Carbon – Science and Technology

Plane-Wave Excitation and Electromagnetic Response of Graphene : Non-Linear Effects

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In graphene, the two-dimensional electrons and holes are described by the Dirac equation with a vanishing effective mass. As a consequence, under particular operative conditions, the electromagnetic response of graphene can be strongly nonlinear. We recently developed a method that accounts for deterministic electromagnetic field dynamics together with the quantum coherent transport in the nanoscale environment. We analyze the response of the system to harmonic plane wave and observe the frequency multiplication effect, resulting from the nonlinearity of the electromagnetic response. Theoretical results are compared with ones reported in the literature.

Introduction :

In view to the new epochal scenarios that nanotechnology discloses, a multitude of exciting research projects based on novel materials and nano-science concepts have been developed to pave the way for a new generation of nanoelectronic devices and systems, yielding not only higher integration densities as well as substantially improved electro-thermal-mechanical properties, but also unprecedented possibilities for novel or drastically improved devices/systems in the microwave / RF engineering domain of applications [1 - 2]. Many nano-scale materials and devices, e.g. carbon-based materials exhibit their most interesting properties over a broad range of applications and operating frequencies, covering the radiofrequency (RF) spectrum, through the microwave up to the optical region.

Among these novel materials, graphene, made of carbon atoms packed in a 2D-honeycomb lattice, is quickly becoming extremely interesting solutions for a wide variety of electronic devices and circuits. It offers the possibility of outstanding performances with much lower power draw, using processing technology compatible to that used in advanced silicon device fabrication (CMOS).

Our analysis is focused on graphene nanoribbons (GNR), that is, a narrow strips of graphene.

In the frequency (energy)-domain, the analysis of graphene and/or graphene nanoribbons (GNR), narrow strips of graphene GNR, can be carried out by discrete models such as tight binding (TB), and continuous models, such as effective mass and $k \cdot p$ approximations, which stems from the approximation of TB around particular points of the dispersion curves.

The $k \cdot p$ model leads, for the graphene/GNR, to a Dirac-like system of equations [3]. Another popular approach for the calculation of quantum transport is given by the use of the Green's function of a GNR region [4]. These techniques are demonstrated to be quite accurate for the analysis of GNR in a variety of problems such as, for example, the effects on charge transport of applied external electric and magnetic fields,

bending, lattice defects and discontinuities, edge terminations and so on.

However, the latter methods requires high computational resources, and can hardly include the effect of i) the self-generated electromagnetic (EM) field, ii) impinging external EM fields. Recently, we have introduced full-wave techniques both in the frequency (energy)-domain [5 - 9], and the time-domain [10 - 13] for the investigation of new devices based on carbon materials, namely carbon nanotube (CNT), multiwall (MW) CNT, graphene, graphene nanoribbon (GNR).

In both the approaches, the quantum transport is described by the Schrödinger equation or its Dirac-like counterpart. The electromagnetic field provides sources terms for the quantum transport equations, that, in turns, provides charges and currents for the electromagnetic field.

In particular, the time-domain full-wave technique introduced in [10 - 13] is aimed for the investigation of the deterministic EM field dynamics together with the quantum phenomena. This is particularly important in the analysis of time transients and in the describing the behavior of high energy carrier bands, as well as the onset of non-linear phenomena due to external impinging electromagnetic fields.

In this contribution, we analyze the electromagnetic response of graphene to an harmonic impinging plane wave. The frequency multiplication effect, resulting from the nonlinearity of the electromagnetic response, is studied under realistic experimental conditions. The first theoretical results are compared to ones predicted in the literature, showing good agreement. This

The combined Maxwell-Dirac equations solution :

In the full-wave time-domain technique introduced in [10 - 13], for each time-step, the Maxwell equations are self-consistently solved together with the Dirac equation, describing the quantum mechanical massless fermions dynamics in ballistic regime. In the Maxwell equations

$$\nabla \times \mathbf{E}(\mathbf{r},t) = -\frac{\partial}{\partial t}\mathbf{B}(\mathbf{r},t) - \mathbf{M}_{eq}(\mathbf{r},t)$$

$$\nabla \times \mathbf{H}(\mathbf{r},t) = \frac{\partial}{\partial t}\mathbf{D}(\mathbf{r},t) + \mathbf{J}(\mathbf{r},t)$$

(1)

the $\mathbf{J}$ electric current source include the quantum-mechanical contribution, and $\mathbf{M}$ equivalent magnetic sources account of external electric field (e.g. plane wave). The Dirac equation, in the presence of an EM field, is defined as

$$i\mathrm{h}\left(\frac{\partial}{\partial t} + \frac{ie}{\mathrm{h}}\varphi\right)\psi_+ = \boldsymbol{\sigma}\cdot\left(\hat{\mathbf{p}} - q\mathbf{A}\right)c\psi_-$$

$$i\mathrm{h}\left(\frac{\partial}{\partial t} + \frac{ie}{\mathrm{h}}\varphi\right)\psi_- = \boldsymbol{\sigma}\cdot\left(\hat{\mathbf{p}} - q\mathbf{A}\right)c\psi_+$$

(2)

In equation (2), $\mathbf{A}$ and ▯ are vector and scalar potentials, directly related to the electromagnetic (EM) field

$$\mathbf{E} = -\nabla\phi - \frac{\partial\mathbf{A}}{\partial t} \qquad \mathbf{B} = \nabla\times\mathbf{A}$$

(3)

through Lorentz gauge :

$$\nabla\cdot\mathbf{A} + \frac{1}{c}\frac{\partial\phi}{\partial t} = 0$$

(4)

In equation (2), $\boldsymbol{\sigma}$ are the Pauli matrices and $\mathbf{p}$ is the linear momentum The solution of the Dirac/graphene equation (1) is the four component spinor complex wavefunction ▯(**r**,t) :

$$\boldsymbol{\psi}(\mathbf{r},t) = \left[\psi_1\ \psi_2\ \psi_3\ \psi_4\right]^T = \left[\psi_+\ \psi_-\right]^T$$

(5)

In particular, in the 2D graphene, equation (2) behaves :

$$H(\mathbf{r},t)\boldsymbol{\psi}(\mathbf{r},t) = i\mathrm{h}\left(\frac{\partial}{\partial t} + \frac{ie}{\mathrm{h}}\varphi(\mathbf{r},t)\right)\boldsymbol{\psi}(\mathbf{r},t)$$

(6)

Where the Dirac Hamiltonian is defined as :

$$H(\mathbf{r},t) = v_F\begin{pmatrix} \sigma_x k_x(\mathbf{r},t) + \sigma_y k_y(\mathbf{r},t) & 0 \\ 0 & -\sigma_x k_x(\mathbf{r},t) + \sigma_y k_y(\mathbf{r},t) \end{pmatrix} + V_p(\mathbf{r})$$

(7)

In equation (6), σ_x and σ_y are two of the three Pauli matrices, V_p is the static potential barrier, $i=-j$ is the imaginary unit, v_F is the Fermi velocity, $\mathbf{r}=(r_x, r_y)$ is the in-plane position vector, $k_x(\mathbf{r},t)$ and $k_y(\mathbf{r},t)$ are the components of the kinematic momentum $\mathbf{k}(\mathbf{r},t)$, related to the linear momentum $\mathbf{p}$:

$$\hat{\mathbf{p}}=-i\hbar\nabla \qquad \hat{\mathbf{k}}=\hat{\mathbf{p}}-q\mathbf{A}(\mathbf{r},t) \qquad (8)$$

The basic requirement for the solutions is the following normalization condition, stating that the probability of the particle being somewhere is one, and also corresponding to the charge conservation.

$$\forall t \geq 0 \quad \int_{-\infty}^{+\infty}\|\psi(\mathbf{r},t)\|^2 d\mathbf{r}=1 \Leftrightarrow q\int_{-\infty}^{+\infty}\|\psi(\mathbf{r},t)\|^2 d\mathbf{r}=Q_T \quad (9)$$

From the wave solution $\psi(\mathbf{r},t)$, it is straightforward to derive the correspondent QM [10 - 12]. The computational scheme develops as in follows :

i) The Maxwell equations are discretized by using the Transmission Line Matrix (TLM) method [11 - 13]. The TLM method, like the finite-difference time-domain (FDTD) method, is a well-known technique that allows the EM full-wave modeling of 3D structures with nearly arbitrary geometry for a wide range of applications from EM compatibility to optics. FDTD is a more general technique, suited for discretizing different kind of equations, e.g. parabolic, hyperbolic etc. Respect to FDTD, TLM is directly related to the discretization of, mainly, hyperbolic equations (Maxwell, Dirac), but it has the advantages that each portion of the segmented space has an equivalent local electric circuit [13]. The TLM scheme is considered as the implementation of the Huygens principle : propagation and scattering of the wave amplitudes are expressed by operator equations. Wave pulses are scattered by nodes (source points), propagate in the conformal (e.g. Cartesian) mesh, and recombine to form new wave front; at each space-time point of the mesh, the combination of incident / scattered pulses gives the EM field components. In this way, the physical correspondence among EM field components and wave pulses is established.

ii) Quantum phenomena are introduced in 2D grapheme / nanoribbon region, and described by the Dirac equation. iii) At each time step, the Dirac equation is solved by accounting for the quantum device boundary conditions, initial conditions (e.g. injected charge); and additional source terms constituted by the EM field, sampled in the domain of the quantum device(s). iv) From the wavefunction (charge) solution of the Dirac equation, we derive the quantum mechanical (QM) current over the device domain. This current is a distribution of local sources for the EM field, that is injected into the TLM nodes, just located on the grid points of the Dirac equation domain. v) At the next time step t+1, the TLM method provide a new updated distribution of field values that are, again, sampled over the device domain, and so on, iteratively.

iv) The contribution of external EM field sources, be they impressed and/or dynamic, are accounted in the Maxwell equations (1).

Results :

In order to perform a comparison with theoretical prediction in the literature, we model and reproduce the configuration reported in [14]. The situation is depicted in Figure (1). An external EM plane wave with amplitude E_0 and frequency ω_0 is impinging on the graphene layer. This EM field onsets the dynamics of graphene carriers, that, in turns, originate a self-induced EM near field. Now, once the surface carrier concentration n_s is fixed, the point is to predict the field amplitude E'_0, as the limit between linear and non-linear regimes. Below E'_0, the EM self-induced field has the same ω_0 frequency of the impinging field; above E'_0, the EM self-induced field shows significant peaks at $2\omega_0, 4\omega_0,\ldots$

In [14], the analytically predicted condition for achieving the non-linear regime of the EM response is

$$\frac{qE_0 v_F}{\omega_0\sqrt{n_s}} \gg 1 \quad \Rightarrow E_0^{NL} \gg \frac{\omega_0\sqrt{n_s}}{qv_F}$$

$$\begin{cases} f = 100 \ GHz \\ n_s \approx 10^{11} \ cm^{-2} \end{cases} \Rightarrow E_0 > 200 \ V/cm \qquad (10)$$

The irradiation of a graphene layer by electromagnetic wave with the frequency ω_0 leads to the higher harmonics generation with the higher harmonics amplitudes falling down very slowly, as $1/|m|$.

It is quite evident that the amplitude of the electric field E_0 that "onsets" the non-linearity strongly depends on the i) surface carrier concentration n_s, that, in turns, depends on the Vg-electrical "doping" of the graphene respect to an electrode, ii) the operating frequency w_0.

$$E_{ext} = E_0 \cos(\omega_0 t)$$
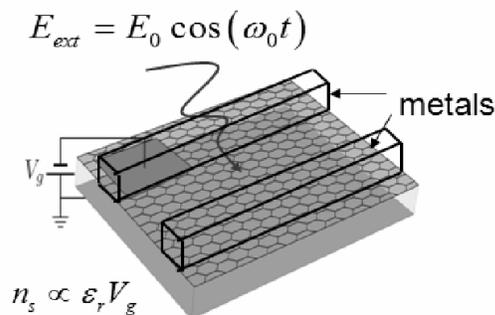
metals

V_g

$$n_s \propto \varepsilon_r V_g$$

Figure (1). The analyzed configuration. A squared (L=100 nm) graphene sheet is placed over a silicon dioxide and electrically tuned for getting n_s. Two metal electrodes are placed over the graphene layer. An external plane wave is impinging on the structure.
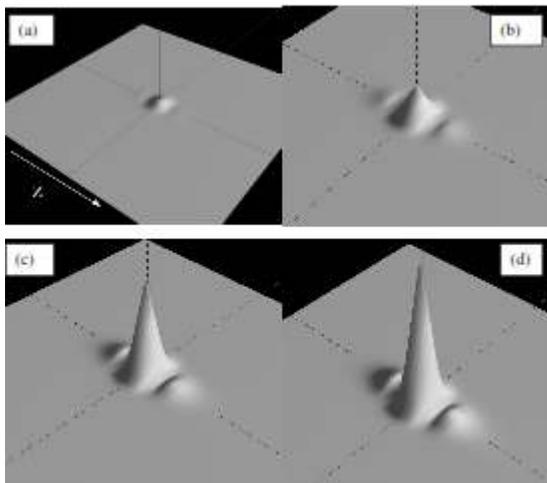

Figure (2). Amplitude of the z-directed field component at t=20 fs (a); t=50 fs (b); t=150 fs (c), t=200 fs (d).

In our simulation, for the same n_s, ω_0 parameters, we found the non-linear condition with $E_0 > 400$ V/cm, that is good results at all. In fact, we consider a more realistic situation, with the presence of two additional electrodes,

representing some kind of circuitry in fabricated environment. In Figure (2), we report the amplitude of the z-directed field component, with E_0=420 V/cm, at t=20 fs (a); t=50 fs (b); t=150 fs (c), t=200 fs (d). It is interesting to observe the presence of metal electrodes (vanishing of the field component) and the behavior of the peak field, that begins to diverge, while resonating between the metal plates.

Conclusions :

We report on the investigation on the non-linear electromagnetic response of graphene taking into account the self-consistent-field effects. Response of the system to a strong pulse excitation is calculated. Numerical results from the present technique are compared to theoretical analysis present in the literature, showing a good agreement. The possibility to use the graphene as natural frequency multiplier paves the way to a new generation of devices, like RFID tag sensors, as discussed in [15].

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