Invited Paper

The linear and non-linear THz properties of graphene

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Abstract: Graphene has recently been the subject of much attraction for THz applications due to its extraordinary material properties arising from its particular band structure. This paper reviews and discusses the linear and nonlinear properties of graphene at microwave/THz frequencies, and it also compares these properties with those of other semiconducting materials. By taking into account the effect of electron momentum relaxation time on intra-band optical transitions, which is significant at microwave and terahertz frequencies, we derive an analytical form for the third order microwave/THz susceptibility in graphene which is found to be $>10^3$ *times* larger than that of other typical nonlinear materials. Finally, current and future applications of such properties in THz devices are discussed, in particular, and we discuss a novel application of graphene into active THz filters.

Keywords: Graphene, Nonlinearities, Charge transport properties, Active devices, Filters

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1. Introduction

With promising applications in many diverse areas of human endeavor, including medicine, biology, communications, security, astronomy, and so on, terahertz (THz) technology has recently turned into a very active area of scientific research [1, 2]. The THz frequency band, usually defined in the 0.1-30 *THz* range, was for decades one of the least explored regions of the electromagnetic spectrum, mainly due to the lack of materials and devices responding to these frequencies in a controllable manner. Even today, there exists a need for devices that can efficiently manipulate THz waves. In this context, graphene has recently arisen as an important material for THz devices due to its extraordinary THz properties inherited from its particular band structure [3]. Under low THz fields graphene exhibits a linear response, where its optical conductivity follows a Drude-like behavior; at these frequencies the optical conductivity heavily depends on graphene Fermi level thus can be modified by means of field effect. Based on this phenomenon many active THz devices have been recently proposed. Under high THz fields, and normal THz-light incidence, graphene exhibits a strong third-order non-linear response which can be orders of magnitude larger than what is achievable in other frequency ranges such as the IR or the visible.

As a result of its ease of integration, low cost, and extraordinary electrical/optical properties, graphene has been the subject of much attention for electronic and photonic applications [4]. Graphene is an intrinsically two-dimensional (2D) semiconductor, which is comprised of sp^2-bonded carbon atoms in a hexagonal lattice. A sketch of graphene's lattice in real space is shown in Fig. 1, where a unit cell, primitive lattice vectors, and vectors between nearest neighbor atoms are highlighted. In the reciprocal space, graphene is also described by a hexagonal lattice. Near the edges of the first Brillouin zone, its tight binding Hamiltonian can be linearized as:

$$H = \hbar v_F \begin{pmatrix} 0 & k_x + ik_y \\ k_x - ik_y & 0 \end{pmatrix}, \tag{1}$$

where $v_F \approx 10^6 m/s$ is the Fermi velocity, and $\hbar$ is the reduced Planck constant. The details of this derivation of Eqn. (1) can be found for instance in Ref. [5]. From the linearized Hamiltonian the following energy dispersion can be obtained for charge carriers in graphene (i.e. electrons and holes):

$$E(k) = \hbar v_F |k| \ . \tag{2}$$

Because of its linear *E-k* dispersion (which resembles that of massless particles such as photons), and since its Hamiltonian has the form of a "Dirac Hamiltonian" for "massless Dirac Fermions" charge carriers in graphene are usually called massless Dirac Fermions. This linear *E-k* dispersion contrasts that of traditional semiconductors, where the dispersion is parabolic. From these properties, in practice, both types of carriers in graphene can achieve very large mobility, exceeding 100,000 $cm^2/V.s$ at room-temperature [6].

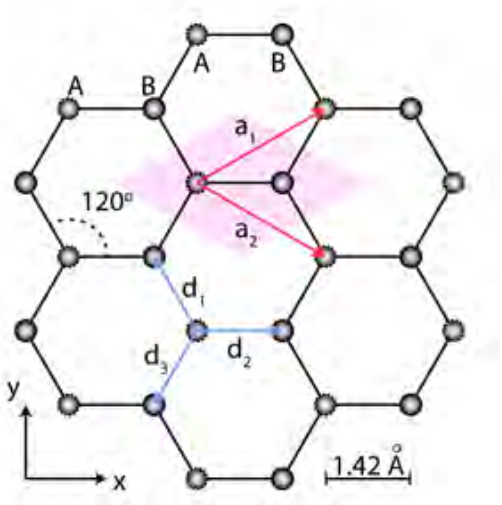

Fig. 1 Sketch of graphene's lattice in real space. A unit cell, comprised of two carbon atoms (A-B) is colored in pink. Primitive lattice vectors and vectors between near atoms are depicted in red and blue respectively. In reciprocal space graphene is also described by a hexagonal lattice. Its band structure at the edges of the first Brillouin zone is linear in accordance with Eqn. (2).

2. Linear response

The optical transitions in graphene include both intra and inter-band transitions. Intra-band transitions require change in *k* thus phonon mediation; thus their properties are closely related to the electrical transport properties of graphene. Based on the linearization of graphene's tight binding Hamiltonian near the edges of the Brillouin zone (Eqn. (2)), its optical conductivity can be expressed as [7-9]:

$$\sigma(\omega) = \sigma_{intra}(\omega) + \sigma_{inter}(\omega) = \frac{ie^2 E_f}{\pi h^2(\omega + i/\tau)} + \frac{ie^2\omega}{\pi}\int_0^\infty \frac{f(\varepsilon - E_f) - f(-\varepsilon - E_F)}{(2\varepsilon)^2 - (h\omega + i\Gamma)^2} d\varepsilon. \quad (3)$$

Here e is electron charge, ω angular frequency, and E_f is Fermi level. $f(\ .\ )$ is the Fermi distribution function, τ momentum relaxation time, and Γ is a parameter describing the broadening of inter-band transitions.

For low photon energies, e.g. the THz range, optical conductivity is determined by the intra-band transition contribution. In contrast, the contribution of inter-band transitions becomes dominant at high photon energies. When ω approaches infinity in Eqn. (3) it can be derived that at high photon energies, the optical conductivity of graphene is $e^2/4\hbar$ thus independent of Fermi level. These photon energies include the visible range, where a universal ~ 2.3 % absorption per graphene layer is observed [10]. As a result of this observation, graphene has been proposed as a platform for transparent-flexible electrodes [11] and UV-transparent electrode for internal photoemission measurements [12].

From Eqn. (3) is it observed that in the IR range optical conductivity can be modified by controlling the Fermi level, but only weakly [13, 14]; nevertheless, this weak dependence is sufficient (in fact superior to what is typically available in conventional semiconductors) for realizing tunable devices such as ultra-fast electro-absorption IR modulators [15, 16]. On the other hand, in the THz range, the contribution from inter-band transitions becomes negligible, thus the real part of optical conductivity (Eqn. (3)) can be rewritten by employing a Drude-like dispersion:

$$Re\{\sigma(\omega)\} = \frac{\sigma_{DC}}{1 + (\omega\tau)^2} \quad , \quad (4)$$

where, unlike in classical semiconductors, σ_{DC} , the DC electrical conductivity of graphene, is a non-linear (square root dependence) function of the charge density (n_s) [17]. The THz optical conductivity, for $f < 1/2\pi\tau$ (where $1/2\ \pi\tau$ depends on the graphene quality and is typically between 1 to 4 *THz*), closely follows the DC electrical conductivity, and thus can be effectively electro-statically tuned [18-21]. As Fermi level becomes closer to Dirac point, the density of states available for intraband transitions decreases thus graphene's optical conductivity. As frequency increases, although optical conductivity decreases, THz absorption still can be

effectively manipulated by employing structures allowing plasmonic resonances as shown by Ju et al. [22], and Yan et al. [23].

3. Nonlinear response

Nonlinear effects are of substantial importance in modern optoelectronics, with applications including ultra-short pulse generation, photo-mixing, ultrafast switching, etc. Nonlinearities arise when the electron motion under an electromagnetic field becomes anharmonic. Since their electric field dependence is super-linear, they are more pronounced under high fields. From this point of view, by means of engineering the material environment to provide field enhancement, nonlinearities can be triggered even with relatively low fields. Such an augmented effective nonlinear optical response can be in practice obtained by employing metamaterials and through plasmonic effects [24, 25]. For instance, coupling of light into surface waves can result in strong local electromagnetic fields, which in turn can significantly enhance the nonlinear optical processes. Local field enhancement at the resonance frequency of metamaterials can also enhance these nonlinearities. This section theoretically provides insight into the nonlinear optical response of graphene at microwave and terahertz frequencies and it briefly discusses the use of metamaterial structures to enable the observation of this phenomenon even at low fields (compatible with table top sources).

Based on its linear properties, graphene has recently become the center of attention for a myriad of terahertz and high frequency applications. Recent studies have predicted very strong optical nonlinearities in graphene, especially at microwave and terahertz frequencies, and observation of nonlinear effects even with rather low fields (~10^3 *V/cm*) [26-30]. This can be understood on the basis of the band structure of graphene. As discussed by Mikhailov in 2007 [26], the nonlinear response of graphene arises from the fact that under an oscillating electromagnetic field the carrier velocity is not proportional to momentum, while in parabolic band semiconductors, velocity (v) and momentum (p) are related by the expression: $v_x = \partial\varepsilon(p)/\partial p_x \propto p_x$, in graphene their relation is given by: $v_x = \partial\varepsilon(p)/\partial p_x \propto p_x/\sqrt{p_x^2 + p_y^2} \propto sgn[p_x]$. Harmonic decomposition of sgn function leads to excitation of all odd high-harmonics (i.e. $3\omega, 5\omega, 7\omega, \ldots$). This natural nonlinear response is of great interest since it can be applied in frequency multiplication and high harmonic generation.

Mikhailov and Zhang's groups theoretically predict a stronger nonlinear response of graphene at these low frequency regions than in the infrared, even to the point that with relatively low fields the nonlinear optical conductance ($\sigma^{(3)}(\omega)$) exceeds the linear one [28-30]. Measurements at microwave frequencies have provided interesting results, with reported conversion efficiencies up to the order of -30 *dB* [31, 32]. These studies showed a flat frequency dependence of conversion efficiency, which contrasts with the expectations from the aforementioned theoretical studies.

In order to accurately model this behavior, one should take into account the effect of electron momentum relaxation time on intra-band optical transitions, which is significant at microwave and terahertz frequencies. The following equation can be derived for the dependence of current with electric field by including this effect:

$$j_x(t) \approx en_s v_F \beta\{(1-3\beta^2/32)\sin(\omega t) + (\beta^2/32)\sin(3\omega t) + \ldots\}, \quad (5)$$

where: $\beta = ev_F\tau E_0/E_F\sqrt{\omega^2\tau^2+1}$. Here E_0 is electric field amplitude, and E_F is Femi level in graphene. It is important to notice that this equation does not asymptotically diverge to infinity when $\omega \rightarrow 0$ as is the case of the equations in the previous theoretical works. Therefore it predicts a frequency independent ratio between $\sigma^{(3)}(\omega)$ and $\sigma^{(1)}(\omega)$ for a fixed electric field at frequencies $f << 1/2\pi\tau$, which is the case in graphene in the microwave and low THz range (e.g. below or around 1 *THz*). The theoretical expressions derived in previous works (e.g. Ref. [26]) are valid either at higher frequencies or for samples with electron densities $n_s < 10^{10}\ cm^{-2}$.

Under low field $\beta << 1$ thus $\sigma^{(3)} << \sigma^{(1)}$; however under high field $\sigma^{(3)}$ and $\sigma^{(1)}$ can become of the same order. At microwave and terahertz frequencies $\omega\tau << 1$ thus $\beta = (ev_F\tau/E_F) \times E_0$. For $10^{12}\ cm^{-2}$ graphene carrier density and 1 *ps* electron momentum relaxation time $\beta \approx 10^{-3} \times E_0[V/cm]$, from where it is estimated that $E_0 > E_c = 2.8kV/cm$ can lead to $\sigma^{(3)}(\omega) > \sigma^{(1)}(\omega)$. The following expression is derived for the third order microwave/THz susceptibility in graphene:

$$\chi^{(3)} = \sigma^{(3)}(\omega)/\omega t_{graphene}\varepsilon_0 = e^4 v_F^4 \tau^3 n_s/32\omega t_{graphene}\varepsilon_0 E_F^2, \quad (6)$$

where $t_{graphene}$ is the thickness of single layer graphene ($0.34nm$) and ε_0 is the vacuum permittivity. For 0.5 *THz*, $\chi^{(3)} \approx 0.5 \times 10^{-7}\ m^2/V^2$, which is 8 orders of magnitude higher than what has been predicted theoretically and measured experimentally in the near-IR (e.g. Ref. [33, 34]). For reference (and comparison) recent studies in liquids (CS2, benzene, CCl4, CHCl3, and CH2I2) showed $\chi^{(3)} \approx 10^{-20}\ m^2/V^2$ [35], $\chi^{(3)} \approx 10^{-17}\ m^2/V^2$ is typical in chalcogenide glasses [36], $\chi^{(3)} \approx 10^{-10}\ m^2/V^2$ was obtained in InAs quantum wells [37], and $\chi^{(3)} \approx 10^{-15}\ to\ 10^{-16}\ m^2/V^2$ for ZnTe, ZnSe, and CdS crystals [38, 39].

Whereas, as previously mentioned, electric fields in the order of ~*kV/cm* are needed for nonlinear effects to be significant, table top THz sources, such as multiplier based and backward wave oscillators, can provide THz powers in the order of >1 *mW* at 500 *GHz* thus electric fields around 10*V/cm*. However, In metamaterials with narrow gaps it is possible to obtain field enhancements in the order of 1,000 X with coupling loss ~ 3 *dB* [40]; therefore sources capable of providing 1 *V/cm* can effectively provide electric fields in the order of 1 *kV/cm* in the metamaterial gap, which is what is required for triggering the nonlinearities in graphene. Thus, use of high power sources might not be a requirement for observing and practically employing this phenomenon.

4. Applications

Applications of graphene into THz devices arise from the possibility of controlling its optical conductivity, and due to its superior carrier transport properties. One of such proposed applications is on THz electro-absorption modulators. In this context, graphene offers several advantages in comparison with other tunable 2DEGs; for instance, graphene devices can achieve a superior depth of modulation, because they exhibit: i) a more favorable dielectric environment; and, ii) a potentially superior conductivity swing in graphene (see Ref. [18]). Experimental demonstration of these devices showed ~ 20 % MD and ~ 0.5 *dB* IL in transmission-mode structures [19]. Although a ~ 20% modulation of THz transmittance may not seem very impressive at a first glance, it is actually remarkable when considering that the active element in this device was only a one atom-thick layer of graphene. However, this performance is still far from the requirements needed for many system applications.

The modulation depth in these early demonstrations of electro-absorption modulators was limited by the conductivity swing achievable in graphene, and therefore by the material quality after chemical vapor deposition (CVD) growth and subsequent transfer process employing poly(methyl-methacrylate) (PMMA) and wet etch methods. However, even with these limited conductivity swings, excellent modulation depth can be achieved by electromagnetically engineering the device structure. One of such ways is via employing reflection-mode structures [41, 42]. In these devices the substrate optical thickness is matched to be an odd multiple of a quarter-wavelength of the incoming THz beam so electric field is enhanced in the active graphene layer. While only a ~ 20 % modulation depth was achieved in the transmittance-based modulator, the modulation depth in the reflection-mode modulator was ~ 70 % for a similar conductivity swing. Based on this enhanced performance, in early 2013 a single-pixel THz imager based on an array of reflection-mode modulators was demonstrated [43].

Tunable THz metamaterials constitute another way to realize THz electro-absorption modulators simultaneously achieving low insertion loss and high depth of modulation (see Ref. [44]). These devices can be constructed by employing passive metallic frequency selective surface (FSS) in conjunction with active graphene layers. Recent experimental studies by Lee et al. [45, 46] and Yan et al. [47] have shown large modulation depths (approaching 90%) in such structures due to electric-field enhancement in graphene at the FSS resonance frequency. Periodically patterned graphene can be employed as a platform for plasmonic applications [22, 23, 48]. Recent studies by Otsuji et al. [49], Ryzhii et al. [50], Sensale-Rodriguez [51], etc. have also discussed the possibility of obtaining THz lasing, detection, and amplification in plasmonic graphene devices.

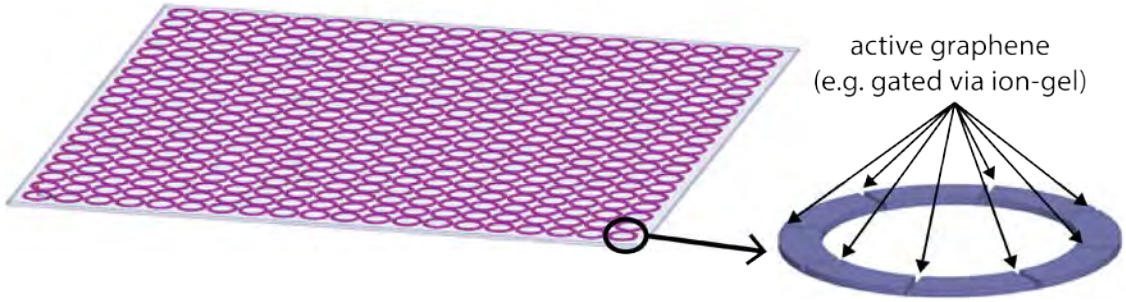

Fig. 2 Sketch of a graphene-based tunable metamaterial THz filter. Sketches of the whole device and a unit cell are depicted in the left and right panel, respectively. Control of the graphene conductivity can be obtained via employing ion-gel or self-gated graphene pairs as proposed in the literature. The unit cell size is 500 μm^2. Inner/outer radii of the rings are 200 and 240 μm, respectively. The gap, where graphene is placed, is 3μm wide. The area of the active region is thus <1% of the total device area, which might boost the response time of the device.

However, there are several important functionality limitations yet to be addressed in terms of graphene THz reconfigurable devices: (i) How to efficiently tune the characteristic resonance of the structure? -in order to develop active filters-, and (ii) How to reduce the area of the active region? -in order to boost the response time of the device-. In addition, one of the drawbacks of the metamaterial devices proposed to date is the tradeoff between degree of reconfigurability and insertion loss. In this section we discuss a new type of active metasurface based on graphene that is capable of operating as an efficient tunable filter thus simultaneously address all these issues. The device geometry and a detailed sketch of the metasurface unit-cell are depicted in Fig. 2 (left and right, respectively). Gating of the graphene can be practically implemented via employing self-gated graphene layers [52] or via employing ion-gel [22]. Full wave finite element simulations were performed employing by HFSS; in these simulations the metal was considered as gold, and graphene was modelled as a very thin conductive layer with associated conductivity swinging between 0.1 and 1mS per layer.

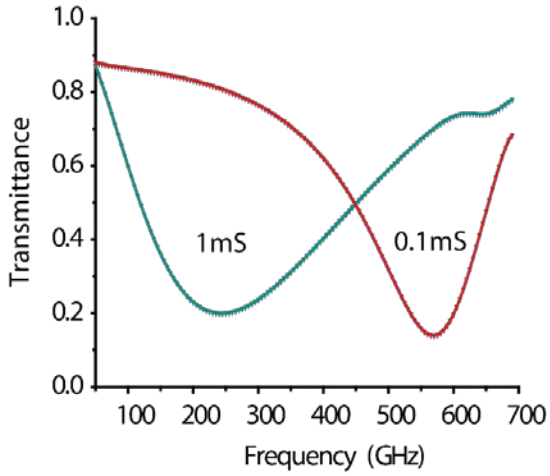

Fig. 3 Simulated transmittance versus frequency. At high graphene conductivities, the structure exhibits a resonance at around 250 *GHz*, which is a result of the current loop through the structure being closed. However, at low graphene conductivities, the current loop opens and the resonance shifts to around 570 *GHz*.

Shown in Fig. 3 are simulations describing the physical mechanism behind the device reconfigurability. If the conductivity of the active region is set to high (1*mS*), i.e. the current loop is closed, the structure presents a resonance at ~250 *GHz* (green curve). At this frequency the transmittance is ~20%. In contrast, if the conductivity of the active region is set to low (0.1 *mS*), then the current loop in the structure is open and this ~250 *GHz* resonance disappears; however, a second resonance at higher frequency (~570 *GHz*) can be excited (red curve). When placing graphene in the active region and controlling its conductivity, e.g. by means of field effect in a graphene self-gated pair, the frequency response of the structure can be tuned between the previously described states as described above.

5. Conclusions

As a result of its particular band structure, graphene poses extraordinary linear and nonlinear THz properties. This paper explored these properties and some of their applications. By taking into account the effect of electron momentum relaxation time on intra-band optical transitions, which is significant at microwave and terahertz frequencies, an analytical form for the third order microwave/THz susceptibility in graphene was derived, which was found to be $>10^3$ times larger than that of other typical nonlinear THz materials. Current and future applications of such properties in THz devices were discussed, in particular, the application of graphene into active THz filters was proposed.

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