# THz bandwidth optical switching with carbon nanotube metamaterial

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**Abstract:** We provide the first demonstration of exceptional light-with-light optical switching performance of a carbon nanotube metamaterial – a hybrid nanostructure of a plasmonic metamaterial with semiconducting single-walled carbon nanotubes. A modulation depth of 10% in the near-IR with sub-500 fs response time is achieved with a pump fluence of just 10 μJ/cm$^2$, which is an order of magnitude lower than in previously reported artificial nanostructures. The improved switching characteristics of the carbon nanotube metamaterial are defined by an excitonic nonlinearity of carbon nanotubes resonantly enhanced by a concentration of local fields in the metamaterial. Since the spectral position of the excitonic response and metamaterial plasmonic resonance can be adjusted by using carbon nanotubes of different diameter and scaling of the metamaterial design, the giant nonlinear response of the hybrid metamaterial – in principle – can be engineered to cover the entire second and third telecom windows, from O- to U-band.



**OCIS codes:** (160.3918) Metamaterials; (130.4815) Optical switching devices.

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## 1. Introduction

Nanoscale sized ultrafast optical modulators are key elements for the development of integrated all-optical signal processing circuits. Ultrafast modulation of light with light on the nanoscale is a challenging task: optical nonlinearities in conventional nonlinear materials are generally too weak to alter the intensity of light significantly on a sub-wavelength scale [1], semiconductor based devices suffer from slow response times [2] and propagating surface plasmon based structures capable of efficient ultrafast light modulation have μm-scale dimensions [3]. Nanoscale sized devices with improved nonlinear optical properties can be engineered with the use of metamaterials – artificial media offering a wide and rapidly expanding range of new photonic functionalities [4]. Few metamaterial-based structures featuring enhanced all-optical switching properties in the THz [5] and near-IR [6, 7] spectral regions have been demonstrated recently. In these studies enhanced switching performance is achieved in metamaterial structures hybridized with active nonlinear media. In such hybrid structures, small changes in the refractive index of the active medium induced by non-resonant photoexcitation tune the metamaterial plasmonic resonance which results in significant modulation of the metamaterial transmission near the resonance. Another powerful opportunity for enhancement of the nonlinear optical response of metamaterial-based structures, not widely implemented so far, is in exploiting effects of resonant concentration of local fields in the vicinity of a metamaterial [8, 9].

Recently we have introduced the carbon nanotube metamaterial [10] – a hybrid structure of a plasmonic metamaterial with semiconducting single-walled carbon nanotubes (CNTs) which employs a combination of both the approaches mentioned above for enhancement of nonlinear optical response. This is achieved by spectral matching of the absorption resonance of the nonlinear active medium (CNTs) with the metamaterial plasmonic resonance. The

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nonlinear optical response of the CNT metamaterial is defined by the effect of the metamaterial transmission modulation as a result of changes in the refractive index of the active medium; however the nonlinear response of the active medium itself is significantly enhanced by a concentration of local fields in the metamaterial under the conditions of resonant excitation. We show that this results in a substantial improvement of the nonlinear optical performance of the CNT metamaterial in comparison with the other previously reported analogous structures.

Our choice of using semiconducting single-walled CNTs as an active medium was dictated by the nanotubes' unique nonlinear optical properties: in addition to high third-order susceptibility at the excitonic absorption resonances, CNTs exhibit sub-picosecond recovery times [11, 12] making them an extremely promising material for applications in ultrafast all-optical modulators.

In our previous study using a single beam measurement configuration we have demonstrated the exceptionally strong nonlinear optical response of the CNT metamaterial in the near-IR [10]. In this letter we present results of the first experimental study of the nonlinear dynamics of the CNT metamaterial and demonstrate ultrafast relaxation of optical nonlinearities with a sub-500 fs recovery time. We also show experimentally that employment of resonant local field concentration in plasmonic nanostructure enables one to substantially decrease the switching intensity of metamaterial-based all-optical modulators.

## 2. Experimental methods

The plasmonic nanostructure used in the CNT metamaterial is formed by an array of asymmetrically split ring resonators and belongs to the class of metamaterials where weak coupling of the plasmonic excitation mode to the free-space radiation modes creates narrow resonances with asymmetric, Fano-like dispersion [13, 14]. This type of metamaterial resonance has a high quality factor and is strongly sensitive to the local environment [14]. The high value of the quality factor is crucial for efficient concentration of local fields while the sensitivity of the metamaterial resonance to the local environment facilitates significant changes of metamaterial transmission in response to small nonlinear changes in the refractive index of the CNT layer. The metamaterial structure shown in Fig. 1(a) was fabricated by focused ion beam milling in a 50 nm thick gold film deposited on a 100 nm thick $Si_3N_4$ membrane. The nanostructure was then covered by a ~50 nm thick layer of CNTs using spray-coating. We used purified single-walled CNTs with an average diameter of 1.4 nm synthesized by the electric arc discharge method. This method produces a natural mixture of CNTs with a 2 to 1 ratio of semiconducting to metallic CNTs. To enable comparison of the nonlinear optical response of the CNT metamaterial with the response of CNTs we have fabricated a square window in the gold film next to the metamaterial array so the layer of CNTs formed during the deposition process had the same thickness on the metamaterial and on the $Si_3N_4$ membrane window. More details on the CNT metamaterial sample fabrication and characterization by scanning helium ion microscopy can be found elsewhere [10]. The spectral position of the plasmonic resonance of the uncovered metamaterial $\lambda_p$ depends on the period of the metamaterial $D$ while deposition of the layer of CNTs on top of the metamaterial results in a redshift of the plasmonic absorption peak to $\lambda_p$*, as can be seen from Fig. 1(b). Using a combinatorial approach we have selected for optical studies the structure with period $D$ = 839 nm where the plasmonic resonance in the hybrid structure (after CNT deposition) is spectrally matched to the CNT excitonic absorption resonance.

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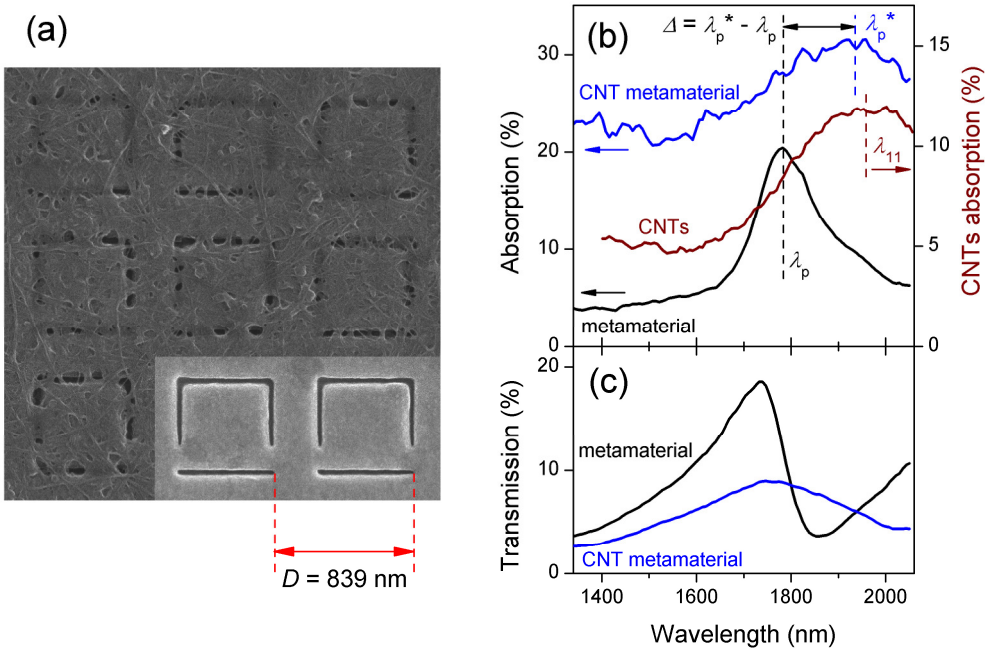


Fig. 1. Carbon nanotube metamaterial. (a) Scanning helium ion microscope image of the plasmonic metamaterial covered by CNTs. Inset shows image of two unit cells of uncovered metamaterial. (b) Left scale – plasmonic absorption resonances of uncovered metamaterial (black) and metamaterial covered by CNTs (blue). Right scale – excitonic absorption spectra of the bare CNT layer (wine). (c) Transmission spectra of uncovered metamaterial (black) and metamaterial covered by CNTs (blue). Presented spectra were measured on a CRAIC microspectrometer and correspond to the linear (low intensity) regime.

The nonlinear optical response of the CNT metamaterial was investigated by degenerate (single-colour) non-collinear pump-probe. Both pump and probe beams from a tunable 200 fs OPO system pumped by a Ti:Sapphire laser operating at 80 MHz were focused to a diameter of ~30 μm. The metamaterial array has a smaller size (22 × 22 $\mu m^2$) so it was illuminated uniformly by both pump and probe beams. The intensity of the probe pulse was 10-20 times weaker than the intensity of the pump pulse. The optical response of metamaterial is polarization sensitive and the direction of linear polarization of both pump and probe beams was chosen to excite the dark-mode resonance in the metamaterial.

### 3. Experimental results

Figure 2 presents pump-probe curves measured on the CNT metamaterial over a wide spectral range covering the overlapped CNT excitonic and metamaterial plasmonic resonances. Using this pump-probe data we have retrieved the spectral dependencies of the nonlinear changes of transmission $\Delta T/T$ of the CNT metamaterial at several delays after the pump pulse (shown in Fig. 3). The differential transmission spectrum measured on the bare CNT layer at the same pump fluence is shown in Fig. 3 by open circles for comparison. From the experimental data shown in Fig. 2 and Fig. 3 one can see that i) relaxation of pump-induced nonlinear changes of transmission of the CNT metamaterial is ultrafast in the whole measured spectral range, ii) the nonlinear optical response of the CNT metamaterial has a complex sign-changing spectral dispersion, and iii) the modulation depth $\Delta T/T$ in the CNT metamaterial is much higher than in the bare CNT layer.

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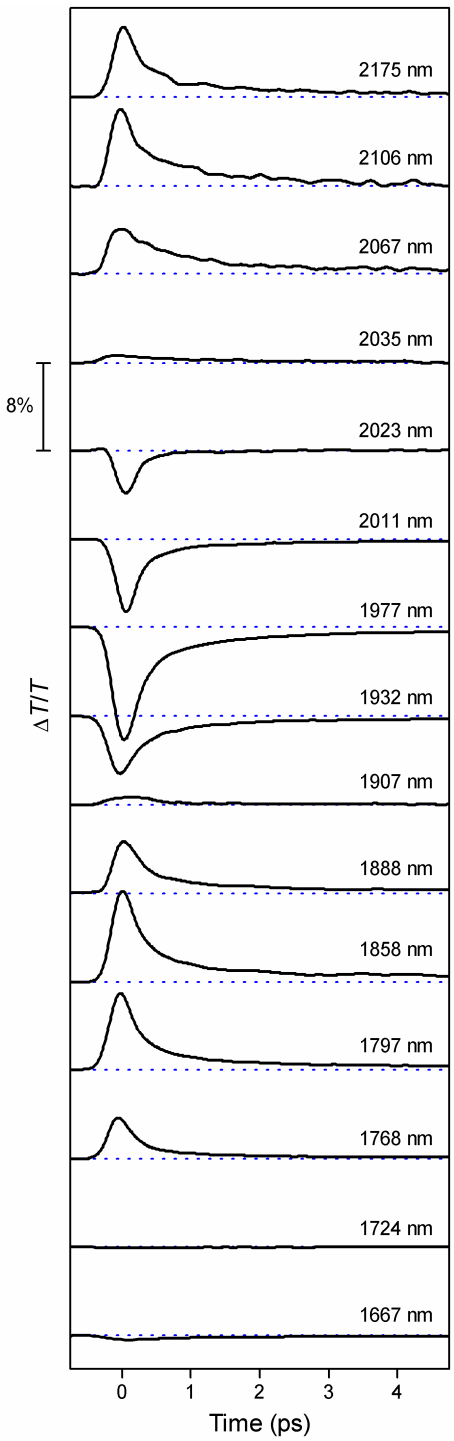


Fig. 2. Spectrally resolved transient changes of transmission of the CNT metamaterial. Pump fluence is 10 μJ/cm$^2$ (power density – 50 MW/cm$^2$).

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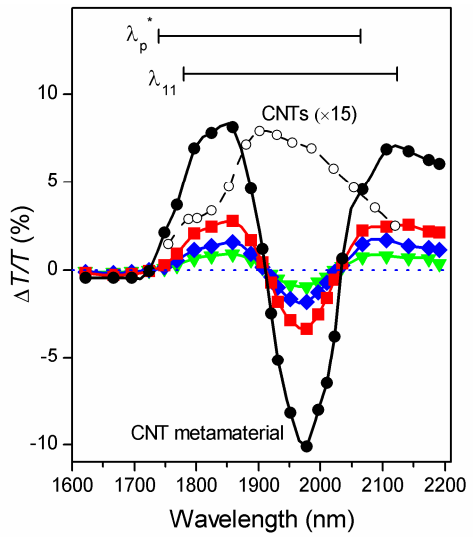


Fig. 3. Differential transmission spectra of the CNT metamaterial (solid symbols) taken at 0 (black circles), 0.5 (red squares), 1 (blue diamonds) and 2 ps (green triangles) after the pump pulse. Differential transmission spectrum of the bare CNT layer measured at 0 ps is shown by open circles. Bars indicate spectral regions (FWHM) of metamaterial (covered by CNTs) plasmonic ($\lambda_p$*) and CNT excitonic ($\lambda_{11}$) resonances.

To further investigate the transient behavior of the nonlinear optical response of the CNT metamaterial we have compared the nonlinear dynamics of the CNT metamaterial with the dynamics of the bare CNT layer. Pump-probe curves measured at two characteristic spectral points 1820 nm and 1960 nm, where $\Delta T/T$ in the CNT metamaterial reaches its extremum values (see Fig. 3), are shown in Fig. 4. For ease of comparison in Fig. 4(b) we present absolute values of $\Delta T/T$, as in the spectral region around 1960 nm nonlinear optical response of the CNT metamaterial is negative (transmission decreases with increasing the power). It is clearly seen that, in terms of absolute values of $\Delta T/T$, the nonlinear dynamics of the CNT metamaterial nearly replicates the dynamics of the bare CNT layer with ~25 and ~15 times higher amplitudes at 1820 nm and 1960 nm, respectively. We have found that each of the pump-probe traces measured on the CNT metamaterial in the spectral range of 1600-2200 nm can be fitted with a bi-exponential decay function where the fast component typically has decay times in the sub-500 fs range while the decay time of the slow component is in the few ps range. The observed nonlinear dynamics of the CNT metamaterial replicate the dynamics of CNTs: bi-exponential decay of the nonlinear optical response of CNTs was reported in a number of studies where the fast component was assigned to the intraband, and the slow component to the interband, carrier relaxation in CNTs [15–17]. For potential applications in all-optical switching it is important that the fast component provides the major contribution to the nonlinear dynamics of both the bare CNT layer and the CNT metamaterial. This is exemplified in Fig. 4(b) where the bi-exponential fit for the CNT metamaterial at 1960 nm (shown by thick dashed blue curve) gives decay times of 410 fs and 3 ps and relative amplitudes of 79.5% and 20.5% for the fast and the slow components, respectively.

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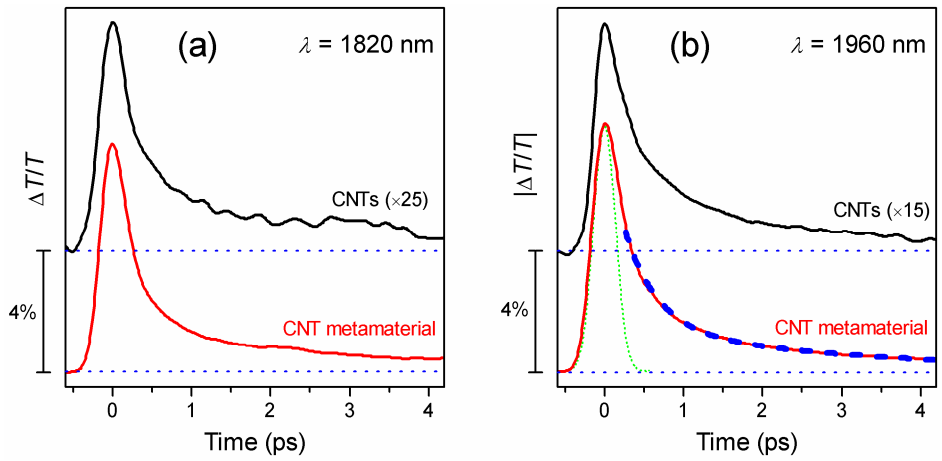


Fig. 4. The time evolution of pump-induced nonlinear changes of transmission measured on the CNT metamaterial (red) and the bare CNT layer (black) at (a) 1820 nm and (b) 1960 nm. Curves shifted vertically for clarity. Pump fluence is 10 μJ/cm$^2$ for all of the presented curves. Dotted green curve on (b) represents autocorrelation trace, thick dashed blue curve shows bi-exponential fit of the pump-probe curve for the CNT metamaterial.

In contrast to previously reported metamaterial-based optical switching devices [6, 7], in the CNT metamaterial the active medium is excited resonantly at the metamaterial plasmonic resonance thus providing an opportunity for enhancement of the nonlinear optical response of the hybrid structure due to field concentration in the plasmonic resonator. To evaluate the effect of local field enhancement we have compared the power dependencies of nonlinear changes of transmission of the CNT metamaterial and the bare CNT layer. Figure 5 shows $\Delta T/T$ of the CNT metamaterial (solid red circles) as a function of the pump pulse fluence measured at 1820 nm and 1960 nm at zero delay (during the pump pulse). Power dependence of the bare CNT layer measured at 1960 nm is shown for comparison in Fig. 5(b) by black open circles. The prime nonlinear optical process in single-walled semiconducting CNTs is saturable absorption, which originates from filling of the exited resonant excitonic states and is associated with third-order optical nonlinearities [18]. Saturable absorption in CNTs can be described by a simple two-level saturable absorber model [19] which gives the intensity-dependent absorption coefficient $\alpha(I)$ as:

$$\alpha(I) = \frac{\alpha_0}{1 + I / I_{sat}} + \alpha_{ns} \qquad (1)$$

where $I$ is the intensity of the incident laser pulse, $\alpha_0$ and $\alpha_{ns}$ are low intensity (linear limit) saturable and nonsaturable absorption components, respectively, and $I_{sat}$ is the saturation intensity (the intensity necessary to reduce the saturable part of absorption to half of the linear limit value). For thin, low-absorbance layers Eq. (1) gives for nonlinear changes of transmission:

$$\frac{\Delta T}{T}(I) = \frac{(\Delta T / T)_{sat}}{1 + I_{sat} / I} \qquad (2)$$

where $(\Delta T/T)_{sat}$ is the maximum possible change of transmission, corresponding to infinitely high intensity of incident pulse. In our study the CNT layer had a thickness of ~50 nm and absorbs about 10% of the incident intensity so we used Eq. (2) to fit experimental data (see dashed curve in Fig. 5(b)). We present the results in terms of fluence of the light excitation for ease of comparison with other reports. For the bare CNT layer, from fitting of experimental power dependence we have estimated the saturation fluence as 15 μJ/cm$^2$ at 1960 nm which is in agreement with previously reported saturation fluences in thin CNT films ranging from 9.5

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μJ/cm$^2$ [17] to 57 μJ/cm$^2$ [20]. From Fig. 5 it can be seen that power dependencies of the nonlinear optical response of the CNT metamaterial follow essentially the same saturation law described by Eq. (2) but with lower $I_{sat}$ and higher absolute values of $(\Delta T/T)_{sat}$. The fit of experimental data for the CNT metamaterial at 1820 nm and 1960 nm gives similar saturation fluences of 5.5 μJ/cm$^2$ and 5.2 μJ/cm$^2$, respectively, which is ~3 times lower than saturation fluence of the bare CNT layer measured at 1960 nm. The observed significant decrease of the saturation intensity in the CNT metamaterial as compared to the bare CNT layer provides experimental evidence for enhancement of the nonlinear optical response of the layer of CNTs placed in the vicinity of metamaterial due to local field concentration in the plasmonic resonator.

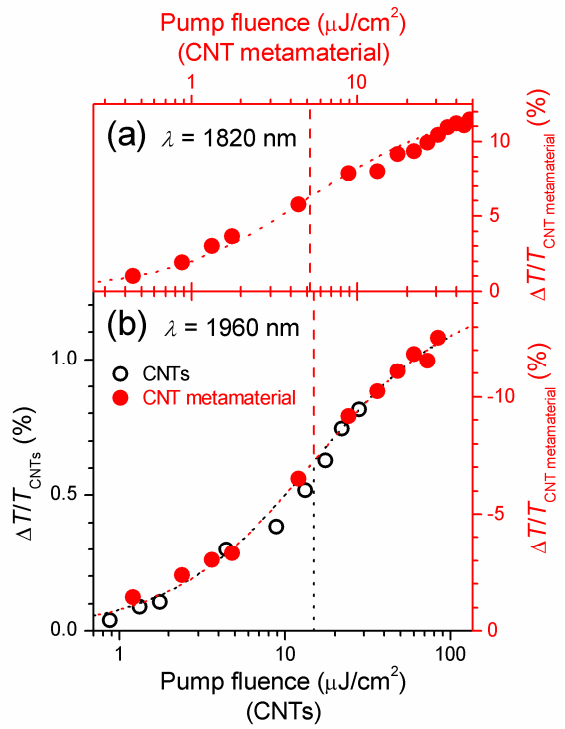


Fig. 5. Power dependencies of nonlinear changes of transmission of the CNT metamaterial (solid red circles, top and right scales) measured at (a) 1820 nm and (b) 1960 nm. Black open circles on (b) show power dependence of the bare CNT layer (left and bottom scales) measured at 1960 nm. Vertical lines indicate saturation fluences for CNTs (black dotted line) and the CNT metamaterial (red dashed line). Dashed curves show theoretical fits using Eq. (2).

## 4. Discussion

The nonlinear optical response of the CNT metamaterial can be explained by taking into account the interaction between the metamaterial plasmonic and CNT excitonic resonances. The plasmonic resonance of the uncovered metamaterial $\lambda_p$ appears at ~1800 nm as a sharp feature in transmission and corresponding peak in the absorption spectrum (see Fig. 1(b) and Fig. 1(c)). Deposition of the CNT layer on the metamaterial results in a redshift $\Delta = \lambda_p^* - \lambda_p$ and damping of metamaterial plasmonic resonance (compare spectra for uncovered metamaterial and metamaterial covered by CNTs in Fig. 1(b) and Fig. 1(c)). Redshift originates from the real part of the refractive index of the highly polarizable CNTs while damping is due to absorption of plasmon evanescent waves in the layer of CNTs and defined by the imaginary part of CNT refractive index. We have checked experimentally that the optical response of the uncovered metamaterial is linear in the range of wavelengths and fluences used in our study. Accordingly, the nonlinear optical response of the CNT

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metamaterial is governed by nonlinear processes in the layer of CNTs. The excitonic absorption resonance $\lambda_{11}$ of the CNTs is spectrally overlapped with the plasmonic resonance $\lambda_p$* of the metamaterial covered by CNTs. In the nonlinear regime this leads to two effects: i) the nonlinear optical response of the CNTs is enhanced due to resonant concentration of the local field in the plasmonic resonator, ii) the decrease of absorption (bleaching) in CNTs and associated changes of the real part of refractive index of CNTs leads to recovery of metamaterial plasmonic resonance towards resonance of uncovered metamaterial. This results in a complex spectral dependence of nonlinear changes of transmission of the CNT metamaterial containing spectral regions with positive and negative values of $\Delta T/T$.

The experimentally observed spectral dispersion of the nonlinear optical response of the CNT metamaterial was reproduced in numerical simulations in COMSOL (see Fig. 6). To do this we have first calculated the linear optical response of uncovered metamaterial using the parameters of real metamaterial structure (thicknesses and dielectric constants of gold and $Si_3N_4$ and the geometry of the metamaterial slits). Calculated transmission and absorption spectra show good correspondence with the experimental data (compare corresponding curves in Fig. 6 and Fig. 1). To calculate optical response of the hybrid structure we have added to the simulations a 50 nm thick layer of dielectric representing the layer of CNTs on the top of metamaterial. The nonlinear optical response of layer of CNTs, which defines nonlinear response of the whole hybrid structure, was taken into account in two different spectral dispersions of refractive index of the CNT layer corresponding to low (linear limit) and high (nonlinear regime) intensities of incident light. The dispersions of the imaginary part of refractive index the CNTs were evaluated from experimental transmission data reasonably assuming low level of reflectivity and scattering losses. Dispersions of real part of refractive index were then calculated using the Kramers-Kronig relation. For the linear limit (low intensity) we used the experimentally measured absorption spectra of the bare CNT layer shown on Fig. 1(b). Absorption spectra of the layer of CNTs on the plasmonic resonator at high intensity, shown in Fig. 6(a) by red dotted line, was estimated taking into account linear absorption and differential transmission spectra of the bare CNT layer (shown in Fig. 3), and rescaling factor originating from field enhancement. The resulting transmission spectra of the hybrid structure calculated for low and high incident intensities are shown in Fig. 6(c) by blue solid and red dotted curves, respectively. The ratio of these two spectra, shown in Fig. 6(d) by solid black line, represents simulated nonlinear changes of transmission of the CNT metamaterial. The calculated dispersion of the nonlinear optical response of the CNT metamaterial contains spectral regions with positive and negative signs of $\Delta T/T$ and is in excellent agreement with experimental data. We note that two fitting parameters were used in our simulations to achieve good quantitative correspondence with the experimental data. These parameters are the average value of the real part of refractive index of the CNT layer across the absorption resonance assumed to be ~1.5 and a rescaling factor ~2 that accounts for higher level of nonlinear absorption of CNTs deposited on the metamaterial. The first parameter defines the spectral shift and the second one the amplitude of the calculated spectral dependence of $\Delta T/T$. The shape of spectral dependence of $\Delta T/T$ does not change much with small variations of fitting parameters. We have also developed an analytical model to describe the interaction between classical (metamaterial) and quantum (CNTs) resonant structures. A detailed description of the model and results will be presented elsewhere [21].

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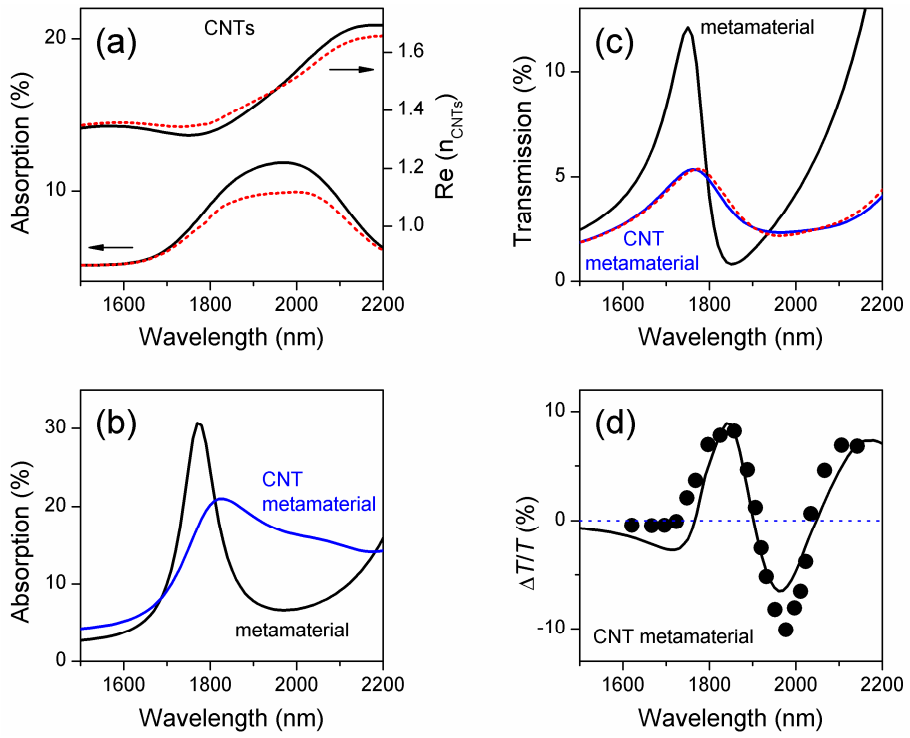


Fig. 6. Modelling of nonlinear optical response of the CNT Metamaterial. (a) Absorption (left scale) and real part of refractive index (right scale) of the layer of CNTs in the CNT metamaterial at low (black solid line) and high (red dotted line) excitation intensities. (b) Absorption of the uncovered metamaterial (black) and the CNT metamaterial (blue) at low intensities. (c) Transmission of uncovered metamaterial (black) and metamaterial covered by CNTs at low (blue solid line) and high (red dotted line) excitation intensities. (d) Comparison of calculated (solid line) and experimental (circles) spectral dispersion of $\Delta T/T$ of the CNT metamaterial.

The considerations discussed above and the results of numerical simulations refer to a steady state corresponding to continuous pumping of the CNT metamaterial. To understand the experimentally observed nonlinear dynamics it is important to point out the fact that the recombination of excited carriers in CNTs is essentially non-radiative. From the microscope image shown in Fig. 1(a) it is seen that most of the CNTs in the hybrid structure are aggregated in bundles. It is well established that while in individual (suspended) CNTs efficient band gap photoluminescence (PL) can be observed, in bundled CNTs PL appears to be strongly quenched [22]. The reason for PL quenching in CNT bundles is suggested to be intertube energy transfer from the semiconducting to the metallic nanotubes within each bundle, followed by rapid non-radiative carrier cooling in the metallic nanotubes [22]. It has been shown recently that in a hybrid structure where the metamaterial plasmonic resonance interacts with optically active resonances in PbS quantum dots, the cavity quantum electrodynamic Purcell effect leads to multifold enhancement of PL intensity [23] and, presumably, to acceleration of the recombination rate in quantum dots. In contrast, the rate of non-radiative recombination in the CNT layer in the CNT metamaterial is not affected by the presence of the metamaterial nano-resonator despite the strong resonant interaction between CNT excitonic and metamaterial plasmonic resonances in the hybrid structure. In the interacting system of the CNT metamaterial energy can be transferred from the plasmonic resonator to CNTs. Manifestation of this energy transfer is the observed decrease of saturation intensity of the nonlinear optical response in the CNT metamaterial as a result of additional pumping of the CNT layer by the local field concentrated in the metamaterial. Energy transfer in the opposite direction (from CNTs to the plasmonic resonator) is negligible as the

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excitation created in the CNTs rapidly dissipates to the surrounding medium non-radiatively. Non-radiative relaxation of excitation in the layer of CNTs on the timescale of few hundreds of fs however is still much slower then the period of plasmonic oscillations in metamaterial (few fs – corresponding to the light period of the laser pulse) and the decay of plasmonic oscillations (few tens of fs as can be estimated from quality factor of plasmonic resonator: $Q \approx 5.5$ for the metamaterial covered by CNTs). We therefore assume that the relaxation of excitation in the CNT layer adiabatically tunes the plasmonic resonance and, as a result, the nonlinear dynamics of the CNT metamaterial replicate the dynamics in the bare CNT layer as is observed in the experiment. We note that, from the point of view of applications for ultrafast light modulators, bundling of CNTs and quenching of PL can be considered as an advantage. Indeed, reported radiative lifetimes in individual CNTs are typically lying in the range from 10 ps to 100 ps [15, 16, 24–26] while in bundled CNTs non-radiative relaxation occurs on sub-picosecond scale [11, 12, 15, 17, 20].

Finally, we compare modulation characteristics of the CNT metamaterial with the characteristics of the analogous nanoscale sized ultrafast optical switching structures reported previously – Ag/α-Si/Ag fishnet nanostructures [6, 7] and nanostructured gold medium [9]. First, we note that all of the structures (including the CNT metamaterial) have comparable thicknesses (100-200 nm) and transmission levels at the wavelength of modulated light (5-15%). As the modulation depth depends strongly on the pump fluence we compare the pump fluences required to induce some fixed change of transmission. In Ag/α-Si/Ag nanostructures and the nanostructured gold medium similar pump fluences of 100 μJ/cm$^2$ [6, 7] and 80 μJ/cm$^2$ [9], respectively, are required to induce a modulation depth of $\Delta T/T = 10\%$. In the CNT metamaterial the same switching ratio is achieved with an order of magnitude lower pump fluence of just 10 μJ/cm$^2$. Such a dramatic decrease of switching fluence is achieved due to: i) use of CNTs as nonlinear medium where third-order nonlinear susceptibility is significantly enhanced due to quantum confinement of electron-hole motion in a one-dimensional space [27] and ii) concentration of local fields in the plasmonic resonator resulting in additional pumping of the nonlinear medium. With increasing pump fluence, modulation depth in Ag/α-Si/Ag nanostructures and nanostructured gold medium increases nearly linearly reaching maximum reported value $\Delta T/T = 70\%$ at 900 μJ/cm$^2$ [7]. In the CNT metamaterial we observed saturation of the modulation depth with the maximum value estimated as $(\Delta T/T)_{sat} = 15\%$ (see Eq. (2) and Fig. 5). Although the saturation of modulation depth limits switching performance of CNT metamaterial we believe it can be significantly improved by optimizing the thickness of CNT layer and, especially, by using selected semiconducting CNTs [28]. Indeed, the value of $(\Delta T/T)_{sat}$ in CNT metamaterial is limited by nonsaturable absorption in CNTs. Metallic CNTs in the natural mixture of CNTs used in this study do not contribute to the nonlinear optical response but induce additional losses. Accordingly, removing metallic CNTs from the mixture can significantly improve switching characteristics of the CNT metamaterial. The recovery time of the CNT metamaterial (<500 fs) which defines switching speed is slightly shorter than recovery times of Ag/α-Si/Ag nanostructures (750 fs [6] and 600 fs [7]) but significantly longer than response time of the nanostructured gold medium (~40 fs [9]). When comparing nonlinear characteristics of nanostructures discussed above it is important to note that Ag/α-Si/Ag nanostructures can be switched only with a visible pump pulse, necessary to photoexcite carriers above the α-Si band gap, while changes of transmission are probed in the near-IR. This eliminates possibility of using Ag/α-Si/Ag nanostructures as nonlinear optical devices working with a single laser beam. On the contrary, a nanostructured gold medium can be used as very efficient optical limiter or saturable absorber, but its capabilities of light with light modulation are limited by requirement for pump and probe beams to be coherent (this requirement originates from the two-photon nonlinear absorption process employed in the nanostructured gold medium [9]). In contrast, the CNT metamaterial can be both pumped and probed with either coherent or frequency shifted beams in the near-IR and it can also work as an efficient nonlinear device with a single beam as was shown in our previous study [10]. From this comparison, one can see that the optical switching performance of the CNT metamaterial has a number of

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significant advantages in comparison with previously reported devices with analogous functionalities.

## 5. Conclusions

In summary, we have demonstrated that the CNT metamaterial possesses exceptional nonlinear optical properties in the near-infrared part of the spectrum – high modulation depth over a nanoscale length with a low switching power and an ultrafast relaxation time, exceeding the characteristics of previously reported materials. We argue that the CNT metamaterial benefits from simplicity of fabrication and is very robust: we observed no noticeable degradation of its nonlinear optical response neither after one year of shelf life nor after many hours of continuous illumination by a high intensity pulse laser during experiments. Another advantage of the CNT metamaterial is in its spectral flexibility. While in the present study we demonstrated optical switching in the spectral region around 1900 nm, resonant nonlinear properties of the CNT metamaterial can be easily tuned in the spectral range of 1-2 μm with CNTs of different diameter [29] and appropriate scaling of the metamaterial, thus covering the entire second and third optical telecom spectral windows.

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