Terahertz relativistic spatial solitons in doped graphene metamaterials

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Abstract
We propose an electrically tunable graphene-based metamaterial that shows a large nonlinear optical response at THz frequencies. The responsible nonlinearity comes from the intraband current, which we are able to calculate analytically. We demonstrate that the proposed metamaterial supports stable 2D spatial solitary waves. Our theoretical approach is not restricted to graphene, but can be applied to all materials exhibiting a conical dispersion supporting massless Dirac fermions.

(Some figures may appear in colour only in the online journal)

1. Introduction
Graphene is a two-dimensional (2D), one-atom thick allotrope of carbon that has been in the spotlight since its experimental discovery and isolation in 2004 [1]. One of the most interesting aspects of graphene, making this material a unifying link between low-energy condensed matter physics and high-energy quantum field theory, is that its 2D quasi-electrons behave like massless (and thus in a certain sense 'relativistic') Dirac fermions, very similar to electrically charged 'neutrinos' [2, 3]. Graphene holds the promise for building advanced nano-electronic devices, due to its unconventional electronic properties [3]. Furthermore, graphene also exhibits very unique optical properties, especially at the terahertz (THz) frequency range. To date, novel photonic devices such as THz devices [4], optical modulators [5], photodetectors [6] and polarizers [7] have been successfully realized. Thus, many researchers now believe that graphene can play a key role and bring us more surprises in photonics and opto-electronics.

In recent years, it has become clear that the potential of graphene structures for nonlinear optical applications is huge, yet largely unexplored. It has been demonstrated that graphene exhibits an extremely strong nonlinear optical response in the THz regime [8, 9]. Preliminary experimental results in this direction include ultrafast saturable absorption [10, 11] and the observation of strong four-wave mixing [12], both of which are very basic phenomena in nonlinear optics [13]. In particular, the existence of four-wave mixing is an indication of the possible existence of modulational instability and optical solitons in graphene, a significant topic that has not been previously investigated.

In this paper, we follow the footsteps of a series of seminal papers by Mikhailov [14] based on the semiclassical kinetic theory, which we use to derive analytically the intraband optical current of a doped layer of graphene. We prove that this theory is also consistent with the more precise quantum approach of Ishikawa [9], based on the Bloch equations derived from the single-electron Dirac equation. In the limit of excitation frequencies in the THz gap, and when neglecting the interband transitions (which is a good approximation for photons below the Fermi energy of doped layers), the two approaches give the same result for the calculation of the intraband current. We apply the above result to describe the self-focusing of 2D spatial solitons in an electrically tunable metamaterial made of several layers of doped graphene, interspaced by layers of silica and silicon, the thickness of which is much smaller than the wavelength of the incident light. This allows us to effectively use an average-medium approach to the description of the propagation and evolution of light. Previous theoretical investigations on the spacetime dispersion of graphene conductivity and quantum transport in graphene also focused on the calculation of the intraband current [15, 16].
A transferred to the electron by the radiation field, in the collisionless approximation is the solution of the Boltzmann–Vlasov kinetic equation:

\[ \mathbf{J} = -\frac{e}{\sqrt{2\pi}} \mathbf{f}(\mathbf{p}) \int \frac{d\mathbf{p}}{p} \mathbf{F}[\mathbf{p} - \mathbf{p}_0(t)] \mathbf{p}, \]

where \( \mathbf{g}_0 = 2 \) and \( \mathbf{g}_e = 2 \) are respectively the spin and valley degeneracy factors. This expression gives the intraband current that is responsible for the strong nonlinearity of graphene in the THz range of frequencies, as demonstrated by Mikhailov \[12, 14\].

More recently, Ishikawa \[9\] introduced Bloch-like equations directly deduced from the one-electron Dirac equation. In his formalism, he starts from the Weyl equation for the charged neutrino, \( i\mathbf{\sigma} \psi = 0 \), where \( \mathbf{\sigma} = (\mathbf{\sigma}_1, \mathbf{\sigma}_2, \mathbf{\sigma}_3) \) is the pseudo-relativistic derivative, \( \psi = (\psi_L, \psi_R) \) is the Pauli matrices vector and \( \mathbf{\sigma}^T = \mathbf{\sigma} \) is the 2 \times 2 identity matrix. Expanding into space and time variables, this translates into the wave equation for the electronic spinor with momentum \( \mathbf{p} \), which reads \( i\hbar \partial_t \psi = \mathbf{v}_F(\mathbf{p}) \psi \equiv \mathbf{H}_0 \psi \), with the momentum operator \( \mathbf{p} \equiv -i\hbar \nabla \). The interacting theory is directly implemented via the minimal substitution: \( i\hbar \partial_t \psi = \mathbf{v}_F(\mathbf{p}) + \exp(\mathbf{A}(t)) \psi = \mathbf{H}_0 \psi \).

If one would be able to solve equation (3), then one can construct the single-electron current \( \mathbf{J}_{1,p} = -e\mathbf{v}_F \mathbf{F}[\psi^\dagger \mathbf{\sigma} \psi] \mathbf{p} \) for a given electronic momentum \( \mathbf{p} \). In his important contribution, Ishikawa \[9\] has demonstrated that in the case of intraband transitions only \( n = -1 \) and \( \rho = 0 \) in the notation of \[9\]), the single-electron intraband current calculated through the Weyl equation would reduce to

\[ \mathbf{J}_{1,p} = -e\mathbf{v}_F \sqrt{(\mathbf{p}_x + e\mathbf{A})^2 + \mathbf{p}_y^2} \mathbf{p} (\mathbf{p}_x + e\mathbf{A}) \mathbf{p}_y, \]

and then the total intraband current is given by \( \mathbf{J} = \int \mathbf{J}_{1,p} \mathbf{F}[\mathbf{p}_x, \mathbf{p}_y] d\mathbf{p} \). This simple result perfectly agrees with the Boltzmann–Vlasov equation approach given above. It is also important to note that, as indicated in \[14\], the intraband current dominates the interband current for photon energies \( h\omega \lesssim \epsilon_F \) and for \( k_B T \ll \epsilon_F \).

In this paper, we calculate analytically the total macroscopic intraband current of a single graphene layer of thickness \( d \) at low temperature (\( T \rightarrow 0 \)). The final result for the \( x \) component (i.e. the only non-vanishing component of the 2D current) is given by

\[ J_{2D}(A) = -\frac{e^2 g_0^2 g_e}{(2\pi)^2} \left( \frac{1/2}{(2\pi)^2} + \frac{eA}{3\epsilon} \right) \left( \frac{4eA}{(p_F + eA)^2} \right) \left( \frac{eA}{(p_F + eA)^2} \right) \times \left( \frac{4eA}{(p_F + eA)^2} \right), \]

where \( \mathbf{F}(\mathbf{p}) \) is the force exerted by the electric field \( \mathbf{E}(\mathbf{r}, t) \) on the electron (\( e > 0 \) here and in the following).

Due to Jeans’ theorem \[18\], which states that any function of the constants of motion of a particle is a solution of the Boltzmann–Vlasov equation, and assuming for simplicity that the system is homogeneous on the \( x, y \) plane (i.e. \( \nabla \mathbf{f}_0 = 0 \)), an exact solution of equation (1) is given by the Fermi–Dirac distribution at temperature \( T \) for interband transitions being negligible, namely \( \mathbf{f}_0(\mathbf{p}) = \mathcal{F}[\mathbf{p}_x - \mathbf{p}_0(\mathbf{r}, t), \mathbf{p}_y - \mathbf{p}_0(\mathbf{r}, t)] \), where

\[ \mathcal{F}[\mathbf{p}_x, \mathbf{p}_y] = \left[ 1 + \exp(\epsilon + \epsilon_F) / (k_B T) \right]^{-1}, \]

where \( k_B \) is the Boltzmann constant, \( \mathbf{p}_0(\mathbf{r}, t) \equiv -e \mathbf{A}(t) \) is the momentum transferred to the electron by the radiation field, \( \mathbf{A}(t) = -\int \mathbf{E}(t) d\mathbf{r} \) is the vector potential and \( \epsilon_F \) corresponds to an electron surface density \( n_e = \epsilon_F / (\hbar^2 v_F^2) \).

2. Background

Let us consider an electrically doped graphene system in which a positive gate voltage \( V_g \) is applied. The energy dispersion of electrons in the conduction band of graphene is given by the well-known Dirac spectrum, \( \epsilon_p = \mathbf{v}_F p \), where \( p \equiv |\mathbf{p}| = \sqrt{p_x^2 + p_y^2} \) is the total electronic momentum, \( \mathbf{p} \equiv (p_x, p_y) \), and \( v_F \approx c/300 \) is the Fermi velocity with \( c \) being the speed of light in vacuum. In figure 1(a), the above spectrum is schematically shown, together with the interband and intraband optical transitions. The Fermi energy \( \epsilon_F \) can be controlled very effectively by changing \( V_g \) applied perpendicularly to the graphene–SiO\(_2\)–Si multilayer, see also figure 1(b). The velocity operator for the quasi-electrons is simply given by \( \mathbf{v} \equiv \nabla \mathbf{f}_0 = \mathbf{v}_F \mathbf{p} / p \). It is important to note that the Dirac point of graphene dispersion is not destroyed by introducing the SiO\(_2\) substrate \[17\].

The momentum distribution function \( f_0(\mathbf{p}, t) \) for electrons in the collisionless approximation is the solution of the Boltzmann–Vlasov kinetic equation:

\[ \partial_t f_0(\mathbf{p}, t) + \mathbf{v} \cdot \nabla f_0(\mathbf{p}, t) + \mathbf{F} \cdot \nabla_p f_0(\mathbf{p}, t) = 0, \]

where \( \mathbf{r} \) and \( t \) are respectively the space and time coordinates, and \( \mathbf{F}(\mathbf{r}, t) \) is the force exerted by the electric field \( \mathbf{E}(\mathbf{r}, t) \) on the electron (\( e > 0 \) here and in the following).

3. Calculation of total intraband current

The total electric current of a collection of electrons that satisfy the Fermi–Dirac statistics is given by

\[ \mathbf{J} = -\frac{e}{\sqrt{2\pi}} \mathbf{f}(\mathbf{p}) \int \frac{d\mathbf{p}}{p} \mathbf{F}[\mathbf{p} - \mathbf{p}_0(t)] \mathbf{p}, \]

where \( g_0 = 2 \) and \( g_e = 2 \) are respectively the spin and valley degeneracy factors. This expression gives the intraband current that is responsible for the strong nonlinearity of graphene in the THz range of frequencies, as demonstrated by Mikhailov \[12, 14\].

Figure 1. (a) Schematics of graphene conical dispersion with doping. Intraband and interband optical transitions are indicated. (b) Geometry of the proposed multilayer metamaterial. The structure is made up of alternating graphene–silica–silicon layers, with total thickness \( L \), much smaller than the wavelength of the THz beam. Each layer of graphene is doped by using an applied gate voltage \( V_g \).
Figure 2. (a) Plot of the analytical intraband current $j_{2D}(\psi)$ (blue solid line), its hyperbolic tangent approximation (black dash–dotted line) and its relativistic approximation (red dashed line). (b)–(d) Plots of analytical intraband current $j_{2D}(t)$ when $\psi(t) = \psi_0 \mathrm{sech}(t/t_0) \cos(5t/t_0)$ (blue solid line), for $\psi_0 = 0.2, 1$ and $3$ respectively, corresponding to the three black dots in (a). The same curves but using the tanh and the relativistic approximations are also shown by the black dash–dotted and the red dashed lines, respectively.

where we have defined the elliptic integrals $E_+(x) \equiv E_+(\tfrac{1}{2}x)$, with $E_+(\theta|x) \equiv \int_{0}^{\infty} \left(1 - x \sin^2 \theta \right)^{-\frac{1}{2}} \, d\theta$.

Let us now define the dimensionless variable $\psi \equiv \epsilon A/\nu_F$, where $\nu_F \equiv \epsilon_F/\nu_F$ is the Fermi momentum, and electric monochromatic fields are scaled with the reference field $E_0 \equiv \omega_0 \epsilon_F/(\nu_F \epsilon_F)$. Figure 4(a) shows the dimensionless quantity $j_{2D}(\psi) \equiv J_{2D}/Jr$ (blue solid line) (where $J_F \equiv -\epsilon_F p_F^2/(\pi \hbar^2)$ is the elementary Fermi current) which in terms of the $\psi$ variable is given by

$$j_{2D}(\psi) = \frac{2}{3\sqrt{\psi}} \left[ 1 + \psi \right] \left( 1 + \psi^2 \right) E_+ \left( \frac{4\psi}{1 + \psi^2} \right) - \left( \psi - 1 \right)^2 E_- \left( \frac{4\psi}{1 + \psi^2} \right),$$

(6)

showing the strong intrinsic nonlinear behaviour of graphene layers when excited by THz radiation. Interestingly, $J_{2D}(\psi)$ can be roughly approximated in several ways, depending on the application one is interested in. For instance, a hyperbolic tangent function approximation, $j_{2D}(\psi) \approx \tanh(\psi)$, is useful for several estimates, for example in the calculation of the linear intraband conductivity, giving the correct $\sigma_{\text{inta}} = e^2 \nu_F^2/(\pi \hbar^2 \omega)$ with $\omega$ being the radiation frequency. We note that the latter function has exactly the same asymptotic behaviour of $J_{2D}$ for $\psi \to 0$ and $\psi \to \infty$. However, the approximation might fail for more precise estimates around the most nonlinear region, namely $\psi = 1$. Here, however, we prefer to use the less precise but more tractable approximation $j_{2D}(\psi) \approx \psi/\sqrt{1 + \psi^2}$, which also shows the pseudo-relativistic nature of the optical nonlinearity treated here. This expression will allow us to treat the transition from real fields to envelopes in a straightforward way in the framework of the paraxial approximation, since the Taylor expansion of equation (6) does not work well due to its fictitious singularity at $\psi = 0$.

In figure 2(a), the full function $j_{2D}(\psi)$ and its two approximated versions are shown for comparison.

In figures 2(b)–(d), we show the analytically calculated current $j_{2D}(t)$ for an example of pulsed excitation $\psi(t) = \psi_0 \mathrm{sech}(t/t_0) \cos(5t/t_0)$, where $t_0$ is the pulse width, for different values of the light field amplitude $\psi_0$, showing the strong nonlinear temporal dependence of the intraband current. Curves obtained by using the tanh and the relativistic approximations above are also shown for comparison.

A quick estimate of the third-order susceptibility $\chi^{(3)}$ is given by expanding $J_{2D}$ in powers of $\psi$ up to the third order: $J_{2D} \approx \psi - \psi^3/8$, giving the nonlinear third-order intraband current $J_{2D}^{(3)}(E) = [eE/(2\omega \nu_F)]^3 [e\nu_F p_F^2/(\pi \hbar^2)] = \omega \epsilon_F \chi^{(3)}(E^3)$. The order of magnitude of such susceptibility for a monochromatic wave is thus given by $\chi^{(3)} = e^2 v_F^2/(8\pi \epsilon_F \hbar^2 \omega^4 \epsilon_F d) \approx 10^{-5} - 10^{-4} \chi_{\text{silicon}}$, depending on the specific parameters used. Note that $\chi^{(3)} \sim \omega^{-4}$, so the intraband nonlinearity rapidly decreases when increasing the frequency. This is consistent with the estimates given in the works by Mikhailov [12, 14]. One must always keep in mind that such estimates, although relevant to get a feeling for the orders of magnitude involved, do not capture the full complexity of the nonlinearity, which is not just a simple Kerr nonlinearity. However, such large third-order coefficient places the graphene nonlinearity in the same category of the resonant nonlinear effects, such as two-level systems [19] or the excitonic nonlinearity [20], but with the great advantage that the bandstructure of graphene is always resonant to optical excitations.

4. Optical propagation

Wave equation for a single graphene layer. The equation that regulates light propagation in the presence of a single graphene layer is the conventional macroscopic wave equation $c^2 \nabla A = J_{2D}(A)$, where $\nabla \equiv (\epsilon_\omega/\epsilon^2) \nabla^2 - \nabla^2$, with $\epsilon_\omega$ is the substrate dielectric function at the selected frequency $\omega$, and $A(r,t)$ is the 3D vector potential. The current circulates in a very thin layer of thickness $d \approx 0.34 \text{ nm}$. Thus, we can model the 3D current with a rectangular function, with the single layer centred at $z = 0$: $J_{2D}(r,t) = J_{2D}(x,y,t) R(z)/d$, where $R(z) \equiv [\sqrt{\sin(z + d/2)} + \sqrt{\sin(d/2 - z)/2}]$, where $\sin(x)$ is the sign function, normalized like $\int_{-\infty}^{\infty} R(z) \, dz = d$.

Average–medium theory of graphene metamaterial. In order to observe THz spatial solitons, we consider a doped graphene metamaterial as shown in figure 1(b). The system that we propose is a periodic multilayer based on graphene–silicon–silicon layers, with total thickness $L$ (see figure 1(b)), where SiO$_2$ and Si are both transparent for THz and optical frequencies. Each graphene layer is doped with an electronic density $n_e$ by applying a gate voltage $V_g$. The size $L$ of the elementary cell is assumed to be much smaller than the incident monochromatic wavelength, $L \ll \lambda$. This means that we can use an average medium approach by expanding the dimensionless vector potential in its Fourier components $\Phi$:

$$\psi(x,y,z,t) = \frac{1}{2} \sum_{m,n} \phi(x,y,z) e^{2\pi i m x/L + 2\pi i n z/d + c.c.}.$$

(7)

In figure 2(a), the full function $j_{2D}(\psi)$ and its two approximated versions are shown for comparison.
where \( k_0 \) is the linear wavenumber, and \( z \) is the direction perpendicular to the layers (see figure 1(b)).

By retaining only the fundamental order in the Fourier expansion, and after using the paraxial approximation for reducing the order of the \( z \) derivative, one obtains

\[
\left\{ \begin{array}{l}
\frac{\varepsilon_i\omega^2}{c^2} - k_0^2 + i\eta^2 + 2i\lambda \frac{\eta}{r}
\end{array} \right\} \frac{\partial \phi}{\partial X} + \left[ -\frac{\varepsilon_i\lambda}{\pi \epsilon_0 \varepsilon_r \lambda^2} \right] f_{2D}(\phi) c_0 = 0,
\]

where \( \sum_{n} c_n \epsilon_2 \pi i c_n e^{2\pi i n/L} e^{2\pi i n X/L} = \sum_{n} c_n \epsilon_2 (e^{2\pi i n/L} e^{2\pi i n X/L})^2, c_0 = \frac{1}{\lambda} \int_{-L/2}^{L/2} R(z) e^{-2\pi i n L} dz, c_0 = \frac{1}{\lambda} \int_{-L/2}^{L/2} R(z) dz = d/L. \]

We now use the relativistic approximation of the full current, namely \( j_{2D}(\phi) \equiv \lambda^2 (1 + |\phi|^2/2)^{1/2} \), inside equation (8).

Paraxial model and soliton solutions. After introducing the rescalings \((x, y) = x_0(X, Y), z = \eta Z, x_0 = \pi h^2 c^2 e_0 L/(e_0 e_f)^{1/2}, \eta = \phi/\lambda^2 \), one obtains the paraxial equation:

\[
i \frac{\partial^2 \eta}{\partial X^2} + \frac{\partial^2 \eta}{\partial Y^2} - \frac{\eta}{\lambda^2 + |\eta|^2} = 0,
\]

which has been previously studied in several contexts [23–25]. For typical parameters \( \omega/(2\pi) = 20 \text{ THz}, \epsilon_i \simeq 4.5 \) [26], \( L \simeq 2 \mu \text{m} \) and \( n_e \simeq 5 \times 10^{12} \text{ cm}^{-2} \), one has \( \epsilon_f \simeq 259 \text{ meV}, x_0 \simeq 15 \mu \text{m} \) and \( \eta_0 \simeq 415 \mu \text{m} \). The scaling for the electric field is \( E_0 \equiv \omega \epsilon_0 \epsilon_f/(\eta_0 e_f) \simeq 30 \text{ MW cm}^{-2} \). For the above parameters, the room temperature is a good approximation of the above results obtained for \( T = 0 \), since \( k_0 T \simeq 25 \text{ meV} \ll \epsilon_f \).

Passing to cylindrical coordinates, one must solve the following ODE:

\[
\frac{d^2 \eta(R)}{dR^2} + \frac{1}{R} \frac{d \eta(R)}{dR} = \left[ g + \frac{1}{\sqrt{1 + \eta(R)^2}} \right] \eta(R) = 0,
\]

where \( g \) is a nonlinear wavenumber, and \( R \equiv \sqrt{X^2 + Y^2} \) is the dimensionless radius, in units of \( x_0 \). Solutions of equation (10) are 2D spatial solitons [27–30] with a rather unconventional relativistic (and thus saturable) nonlinearity. The fundamental and some of the higher order soliton profiles are shown in figure 3 for different values of \( q \). The fundamental soliton shown in figure 3(c) is stable, since the saturation of the nonlinearity gives it stability (which can also be confirmed by the Vakhitov–Kolokolov criterion), while the higher order solitons (which exhibit rings) are typically unstable and will decay into multiple fundamental solitons [31].

5. Conclusions

In this paper, we have proposed an electrically tunable metamaterial based on graphene–silicon–silica multilayers. We have calculated the intraband current of doped graphene analytically, which dominates the electron dynamics for THz excitations. Finally, stable 2D spatial solitary waves have been found to propagate in the longitudinal direction for realistic parameters. This will pave the way for a more extensive analysis of the mathematical structure and the physical content of the graphene Bloch equations for useful nonlinear optical applications. Our theoretical approach is not restricted to graphene, but can be applied to all materials exhibiting a conical dispersion, supporting massless Dirac fermions (for instance, HgTe [32]).

It is important to note that in the above analysis, we have neglected losses in the calculation of the current. It has been demonstrated by Mikhailov [33] that this is acceptable when the number of graphene layers is between 25 and 35. In that case, the nonlinear effects are maximized and the losses are optimized, and the approximations used above are reasonable.

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