather high.

Here we show that arrays of metallic carbon nanotubes (CNT) behave as ultra wide-band and low-loss indefinite materials in the THz and MIR ranges. This collective property of arrays results from the quantum properties of individual CNT, namely their high quantum inductance, so-called kinetic inductance [16], which leads to suppression of spatial dispersion in arrays of parallel CNTs. This can open a possibility for realization of interesting devices, which we discuss here.

Two-dimensional periodic arrays of carbon nanotubes are fabricated by many research groups. Such CNT arrays form finite-thickness slabs which are used already as field emitters [17], biosensors [18] and antennas [19, 20]. We assume that all nanotubes possess metallic properties that is a realistic assumption in view of recent studies of single-wall CNTs [21–23]. Usually carbon nanotubes form hexagonal lattices in arrays in processes of fabrication. However, for low-density arrays for which arrangement of nanotubes is not important. By this reason and for simplicity we assume lattices to be square.

For eigenwaves propagating in arrays of infinitely long carbon nanotubes we take the space-time dependence of fields as

$$\Psi = e^{j(\omega t - k_z z)}.$$

Effective electromagnetic properties of CNT arrays can be understood from the nonlocal quasistatic model of wire media (WM) [15]. In the framework of the effective medium model the CNT array can be considered as a uniaxial material with the permittivity dyadic

$$\varepsilon = \varepsilon_{xx} \mathbf{z}_0 \mathbf{z}_0 + \varepsilon_0 (\mathbf{x}_0 \mathbf{x}_0 + \mathbf{y}_0 \mathbf{y}_0),$$

where $$\varepsilon_0$$ is the permittivity of vacuum (we consider CNTs placed in vacuum). As was shown in [15],

$$\epsilon_{xx} = 1 - \frac{k_p^2}{k^2},$$

where $$k_p$$ is the effective plasma wavenumber $n^2 = \frac{\omega^2}{C_{cnt}^2 \varepsilon_0}$, $C_{cnt}$ is the effective inductance and capacitance per unit length, respectively, and the parameter

$$\xi = \frac{R_{cnt}}{L_{cnt}} \sqrt{\frac{\sigma_{xx}}{\varepsilon_0}}$$

is responsible for losses. Distributed parameters $L_{cnt}$, $C_{cnt}$ and $R_{cnt}$ for separate carbon nanotube can be obtained using the model of impedance cylinder and effective boundary conditions developed in [24]. According to this model the simple approximate expression for the complex surface conductivity, which is valid for metallic zigzag CNTs at frequencies below optical transitions looks as:

$$\sigma_{zz} \approx -j \frac{2 \sqrt{3} \varepsilon_0 \Gamma_0}{3q \pi \hbar^2 (\omega - j \nu)}.$$

Here $e$ is the electron charge, $\Gamma_0 = 2.7$ eV is the overlapping integral, $\tau = 1/\nu$ is the relaxation time, $\hbar$ is the reduced
Planck constant. The radius of metallic zigzag CNT, taken as an example, r is determined by an integer q as \( r = 3\sqrt{3q}b/2\pi \), where \( b = 0.142\) nm is the interatomic distance in graphene. Since the wall of a single-wall CNT is a monoatomic sheet of carbon, (3) can be considered as the surface conductivity of the carbon nanotube. The surface impedance per unit length

\[
z_i = \frac{1}{2\pi r \sigma_{zz}} = \frac{\sqrt{3q}h^2}{4\varepsilon^2 \Gamma_0 r} + j\omega \frac{\sqrt{3q}h^2}{4\varepsilon^2 \Gamma_0 r} = R_0 + j\omega L_0. \tag{4}
\]

Let us find \( L_{\text{cnt}} \), \( C_{\text{cnt}} \) and \( R_{\text{cnt}} \) for a separate carbon nanotube. The total inductance per unit length \( L_{\text{cnt}} = L_0 + L_{\text{em}} \), where \( L_0 \) is the kinetic inductance [16] defined by the formula (4), which has a quantum nature, and \( L_{\text{em}} \) is the electromagnetic inductance per unit length. For thin CNTs the kinetic inductance strongly dominates over the electromagnetic inductance, e.g., for the zigzag CNT, having the radius \( r \approx 1.53\) nm (\( q = 13 \)), \( L_{\text{em}} = 5.6 \times 10^{-7}\) H/m and \( L_0 = 3.7 \times 10^{-5}\) H/m. 

This shows that the electromagnetic inductance can be neglected. The total capacitance of CNTs \( C_{\text{cnt}} \) includes the electrostatic capacitance defined as \( C_s = \epsilon_0 \mu_0 / L_{\text{em}} \) and the quantum capacitance \( C_q = 2e^2/(\hbar \nu_F) \), where \( \nu_F \) is the Fermi velocity which is equal to \( 8 \times 10^5 \) F/m for graphene and CNT [25]. The two effective capacitances are connected serially. For the considered example \( C_s \approx 1.98 \times 10^{-11}\) F/m, \( C_q \approx 9.66 \times 10^{-11}\) F/m and \( C_{\text{tot}} = C_s/C_q / (C_s + C_q) = 1.64 \times 10^{-11}\) F/m. Let us estimate the terms entering formula (2) at the frequency \( \omega/2\pi = 1\) THz. One obtains \( k_p^2 = 4.32 \times 10^{10}\) m/\( \nu^2 \), \( k_p^2 = 4.39 \times 10^8\) m/\( \nu^2 \) and dimensionless \( n^2 = 4.32 \times 10^7\), so the last term in the denominator of formula (2) can be neglected.

Thus strong spatial dispersion is suppressed and the material behaves as a uniaxial free-electron plasma, where electrons can move only along \( z \)-direction. Apparently, this corresponds to an indefinite medium at frequencies below the plasma frequency, because the permittivity in the directions orthogonal to the tubes is close to unity. Dispersion equation for waves propagating in a uniaxial crystal is [26]

\[ \epsilon_0 k_{\perp}^2 = \epsilon_{zz}(k^2 - k_p^2), \tag{5} \]

where \( k_{\perp}^2 = k_x^2 + k_y^2 \). Substituting (2) with \( k_p^2/n^2 = 0 \) into Eq. (5) we obtain a simple formula:

\[ k_x^2 = \frac{k^2(k^2 - k_s^2 - k_p^2)}{k^2 - k_p^2} \] \[ k_y^2 = \frac{k^2(k^2 - k_s^2 - k_p^2)}{k^2 - k_s^2} \tag{6} \]

which describes a typical conic-type dispersion, see Fig. 1. Applicability of the described effective medium model was verified comparing with the results of the electrodynamic model, based on Green’s function method [27]. This model takes into account the array periodicity and demonstrates that the effective medium model gives a good agreement with the more accurate theory for small transversal wave numbers \(|k_x, k| < 0.15\pi\).

In a series of papers published in 2003-2004 by the groups of G. Eleftheriades, C. Caloz and T. Itoh, Pendry’s concept of perfect lens [6] was experimentally confirmed for a planar analogue of the perfect lens (see also books [28] and [29]). Planar networks composed of backward-wave transmission lines (TLN) are surface analogues of bulk double-negative materials. Surface formed by TLN acts as a layer of an indefinite medium, where the spatial dispersion is suppressed [30]. Based on the results of the previous discussion, we expect that in the THz and MIR ranges a similar backward-wave structure can be realized as an array of aligned CNTs.

Let us consider waves propagating in a finite-thickness slab of CNTs assuming for simplicity that the slab is placed between perfect electric conductor (PEC) and perfect magnetic conductor (PMC) planes, where the PMC boundary models the open-ended interface with free space. The resonant condition for the longitudinal wavenumber \( k_z \) reads \( k_z = \pi/(2h) \) where \( h \) is the thickness of the slab. The relation between the transversal wave vector components and the wavenumber in free space is the following:

\[ k_{\perp}^2 = k_x^2 + k_y^2 = \frac{(k^2 - k_p^2)(k^2 - k_s^2)}{k^2}. \tag{7} \]

One can easily show that the derivative \( \frac{dk_{\perp}^2}{dk^2} < 0 \) if \( k_z/k > 1 \) and \( k_p/k > 1 \). Thus, in this regime the finite-thickness slab supports propagation of backward waves, because the group velocity is in the opposite direction to the phase velocity.

This property can be understood also from considering a
planar waveguide filled with a CNT array (the array axis is orthogonal to the walls of the waveguide). The propagation constant along the waveguide is equal to

$$k_\perp = \sqrt{\varepsilon_{zz} [k^2 - (m\pi/2h)^2]}$$

(8)

where $m$ is a positive integer and $h$ is the height of the waveguide [11]. If $\varepsilon_{zz} < 0$, backward-wave propagation is allowed when $k < m\pi/2h$ and forbidden for $k > m\pi/2h$.

Fig. 2 illustrates dispersion properties of three modes, corresponding to $m = 1, 2$ and $m = 3$. There are three embedded conical surfaces. The internal cone corresponds to $m = 1$, and the external one to $m = 3$. One can see that backward waves propagate in the slab in a very wide frequency range. Their properties are quite isotropic in the $xy$-plane due to a very small period of the CNT lattice. It follows from formula (8) that there is no low-frequency cutoff for any mode, which can propagate at low frequencies with very large transversal wavenumbers $k_\perp$. The effective medium model becomes unacceptable if $k_\perp d > 0.15\pi$ (compare with results of [27]).

To suppress the spatial dispersion we do not need any loadings because CNTs possess very high kinetic inductance which ensures that $k^2/\gamma^2 \ll k^2$. Comparing to the microwave backward-wave structures based on loaded transmission lines and mushroom layers we note that in CNT arrays there is no parasitic inductance between the cells and parasitic capacitance between the tube ends and the ground plane is very small due to the small cross-section area of the tubes. This leads to a dramatically increased bandwidth of backward-wave propagation and gives the ground to consider CNT arrays as perfect backward-wave metamaterials.

For any optical material at terahertz and infrared ranges one of the most important issues is the level of losses. In CNTs it is determined by the relaxation time $\tau$ at the frequencies below optical transitions. At low frequencies, including values close to a few terahertz, $\tau$ at room temperature usually is estimated as $3 \times 10^{-12}$ s [24, 31]. Such a relaxation time gives the ratio $\text{Im}(k_{\perp})/\text{Re}(k_{\perp}) \simeq 10^{-3}$ at frequencies around 50 THz. However, in other sources this value is taken to be $10^{-13}$ s [32], which corresponds to considerably higher losses, namely, $\text{Im}(k_{\perp})/\text{Re}(k_{\perp}) \simeq 0.025$ in the same frequency range.

Here we point out one potential application of indefinite dielectric materials based on the conversion of incident evanescent waves into propagating transmitted waves by samples of an indefinite medium. Such a conversion is possible up to very high, in the limiting case infinite spatial frequencies. The idea is illustrated by Fig. 3.

Consider a prism with the wedge angle $2\alpha \approx \pi/2$ filled with an indefinite medium with components of the permittivity tensor $\varepsilon_{zz} < 0$, $\varepsilon_{yy} = \varepsilon_{xx} > 0$. For a wedge made of a CNT array this would correspond to the $z$-oriented tubes. Let an evanescent wave attenuating along $z$ and having transversal wave vector component $k_y > k$ contribute $\omega\varepsilon_{0}\mu_{0} Q_{\perp}$ impinges on the left side of the prism. In the isofrequency diagram $(k_z - k_y)$ which is depicted in Fig. 3 (a) the wave vector of the refracted wave selects the point at the medium isofrequency contour so that the component of the refracted wave vector which is parallel to the first asymptote of the hyperbolic isofrequency contour preserves. On the second side of the prism the compo-
backward waves, which are characterized by low levels of losses in the terahertz and mid-infrared ranges. The finite-thickness carbon nanotube array can be considered as the perfect isotropic backward-wave metamaterial. We have theoretically demonstrated that these materials can be used as transformers of full spectrum of evanescent electromagnetic waves into propagating ones, and as devices for emission enhancement for subwavelength-sized radiators.

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