Plasmonic Band Gaps and Waveguide Effects in Carbon Nanotube Arrays Based Metamaterials

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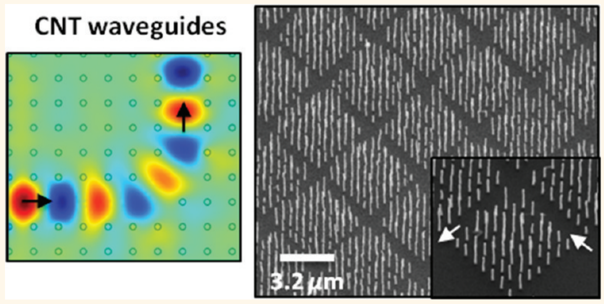
ABSTRACT



Highly dense periodic arrays of multiwalled carbon nanotubes behave like low-density plasma of very heavy charged particles, acting as metamaterials. These arrays with nanoscale lattice constants can be designed to display extended plasmonic band gaps within the optical regime, encompassing the crucial optical windows (850 and 1550 nm) simultaneously. We demonstrate an interesting metamaterial waveguide effect displayed by these nanotube arrays containing line defects. The nanotube arrays with lattice constants of 400 nm and radius of 50 nm were studied. Reflection experiments conducted on the nanoscale structures were in agreement with numerical calculations.

KEYWORDS: metamaterials · carbon nanotube arrays · plasmonic band gaps · optical waveguides

Advances in nanoscale fabrication allow for the realization of artificial materials with properties that do not exist in nature, metamaterials.[1] They are composed of subwavelength magnetic structures, placed at close proximity to each other.[2] Due to mutual coupling between individual structures, they present properties to incident electromagnetic radiation that are different from those associated with the material from which the structures are comprised of. We report the use of multiwalled carbon nanotubes (MWCNTs) as subwavelength structures to produce optical metamaterials that exhibit artificial dielectric properties and band gaps within the optical regime, paving the way toward interesting waveguide effects.

Multiwalled carbon nanotubes first reported in ref 3 are very interesting materials and have been the focus of enormous research in the past decade. They are mostly metallic and are able to carry high current densities along their axis.[4] Apart from their myriad applications such as in field emission displays,[5] as electrodes in rectifiers,[6] solar cells,[7] and optical antenna arrays,[8] periodic arrays of vertically aligned carbon nanotubes have also been used as photonic crystals.[9] Photonic crystals present periodicity in their dielectric constant, which causes Bragg scattering of the incident electromagnetic waves with comparable wavelengths. This introduces band gaps, regions where no wave propagation is allowed. If these band gaps fall in the optical frequency range, they are called photonic band gaps. Therefore, for the carbon nanotube arrays to display photonic band gaps (in the optical regime), their lattice constants should be on the order of a few hundred nanometers. To the best of our knowledge, the carbon nanotubes based photonic crystals reported so far have lattice constants on the order of 1 μm, and they display photonic bands well outside of the optical regime in the terahertz range. Due to the technical difficulties in measuring the band gaps in these regimes, we see that so far only the optical diffraction and scattering measurements from periodic arrays of CNT have been reported.[9−11] We, however, report highly dense periodic arrays of multiwalled carbon nanotubes having a tube radius of 50 nm and lattice constants of 400 nm. When grown at such a close proximity, the metallic properties of the nanotube arrays can be used to achieve metamaterials displaying an artificial negative dielectric constant toward incident light, causing reflection. We, for the first time, report the computational

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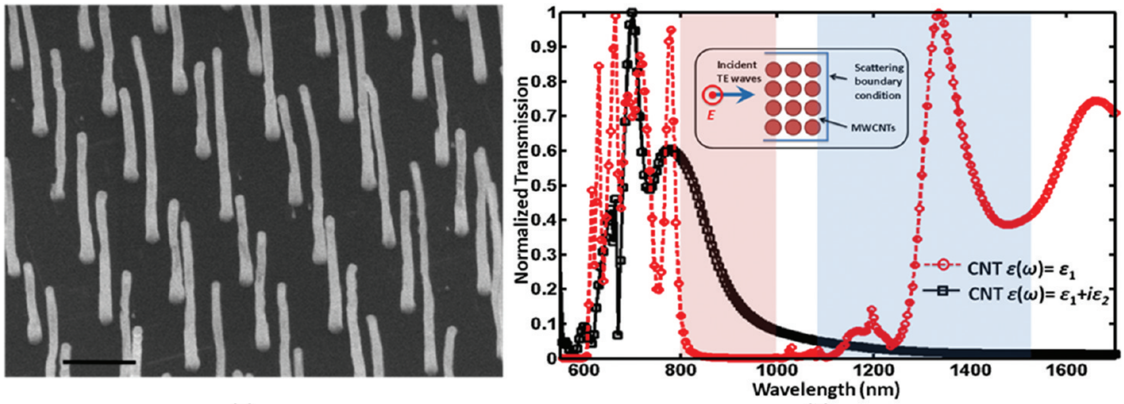

(a) (b)

Figure 1. (a) SEM image of CNT arrays with $a = 400$ nm and $r = 50$ nm. The marker represents 400 nm. (b) Calculated TE_{01} mode transmission spectra through the CNT arrays. The simulation considering the CNTs as dielectric rods (red curve) shows a photonic crystal like response, while the calculations considering the metallic character of CNTs (black curve) shows the metamaterial effect and an infinitely extending plasmonic band gap. The inset shows the simulated model geometry.

analysis of these metamaterials as optical waveguides and filters operating in the 850 and 1550 nm windows used for optical communications. Effective fabrication of these metamaterials was performed. A wavelength selective waveguiding effect was observed and is in agreement with simulation results.

RESULTS AND DISCUSSION

Metamaterial Theory. Pendry was the first to propose such plasmonic metamaterials consisting of metallic wires with very small diameters compared to both the lattice constant and the wavelength.[2,12] The electromagnetic response of an array of thin metallic wires, excited by an electric field parallel to the wires (transverse electric (TE) mode) is similar to that of a low-density plasma of very heavy charged particles, with a plasma frequency ω_p:

$$\omega_p{}^2 = \frac{2\pi c_0{}^2}{a^2 \ln(a/r)} \qquad (1)$$

where c_0 is the velocity of light in vacuum, a is the lattice constant of the 2D wire array, and r is the radius of the wires. This concept can be used for lowering the plasma frequency in nanotube-based applications and achieving negative dielectric constants for producing metamaterials. The lowering of the plasma frequency is due to the increase in the effective electronic mass on the nanotubes due to the induced current and corresponding coupling of the magnetic fields around them.[2]

High-density periodic arrays of carbon nanotubes display interesting band gap regions called plasmonic photonic band gaps that extend from zero-frequency toward the plasmon frequency, ω_p.[13] A similar band gap is also observed for metals due to plasma resonances at optical frequencies. The electromagnetic waves of frequencies lower than the plasmon frequency, ω_p, are reflected, while the frequencies greater than plasmon frequency, ω_p, are allowed to propagate through. This allows the use of these metamaterials as nanoscaled high pass filters, as reported in our previous work.[14]

According to eq 1, the effective plasma frequency strongly depends on the nanotube radius and lattice constant. To obtain these plasmonic band gaps and the plasmon frequency within the optical regime, carbon nanotube arrays with nanoscale dimensions are required. Vertically aligned multiwalled carbon nanotubes were grown at precisely determined locations by the process of plasma-enhanced chemical vapor deposition (PECVD).[15,16] A uniform array of multiwalled carbon nanotubes, 50 nm in diameter, 2 to 3 μm in height, with 400 nm spacing is shown in the scanning electron microscopy (SEM) image of Figure 1a.

The propagation of light and photonic band gaps displayed by the carbon nanotube arrays were analyzed through simulation. The transmission spectrum of the array for light polarized parallel to the nanotubes (transverse electric (TE_{01}) mode) was studied using finite element method (FEM) simulation. A 2D lattice of nanotubes was modeled as thin rods of infinite length in air. The lattice consisted of 8 rows of 15 CNTs with a radius of 50 nm and lattice constant of 400 nm. The dielectric constant of individual MWCNTs is defined by their frequency-dependent dielectric function $\varepsilon(\omega)$, which is anisotropic in nature[11,17] and matches very closely with that of bulk graphite.[18,19] Like other metals MWCNTs have a complex dielectric function, $\varepsilon(\omega) = \varepsilon_1 + i\varepsilon_2$, with an imaginary loss factor related to their conductivity. The frequency-dependent dielectric constant for graphite was incorporated in the simulations.

To study this metamaterial effect, the transmission spectrum through the lattice was simulated with ($\varepsilon(\omega) = \varepsilon_1 + i\varepsilon_2$) and without ($\varepsilon(\omega) = \varepsilon_1$) considering the imaginary part of the dielectric constant, as shown in Figure 1. The calculations with the CNT dielectric constant, $\varepsilon(\omega) = \varepsilon_1$, modeled the dielectric characteristics of only the CNTs. The calculated transmission

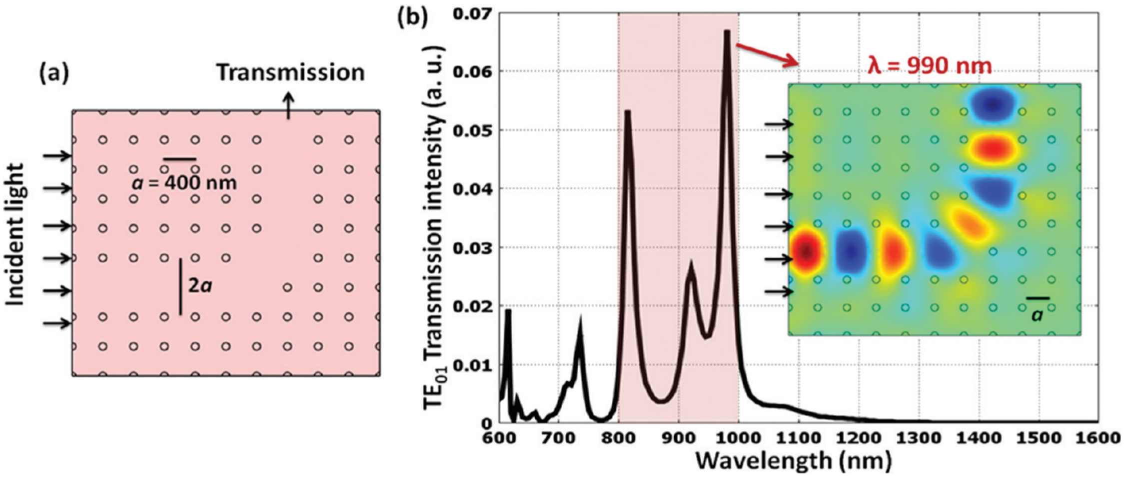

Figure 2. (a) Simulated MWCNT-based photonic waveguide, with a line defect of 2*a* = 800 nm and tube radius = 50 nm. (b) Calculated (TE_{01} mode) light propagation through the waveguide. (c) Propagation of a 990 nm wavelength wave through the 800 nm wide waveguide.

spectrum displayed a dielectric rod based photonic crystal like behavior presenting a succession of transmission bands (from 600 to 800 nm and above 1300 nm) and band gaps (<600 nm and from 800 to 1300 nm). In the first transmission window, which extends from 600 to 800 nm, the dielectric rod based photonic crystal behaves as an ensemble of Fabry–Perot cavities coupled to one another along the propagation direction.[13] The band gap extending from near 800 nm to approximately 1300 nm is associated with the Bragg condition $\lambda/2 \approx a$, where λ represents the wavelength in the photonic crystal and *a* represents the lattice constant (400 nm). The frequency modes with wavelengths larger than 1300 nm are seen to propagate through the lattice.

However, the simulation with the CNT dielectric constant, $\varepsilon(\omega) = \varepsilon_1 + i\varepsilon_2$, takes the metallic characteristics into consideration, displaying metamaterial filtering effects. A similar transmission band extending from 600 nm to about 850 nm is observed, along with a plasmonic photonic band gap extending until infinite wavelengths (zero frequency). The overall plasmonic effect predicted by metamaterial theory can be observed from the simulation results. The carbon nanotube arrays with nanoscale spacing display an effective metallic character, reflecting the frequency modes belonging to the infinitely extending plasmonic band gap. This is of interest here, as it shows that the plasmonic band gaps encompassing several important optical communication windows such as 850 and 1550 nm can be obtained from the engineered carbon nanotube array based metamaterials. The observed effect paves the way toward utilization of these structures for accomplishing further metamaterial applications such as waveguiding and wavelength-dependent filtering (diplexer) in these optical windows.

Linear defects in a photonic band gap material can support the propagation of linearly localized frequency modes that fall inside the band gaps.[20] Such defects can act as a waveguide for electromagnetic waves, without relying on total internal reflection. We demonstrate the waveguiding effects that can be obtained from the carbon nanotube based plasmonic metamaterials. By introducing line defects within the nanotube arrays, waveguide structures with 90° bends were modeled and transmission through them was numerically calculated, as show in Figure 2. Line defects with widths 2*a* (800 nm) and 3*a* (1200 nm) were introduced by removing one or two rows of carbon nanotubes from the lattice, respectively. Figure 2 shows the computational analysis of the optical transmission (TE_{01} mode) through a 800 nm wide line defect. The transmission spectrum for the waveguide (Figure 2b) shows that that only the frequency modes falling within the plasmonic band gap are guided through the line defect. A 2D localized transmission of light (ranging from 800 to 1000 nm) through the line defect is observed. No wave propagation takes place out of the waveguide, as there are no extended modes into which the propagating mode can couple. Within the plasmonic band gap the CNT array around the waveguide displays an artificial negative dielectric constant to the propagating light, due to which a localized propagation takes place only through the defect. An average propagation loss of 72 dB/cm was calculated for the waveguide structures. The light having wavelengths (800–1000 nm) comparable to the waveguide dimensions (800 nm) was effectively transmitted through it. The 820 and 990 nm waves were transmitted through the waveguide with the highest transmission intensity.

The simulated results of optical transmission through the 1200 nm (3*a*) wide waveguides are shown in Figure 3. Transmission of the frequency modes with wavelengths on the order 1100–1550 nm is observed. However, the modes with wavelengths on the order of

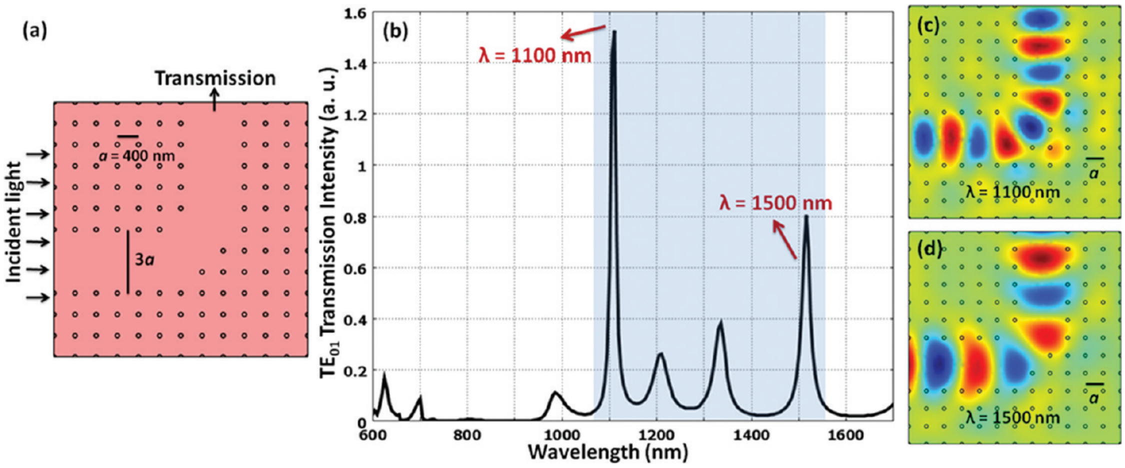

Figure 3. (a) Simulated MWCNT-based photonic waveguide, with a line defect of $3a$ = 1200 nm and tube radius = 50 nm. (b) Calculated (TE_{01} mode) light propagation through the waveguide. The mode with wavelength comparable to the waveguide dimensions propagates efficiently. The propagation of (c) 1100 nm and (d) 1500 nm wavelength waves through the waveguide.

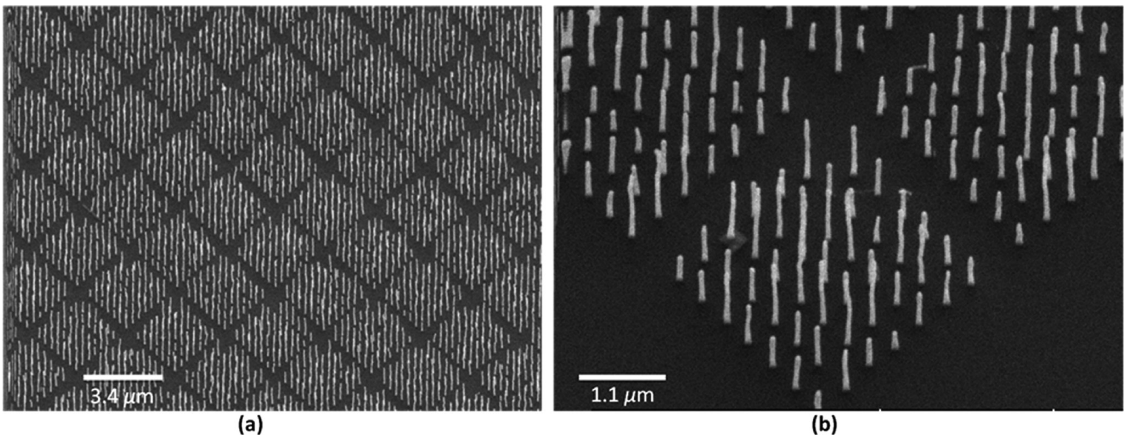

Figure 4. (a) Electron microscopy image of a 2D square lattice array of MWCNTs having a radius of 50 nm and lattice constant of 400 nm, consisting of $2a$ = 800 nm wide line defects with 90° bends. (b) The same in higher resolution.

800 to 1000 nm show significantly reduced propagation through the 1200 nm (3*a*) wide waveguide. Therefore, the simulated spectra from the two waveguides show efficient transmission of the frequency modes that have wavelengths comparable to the waveguide widths.

This phenomenon displayed by plasmonic band gap metamaterials can also be used to demonstrate wavelength-dependent beam splitters and band-pass filters at micrometer scale.[21] The unique plasmonic band gap displayed by the high-density array of metallic nanotubes extends from about 850 nm to infinite wavelength. Unlike the conventional dielectric photonic crystal band gaps, it simultaneous covers multiple optical windows (850 and 1550 nm) used for optical communication. Efficient filtering of these two windows can be achieved by introducing multiple line defects with 2*a* or 3*a* dimensions in the MWNT-based photonic crystals.

Fabrication. To achieve these metamaterial waveguide effects in the optical domain, metallic carbon nanotube arrays of nanoscale dimensions and interspacing were fabricated. MWCNTs being metallic are promising materials to establish such metamaterial structures, and the development of deterministic growth methods for vertically aligned MWCNTs[14,15] enables high a/r aspect ratio nanotube arrays to be realized. The process of PECVD was used to fabricate uniform square lattice arrays of carbon nanotubes with a spacing of 400 nm and average radius on the order of 50 nm. By omitting complete rows of CNTs, 800 nm (2*a*) wide line defects were introduced with 90° bends. The numerical studies suggested that interesting waveguiding effects can also be achieved through the line defects that are 2*a* or 3*a* wide. However, considering the spectral range of our optical characterization equipment we fabricated 800 nm (2*a*) wide waveguides. The fabricated nanotube array consisting of waveguides is shown in the scanning electron microscopy image of Figure 4a and with higher magnification in Figure 4b.

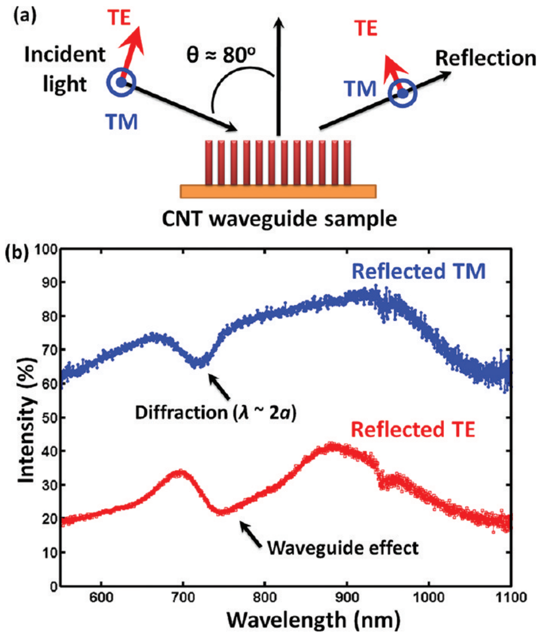

Figure 5. (a) Schematic representation of the experimental setup. (b) Reflection measurement from the sample for light polarized parallel (TE) and perpendicular (TM) to the multi-walled carbon nanotubes. A drop in the reflected TE mode is observed extending from near 700 to 890 nm, showing the waveguiding effect, which significantly matches the theoretical result, whereas only the diffraction dip at 740 nm is observed for the TM mode.

Due to the small lattice constant, the nanotubes were tangled in some regions of the substrate. Additionally, some array defects were produced due to inhomogeneous depth of the nickel catalyst layer, affecting the decomposition of the carbon source and, hence, producing shorter MWCNTs. However, a steady periodicity required for a 2D square lattice was common throughout.

Characterization. The CNT-based waveguides were characterized using a spectrometer setup to obtain the reflection spectra. An Ocean Optics white-light source with an optical spectrum from 450 nm up to 1100 nm was utilized for illumination. The polarization of the white-light beam was selected before the beam was guided onto the sample. The setup had no limit to the positive incident angles it could record, and near-normal incident angles (~80°) were used, as shown in Figure 5a. Microscope objectives were used to collimate the light beam coming out of the fiber and to focus it onto the sample. The sample size of 1.25 mm square was sufficient for these experiments, as it was much larger than the incident beam's spot size. All the reflected light was focused back into the fiber by placing a series of lenses and objectives very close to the sample. An Ocean Optics 2000 spectrometer was utilized to capture the reflected signal for the spectral range of almost 550 to 1050 nm, with a resolution of 0.2 nm.

The measured reflection spectrum at near-normal incidence for light polarized parallel (TE) and perpendicular (TM (transverse magnetic)) to the nanotubes is shown in Figure 5b. The reflected spectrum for TM light displayed a higher reflection intensity (about 40%) compared to TE due to the negligible interaction with the carbon nanotubes. The TM spectrum also displays a dip near 720 nm. This dip is due to the diffraction of light caused by the periodicity of the nanotube arrays. According to the Bragg condition ($\lambda \approx 2a$), the square lattice CNT array with a = 400 nm must cause the diffraction of light with a wavelength of about 800 nm. However, due to the shorter angle of incidence (<90°) used in the experiments, the effective a presented to the incident light is different, causing a blue shift in the diffraction dip at 720 nm.

Such a blue shift was also observed in the reflection spectrum of TE light. A significant drop in TE transmission (relative to the TM transmission) was observed in the region from 700 to 890 nm, with the lowest intensity at 740 nm, showing the light in this region is being coupled and guided through the carbon nanotube based waveguides. The spectrum with light polarized perpendicular (TM) to the nanotubes did not show such an extended dip in this regime, showing that most of the light was reflected. However, a drop in reflection for parallel polarized light confirms the metamaterial effect. According to the simulated waveguiding effects in Figure 2, light in the range of 800 to 1000 nm is guided through the 800 nm wide defect in the CNT array. The TE mode reflection spectrum shows an extended dip for the guided (and scattered) modes extending from 700 to 890 nm. A blue shift on the order 100 nm is expected due to the different incident angle used and is consistent with the diffraction dip in the TM mode spectrum. The experimentally measured spectrum shows a good agreement with the corresponding numerical simulations of waveguiding effects shown in Figure 2.

The plasmonic band gaps displayed by the metamaterial can be further brought into the optical regime (to comprehensively encompass multiple optical windows) by increasing the material density of the sample, *i.e.*, by increasing the radius of the tubes and decreasing the lattice constant as presented in eq 1. Growth of well-aligned MWCNTs at an interspacing of less than 400 nm can be achieved by optimizing the catalyst layer and growth time in the PECVD process.[22] To improve the device prospect and feasibility of carbon nanotube based metamaterials, these arrays need to be fabricated with superior periodicity and device geometry to reduce the scattering losses and increase the optical confinement. A better characterization method with an in-plane optical transmission will be employed in the future for an in-depth analysis of the proposed waveguide structures.

CONCLUSION

In conclusion, we, for the first time, demonstrated a numerical and experimental study on the metamaterial waveguiding effects displayed by high-density periodic arrays of carbon nanotubes with line defects. The arrays effectively act as a pseudoplasma, displaying an artificial negative dielectric constant in the plasmonic band gap extending from the plasma frequency to infinite wavelength. These very wide plasmonic band gaps encompass several crucial optical windows simultaneously, which can be efficiently coupled and guided through the line defects (of comparable dimensions) in carbon nanotube arrays. The measured coupling response of 800 nm wide waveguides was in good agreement with numerical calculations. The plasmonic band gaps of the structure can be engineered by varying the geometrical parameters. Plasmonic characteristics of nanotube arrays also have great potential in nanoscale optical wave guiding and wavelength-selective filtering.

METHODS

Carbon Nanotube Growth Process. Square lattice arrays with a spacing of 400 nm and tube radius of 50 nm were grown on silicon substrates. Electron-beam lithography was used to pattern a 5 nm thick nickel catalyst layer into an array, with each dot being 100 nm in diameter. This allowed the growth of a single MWCNT of 50 nm radius on each dot. The substrate was heated by dc current under vacuum of 10−2 mbar to 650 °C at a ramping rate of 100 °C per minute. This mild heating process is preferred to prevent the catalyst dots from cracking. Ammonia gas was then introduced to etch the surface of the nickel catalyst islands. Acetylene was chosen to be the carbon source and was imported into the deposition chamber after the temperature reached 690 °C, followed by a dc voltage of 640 V between the gas shower head and the heating stage to create plasma of 40 W in power. The growth process lasted for 10 to 15 min at 725 °C, which gives multiwalled carbon nanotubes of nearly 2 to 3 μm in height.

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