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Nonlinear reshaping of THz pulses with graphene metamaterials

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We study the propagation of electromagnetic waves through a slab of graphene metamaterial composed of the layers of graphene separated by dielectric slabs. Starting from the kinetic expression for two-dimensional electric current in graphene, we derive a novel equation describing the nonlinear propagation of THz electromagnetic pulses through the layered graphene-dielectric structure in the presence of losses and nonlinearities. We demonstrate strong nonlinearity-induced reshaping of transmitted and reflected THz pulses through the interaction with the thin multilayer graphene metamaterial structure.

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Graphene, a two-dimensional lattice of carbon atoms, is known to exhibit a wide range of unique electronic and optical properties [1, 2]. One of the key features of graphene is the linear dispersion of electrons close to the band-edges which is similar to the dispersion of ultra-relativistic Dirac fermions [1, 2]. In particular, this property leads to the square-root dependence of the electron cyclotron frequency on the magnetic field, much larger separation of the Landau levels in graphene, and consequently to the possibility to observe quantum Hall effect at room temperatures.

Graphene structures have been suggested for many applications in tunable THz devices for control of both electrons and electromagnetic waves [3–10]. It was also suggested that graphene can be employed as a material with remarkable nonlinear response in THz and optical frequency ranges [8], and the study of propagation of linear and nonlinear electromagnetic waves in the structures with graphene has attracted a great interest [3, 4, 6–9].

We would like to notice that a recent concept of metatronics [1] is merging with graphene physics. The three-level approach to graphene metamaterials suggested in Refs. [5–7] includes a metamaterial-inspired study of quantum-mechanical electron probability waves [5–7] employing highly controllable electron waves for creating devices of solid-state graphene electron optics [7], the use of such elements as electronic resonators [5–7] and waveguides, diffraction gratings [7], electron lenses [11], as well as the electronic-based tuning of the electromagnetic characteristics of such structures as, for example, in carbon nanotubes-graphene-insulators [6, 7], and finally the interaction between electronic modes and electromagnetic waves.

Recently, a novel concept of metamaterials based on multilayer graphene structures has been suggested [12], and it was shown theoretically that such graphene structures can demonstrate a giant Purcell effect that can be used for boosting the THz emission in semiconductor devices. In this Letter, we demonstrate the importance of nonlinear effects in the propagation of electromagnetic pulses through layered graphene-dielectric structures, where we take into account the nonlinear response of graphene. In particular, we predict strong reshaping of THz pulses in the transmission regime.

We are interested in the possibility of reshaping and self-action effects for short THz pulses propagating through a graphene metamaterial composed of a multilayered graphene-dielectric structure, as shown in Fig. 1. The alternating graphene layers and dielectric slabs have their interfaces parallel to the plane (x, y), while electromagnetic waves propagate in the z direction. We assume that the wave is linearly polarized having the E_x component, $\mathbf{E} = (E_x, 0, 0)$, and it propagates normally to the structure.

Next, we assume that two-dimensional graphene layers have the equilibrium electron concentration $n_{20} \sim \sim 10^{12}\,\mathrm{cm}^{-2}$, and they possess electrodynamic nonlinear response that originates from electron nonlinearity due to the pseudo-relativistic dispersion. The corresponding

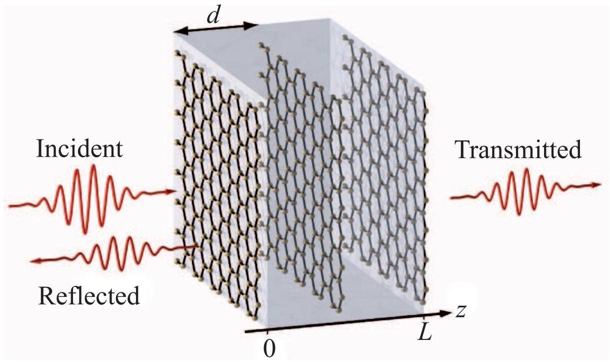

Fig. 1. Schematic of the pulse propagation through a graphene metamaterial composed of graphene layers separated by dielectric slabs. Spacing between the layer is d, and the total thickness of the structure is L

expression for the surface electron current can be obtained from the kinetic equation in the following explicit form [13],

$$i_s = \frac{ev_\mathrm{F}k_\mathrm{F}}{\pi}\frac{\Psi}{\sqrt{1+\Psi^2}}, \quad (1)$$

where $\Psi = eA/p_\mathrm{F}$ is the normalized vector potential of the electromagnetic field, $A = \partial E/\partial t$, $p_\mathrm{F} = hk_\mathrm{F}$ is the Fermi momentum, where $k_\mathrm{F} = (\pi n_{20})^{1/2}$, and $v_\mathrm{F} = 10^6$ m/s is the Fermi velocity in graphene. We introduce an analogue of the effective electron mass $m^* = p_\mathrm{F}/v_\mathrm{F} \approx (0.01–0.03)m_0$, where m_0 is the mass of free electron. The electron nonlinear response demonstrates saturation, and Eq. (1) is similar to the volume nonlinearity of narrow gap semiconductors such as n-InSb.

Next, we substitute Eq. (1) into the equation describing the wave propagation in a layered structure,

$$c^2\Delta E - \frac{\partial^2 D}{\partial t^2} - \\ - \omega_{pg}^2 l_n \sum_j \frac{\partial}{\partial t}\left[\frac{A}{\sqrt{1+(eA/p_\mathrm{F})^2}}\right]\delta(z-z_j) = 0, \quad (2)$$

where the sum is taken over all graphene layers with positions $z+j$; D is the linear electric induction in the dielectric layers, and $\omega_{pg}^2 l_n = (4\pi e^2 n_{20}/m^*)$. The typical spatial scale l_n used below in numerical simulations is assumed to be in the THz frequency range, $l_n = 10^{-2}$ cm.

We consider moderate nonlinearities and look for solutions of Eq. (2) in the form of asymptotic series by extracting the slowly varying envelope,

$$E = \frac{1}{2}C(z,\rho,t)\exp(i\omega t) + \text{c.c.}, \quad (3)$$

where C is the amplitude slowly varying in time t, but with an arbitrary dependence on z and ρ. Using Eq. (3), we obtain the following expression for D in the dielectric layers,

$$D \approx \frac{1}{2}\left(\varepsilon C - i\frac{d\varepsilon}{d\omega}\frac{dC}{dt} - \frac{1}{2}\frac{d^2\varepsilon}{d\omega^2}\frac{d^2C}{dt^2}\right)e^{i\omega t} + \text{c.c.} \quad (4)$$

After substituting Eqs. (3), (4) into Eq. (2), we derive the nonlinear parabolic equation for the wave amplitude C,

$$\frac{\partial C}{\partial t} + \frac{ic^2}{2\omega\varepsilon^{(1)}}\left(\frac{\partial^2 C}{\partial z^2} + \Delta_\perp C\right) + \\ + \frac{i}{2\omega\varepsilon^{(1)}}\left(\left[\omega^2\varepsilon - \sum_j \omega_{pg}^2 l_n \delta(z-z_j)(1+i\nu/\omega)\right] + \right. \\ + \omega_{pg}^2 l_n \sum_j \left\{1 - \frac{1}{\sqrt{Q_g}}\left[1 - \frac{1}{8}\frac{e^2|C|^2}{(m^* v_\mathrm{F}\omega)^2}\right]\right\} \times \\ \left. \times\, \delta(z-z_j)\right)C = 0, \quad (5)$$

where

$$Q_g = 1 + \frac{e^2|C|^2}{2(m^* v_\mathrm{F}\omega)^2},$$

and $\varepsilon^{(1)} = \varepsilon + (\omega/2)(d\varepsilon/d\omega)$ is the expansion of the dielectric function due to the frequency dispersion. The derivation takes into account the frequency ν of electron collisions in the graphene layers that is typically about $\nu = (0.3–3)\cdot 10^{12}$ s^{-1}.

As follows from Eq. (5), the nonlinearity in graphene is of the self-focusing type, in both longitudinal z and transverse ρ directions. Equation (5) should be accompanied by boundary conditions derived from the continuity condition for the tangential components E_x and H_y. We assume that the layered structure is surrounded by vacuum. At $z < 0$, both the incident and reflected waves are present, at $z > L_z$ only the transmitted wave exists. In the parabolic approximation, the boundary conditions can be obtained in the form,

$$\left.\frac{\partial C}{\partial z}\right|_{z=0} - ik_0 C|_{z=0} = -2ik_0 E_i(\rho,t), \quad (6)$$

$$\left.\frac{\partial C}{\partial z}\right|_{z=L_z} + ik_0 C|_{z=L_z} = 0, \quad (7)$$

where $k_0 = \omega/c$, and $E_i(\rho,t)$ is the amplitude of the incident wave at the input. The derivation method employed here is similar to the approach used earlier for the analysis of magnetostatic solitons in magnetic films [14–16] and strongly nonlinear waves in nonlinear metamaterials [17, 18], but the system of equations (5)–(7) describes a novel type of nonlinear model not known previously.

First, we study the normal linear transmission through the metamaterial structure that is characterized by the standard transmission and reflection coefficients presented in Fig. 2. Due to periodicity of the

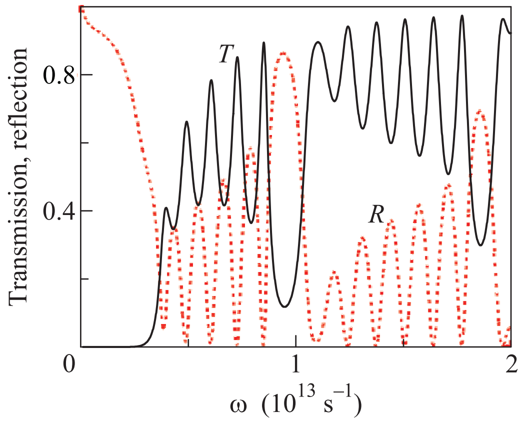

Fig. 2. Transmission T (solid line) and reflection R (dashed line) of linear THz waves vs. frequency, for relatively small dissipation, $\nu = 3 \cdot 10^{11} \mathrm{s}^{-1}$

structure, the transmission is characterized by the Bragg reflection gap, and the results correspond qualitatively to the analysis recently presented in Ref. [19].

Next, we study the nonlinear propagation of short THz pulses through this graphene metamaterial. For this case, we select the incident wave in the form of a super-Gaussian pulse,

$$E_i(\rho, t) = E_{i,0} \exp\left(-\frac{\rho^4}{\rho_0^3}\right) \exp\left[-\frac{(t-t_1)^4}{t_0^4}\right], \qquad (8)$$

and assume that the structure includes seven dielectric layers ($\varepsilon = \varepsilon^{(1)} = 4$) with the thickness of each layer $h = 5 \cdot 10^{-3}$ cm. As a result, there are eight graphene layers placed at the positions $z = 0, h, 2h$ etc, as shown in Fig. 1. We also assume the two-dimensional electron density of $n_{20} = 2.5 \cdot 10^{12}\,\mathrm{cm}^{-2}$, and consider THz pulses with the carrier frequency of $\omega = 10^{13}\,\mathrm{s}^{-1}$ and the half-width of $\rho_0 = 0.025$ cm. The pulse amplitude $|E|$ is normalized to 20 kV/cm.

Figures 3 and 4 present the characteristic examples of our numerical results for the simulation of nonlinear propagation of THz pulses through the metamaterial structure. In Figs. 3a–d, the curve *1* (solid) shows an incident pulse, the curve *2* (dashed) shows a reflected pulse; and the curve *3* (dotted) is used to show the transmitted pulse. In Fig. 4, we show the nonlinear transmission and reflection coefficients $T_{nl} = |E|^2_{t,\max}/|E|^2_{in,\max}$ and $R_{nl} = |E|^2_{r,\max}/|E|^2_{in,\max}$, both normalized to the maximum value of the intensity of an incident pulse $|E|^2_{in,\max}$. Here $|E|^2_{t,\max}$ and $|E|^2_{r,\max}$ are the maximum values of the intensities of transmitted and reflected pulses, respectively, taken at the axis of symmetry ($\rho = 0$); the index "nl" emphasizes the role of nonlinearity.

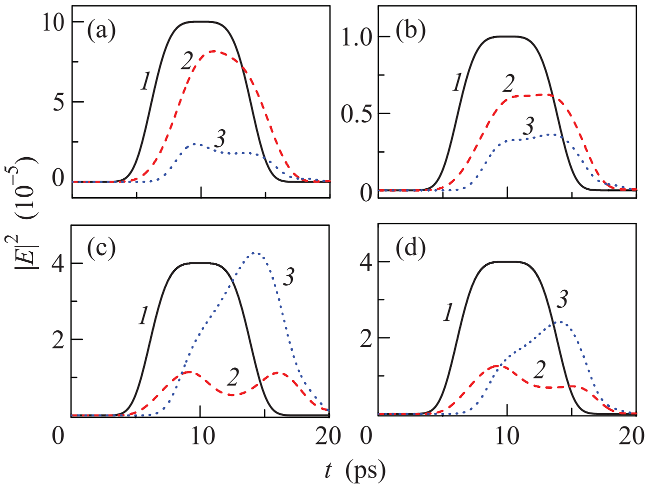

Fig. 3. Propagation of THz pulses with different input amplitudes through the graphene metamaterial structures with different strength of dissipation. Curve *1* – incident pulse, curve *2* – reflected pulse, and curve *3* – transmitted pulse; (a)–(c) graphene metamaterial with lower dissipation, $\nu = 3 \cdot 10^{11}$ s^{-1}. (d) – Graphene metamaterial with higher dissipation, $\nu = 2 \cdot 10^{12}$ s^{-1}. (a) – Linear regime with relatively large reflection and small transmission. (b) – Input pulse with larger input amplitude, but still in the quasi-linear regime, (c), (d) – strongly nonlinear regime with dominating transmission. The amplitude $|E|$ is normalized to 20 kV/cm

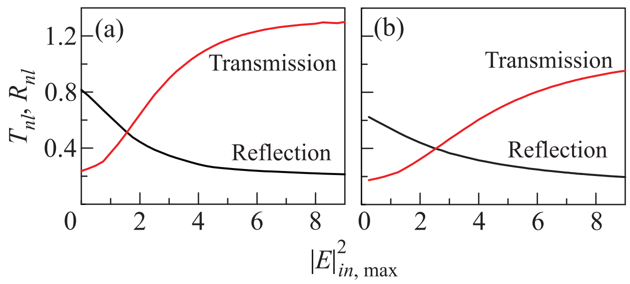

Fig. 4. Transmission $T_{nl} = |E|^2_{t,\max}/|E|^2_{in,\max}$ and reflection $R_{nl} = |E|^2_{r,\max}/|E|^2_{in,\max}$ coefficients as functions of the maximum value of the input pulse intensity $|E|^2_{in,\max}$. The values $|E|^2_{t,\max}$ and $|E|^2_{r,\max}$ are the maximum values of the intensity of transmitted and reflected pulses, respectively. Incident pulse duration is 10 ps. (a) – Graphene metamaterial structure with lower dissipation, $\nu = 3 \cdot 10^{11}\,\mathrm{s}^{-1}$. (b) – Graphene metamaterial structure with higher dissipation, $\nu = 2 \cdot 10^{12}\,\mathrm{s}^{-1}$. The amplitude $|E|$ is normalized to 20 kV/cm

As follows from a comparison of Figs. 3c, d and a, b, nonlinear response leads to a strong reshaping of all pulses when the input amplitude exceeds some critical value, the nonlinear transition with the dominating transmission is clearly observed in this regime. This effect is due to the nonlinear-induced shift of the Bragg reflection gap that allows a larger part of the input pulse propagates through the structure in the nonlinear regime. Interestingly, the normalized transmission coefficient T_{nl} may exceed the unity in the case of strong nonlinearity, because this value characterizes a ratio between the maxima of the corresponding pulses, and it is not directly linked to the energy characteristics of the pulses.

As is seen from Figs. 3a, b, the value of the input pulse amplitude required for the transition to the strongly nonlinear regime with the dominant transmission is of the order of $E_{i0} \sim 20\,\mathrm{kV/cm}$. For the graphene structures with larger dissipation (see the example in Fig. 4b), the incident pulse amplitude required for the transition to the strongly nonlinear regime of enhanced transmission is larger than that corresponding to the lower dissipation, see the example in Fig. 4a. At the same time, at the transition point where $T_{nl} = R_{nl}$, the value of the transmission coefficient T_{nl} is larger than for the structures with the smaller dissipation (see Fig. 4a).

Therefore, the main effect revealed by our study is a strong pulse reshaping in the nonlinear transmission of THz pulses observed in an interplay of the reflection and transmission regimes due to a nonlinearity-induced shift of the Bragg reflection gap induced by only a few subwavelength layers of graphene of the metamaterial. The effect itself and the shape of the transmitted pulse depend on the width of the input pulse.

The next step would be to analyze an oblique incidence and a possibility of nonlinear switching of the transmission coefficient as a function of the input pulse amplitude for longer pulses. In addition, we have found an interesting dependence of nonlinear output on transverse nonlinear effects and even competition between transverse and longitudinal self-focusing; the results will be reported elsewhere.

In conclusion, we have studied the propagation of electromagnetic pulses through a finite-length graphene metamaterial composed of a periodic array of graphene layers separated by dielectric slabs. We have derived a novel nonlinear model of the wave propagation taking into account the electron nonlinearity in graphene. We have predicted a possibility of nonlinear reshaping of THz pulses in both reflection and transmission regimes, for short pulses interacting with the multi-layer graphene metamaterial.

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