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Nonlinear optical conductance in a graphene pn junction in the terahertz regime

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We show that the optical responses of a graphene pn junction is dominated by nonlinear intraband and interband processes. At the experimentally relevant electric field intensity, nonlinear conductance is an order of magnitude larger than the linear conductance. Furthermore, the total conductance is negative in the terahertz to far infrared regime. The negative conductance provides a unique mechanism for photon generation in graphene and could be used for developing coherent terahertz radiation sources. © 2010 American Institute of Physics. [doi:10.1063/1.3462972]

Graphene is a one-atom-thick planar sheet of carbon atoms that are densely packed in a honeycomb crystal lattice.¹ The fabrication of graphene overthrows the prediction that strictly two-dimensional crystal cannot exist in finite temperature because of thermal disturbance.² Graphene has many peculiar properties and potential applications. For example, the prediction and observation of half-integer quantum hall effect,³ finite conductivity at zero charge carrier concentration,⁴ perfect quantum tunneling effect,⁵ and ultra-high carrier mobility.⁶

The optical conductance of graphene based systems in the terahertz to far infrared regime has been a topic of intense interest due to the ongoing search for viable terahertz detectors and emitters. A series of work searching for application of graphene in terahertz science and technology have been carried out and suggest that graphene can be used in building innovative devices for terahertz optoelectronics. Within the classical picture,^{7,8} it has been shown that graphene is a strong nonlinear material and frequency upconversion can be achieved. This strong nonlinear effect has been confirmed using a quantum mechanical approach.^{9,10} Strong terahertz response of graphene nanoribbon^{11,12} has also been found. Graphene based terahertz plasmon oscillators,¹³ lasers,^{14,15} and detectors¹⁶ have also been proposed and investigated.

The purpose of this paper is to demonstrate that strong nonlinear optical response can exist in a graphene pn junctions in the terahertz to infrared frequency regime under a moderate electric field. First we study the linear and nonlinear optical conductivities associated with intraband transitions based on Boltzmann transport equation. Then we adopt quantum mechanical approach to study the linear and nonlinear optical conductivities associated with interband transitions. At last we obtain the total optical conductivity including the intraband and interband absorption and radiation processes.

In a graphene one can use a gate voltage to control the Fermi energy to form p and n regions. The linear optical conductivity associated with intraband and interband transitions has been calculated in Ref. 14. For a single layer

graphene with the substrate thickness of SiO₂ layer 300 nm and the gate voltage V_g , carrier sheet density is $7.3 \times 10^{10} V_g$ (cm⁻²).⁴ From the sheet density the Fermi energy can be obtained. Here we adopt the same model as the one used in Ref. 14 with the gate voltage of the p region $V_g/2$ and n region $-V_g/2$, and with a forward bias voltage V_b between p and n leads.

Now we proceed to calculate the optical conductance associated with intraband transitions of graphene pn junction. For a junction under a time dependent electric field $\mathbf{E}(t) = \mathbf{E}_0 e^{i\omega t}$ along the x direction, the electron distribution function can be calculated from the Boltzmann equation,

$$\frac{\partial f}{\partial t} - \frac{\partial f}{\partial \mathbf{p}} \times e\mathbf{E} = -\frac{f - f_0}{\tau}, \quad (1)$$

where $f_0(v_F p) = [1 + \exp(v_F p - \varepsilon_F / k_B T)]^{-1}$ is the equilibrium Fermi distribution function, f is the nonequilibrium Fermi distribution function, τ is the relaxation time of electron and hole caused by impurity and phonon scattering. Letting $F = f - f_0$ and substituting $\partial F / \partial t = i\omega F$ into Eq. (1), we obtain $F_n = (e\tau E_0 / (1 + i\omega\tau)) \times (\partial F_{n-1} / \partial p_x)$ ($n \geq 1$), where $F_0 = f_0$. The corresponding current density is $\mathbf{J}_n^{\text{intra}} = 1/2 \pi^2 \hbar^2 \int -e \mathbf{v}_x \cdot \mathbf{F}_n \cdot d\mathbf{p}$, where $\mathbf{v}_x = v_F \cos \theta$, with the Fermi velocity $v_F \approx c/300$. Because $\int d\mathbf{p} = \int_0^{2\pi} d\theta \int_0^\infty p dp$ and $(\partial F_{n-1} / \partial p_x) = (\partial F_{n-1} / \partial p) \cos \theta$, the n^{th} order optical conductivity associated with intraband transitions $\sigma_n^{\text{intra}} = \mathbf{J}_n^{\text{intra}} / \mathbf{E}_0$ can be written as,

$$\sigma_n^{\text{intra}} = \frac{e^2 v_F \tau}{2\pi^2 \hbar^2 (1 + i\omega\tau)} \int_0^{2\pi} \cos^2 \theta d\theta \int_0^\infty -\frac{\partial F_{n-1}}{\partial p} p dp. \quad (2)$$

Here we are only interested in the real part of the optical conductivity, which is defined as optical conductance. The linear optical conductance associated with intraband transitions is given as,

$$\text{Re}(\sigma_1^{\text{intra}}) = \frac{e^2}{2\hbar} \times \frac{v_F \tau}{\pi \hbar (1 + \omega^2 \tau^2)} \int_0^\infty -\frac{\partial f_0}{\partial p} p dp. \quad (3)$$

By induction, we substitute F_1 to Eq. (2) and obtain the second order optical conductivity associated with intraband transitions. As expected σ_2^{intra} equals to zero due to the time-

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reversal symmetry. The third order conductance associated with intraband transitions can be written as

$$\text{Re}(\sigma_3^{\text{intra}}) = \frac{e^2}{2\hbar} \cdot \frac{3e^2 v_F \tau^3 E_0^2 (1 - 11\omega^2 \tau^2)}{4\pi\hbar [(1 - 11\omega^2 \tau^2)^2 + (6\omega\tau - 6\omega^3 \tau^3)^2]} \times \int_0^\infty \left(\frac{1}{p} \frac{\partial f_0}{\partial p} - \frac{\partial^2 f_0}{\partial p^2} - p \frac{\partial^3 f_0}{\partial p^3} \right) dp. \quad (4)$$

To calculate the nonlinear interband optical conductance, we adopt a quantum-mechanical approach⁹ and extend it to non-equilibrium graphene pn junction. The tight binding Hamiltonian in the low energy regime is given by

$$H = v_F \begin{pmatrix} 0 & p_- + eA_- \\ p_+ + eA_+ & 0 \end{pmatrix}, \quad (5)$$

where $p_\pm = p_x \pm ip_y$ and $A_\pm = A_- = \frac{E_0}{i\omega} e^{i\omega t}$. The time-dependent two-component wave functions can be expanded in the basis set

$$\psi(\mathbf{p}, t) = \sum_{n=0}^\infty \phi_n(\mathbf{p}) e^{in\omega t} e^{-i\epsilon t/\hbar}, \quad (6)$$

where $\epsilon = v_F |\mathbf{p}|$ and $\phi_n(\mathbf{p})$ is a spinor given by $\phi_n(\mathbf{p}) = [\alpha_n, \beta_n]^T$ representing the transpose of $[\alpha_n, \beta_n]$. By substituting Eq. (6) into the Schrödinger equation $i\hbar \partial \psi / \partial t = H \psi$, we obtain the information of all multiple photon processes. Due to the orthonormal relation of $e^{in\omega t}$, we can write the coupled recursion relations for the spinor components,

$$\alpha_n = \frac{eE_0 v_F}{i\omega\hbar} \times \frac{v_F p_- \alpha_{n-1} + (v_F p - n\hbar\omega) \beta_{n-1}}{n\omega(n\hbar\omega - 2v_F p)},$$

$$\beta_n = \frac{eE_0 v_F}{i\omega\hbar} \times \frac{v_F p_+ \beta_{n-1} + (v_F p - n\hbar\omega) \alpha_{n-1}}{n\omega(n\hbar\omega - 2v_F p)}. \quad (7)$$

The recursion relations couple the n photon processes to the $n-1$ photon processes. When the electric field is zero, only $n=0$ terms are nonzero and the solution to Eq. (7) is the usual wave functions for the massless Dirac fermions. To assure normalization we let $\alpha_0 = p_- / \sqrt{2p}$, $\beta_0 = 1 / \sqrt{2}$. From the solutions to Eq. (7) we can calculate the n th order current given by $\mathbf{J}_n = \frac{1}{4\pi} \int \mathbf{j}_n N(\epsilon) d\mathbf{p}$, where $N(\epsilon) = [1 + \exp(-\epsilon - \epsilon_F + eV_b/k_B T)]^{-1} - [1 + \exp(\epsilon - \epsilon_F/k_B T)]^{-1}$ is the thermal factor and $\mathbf{j}_n = -e\psi^\dagger \hat{v}_x \psi$ is the current operator, where $\hat{v}_x = \partial H / \partial p_x$. The real part of the n th order optical conductivity associated with interband transitions is given as,

$$\sigma_n^{\text{inter}} = \frac{1}{2\pi^2 E_0} \int_0^{2\pi} d\theta \int_0^\infty N(\epsilon) p dp \text{Re} \left(\sum_{i=0}^n \alpha_i^* \beta_{n-i} + \beta_i^* \alpha_{n-i} \right). \quad (8)$$

The current calculated with $n=1$ terms is equivalent to the linear response result,

$$\sigma_1^{\text{inter}} = \frac{e^2}{4\hbar} \times N\left(\frac{\hbar\omega}{2}\right) = \frac{e^2}{4\hbar} \left[1 - f_0\left(\frac{\hbar\omega}{2}\right) - f_1\left(\frac{\hbar\omega}{2}\right) \right], \quad (9)$$

where we make $f_i(v_F p) = [1 + \exp(v_F p + \epsilon_F - eV_b/k_B T)]^{-1}$. This is same as the result in Refs. 14 and 17 calculated with the Green function method. The second order optical con-

FIG. 1. (Color online) The real part of linear optical conductivity of graphene pn junction as a function of frequency for different relaxation times with $\epsilon_F = 24$ meV ($V_g = 18.56$ V) and bias voltage $V_b = 40$ mV.

ductivity associated with interband transitions is also zero because of time-reversal symmetry. The third order optical conductivity associated with interband transitions is obtained as follows:

$$\sigma_3^{\text{inter}} = \frac{e^2}{4\hbar} \times \frac{e^2 v_F^2 E_0^2}{\hbar^2 \omega^4} [N_3(\omega) \times e^{i3\omega t} + N_1(\omega) \times e^{i\omega t}], \quad (10)$$

where $N_3(\omega) = (13/48)N(\hbar\omega/2) - (2/3)N(\hbar\omega) + (45/48)N(3\hbar\omega/2)$ and $N_1(\omega) = 2N(\hbar\omega)$. σ_3^{inter} consists of contributions due to two distinctive processes. The term proportional to $e^{i3\omega t}$ corresponds to a process of simultaneous absorption of three photons. The term proportional to $e^{i\omega t}$ corresponds to a process of absorbing two photons and immediately emitting one photon. In this paper we consider nonlinear responses up to the third order processes. The total optical conductivity is the sums of intraband and interband linear terms and the third order intraband nonlinear term and the third order interband term oscillating with ω .

In Fig. 1, we plot the frequency dependent linear optical conductance of a graphene pn junction in unit of $e^2/2\hbar$. When the Fermi energy ϵ_F is 24 meV ($V_g = 18.56$ V) and the bias voltage V_b is 40 mV, there exists a frequency region of 0–10 THz where the conductance are negative. The negative conductance is due to the interband transitions. From Eq. (9) we can conclude $N(\hbar\omega/2) < 0$ will lead to a negative σ_1^{inter} . That is to say, $\text{Re}(\sigma_1^{\text{inter}})$ will be negative under the condition $\omega < eV_b/\hbar$. When $V_b = 40$ mV, $\text{Re}(\sigma_1^{\text{inter}})$ will be negative in the range of frequencies below 10 THz. The sums of the linear optical conductivities $\text{Re}(\sigma_1^{\text{inter}})$ and $\text{Re}(\sigma_1^{\text{intra}})$ are negative in some part of the terahertz regime. We found that when the frequency increases the relaxation time will have a negligible effect on the linear optical conductivity.

Figure 2 shows the electric field intensity dependence of the nonlinear optical conductance. It is shown that when the electric field is weak, the nonlinear process is negligible. The ratio of nonlinear and linear optical conductance increases with electric field intensity. With the same electric field intensity the ratio decreases with increasing frequency. For weak fields and high frequencies, the linear term dominates, but for strong fields and low frequencies, the nonlinear term dominates. The nonlinear conductance can be an order mag-

FIG. 2. (Color online) The ratio of the real part of nonlinear and linear optical conductivity vs the electric field intensity with $\epsilon_F=24$ meV, bias voltage $V_b=40$ mV, and the relaxation time 10 ps.

nitude larger than the linear conductance when the electric field intensity is around 10^3 V/cm. Our analysis indicates that the relaxation time only affect the nonlinear processes very weakly.

Figure 3 depicts the total conductance of a graphene pn junction under a forward bias as a function of frequency with the relaxation time fixed. It is found that at a moderate field strength the real part of the total conductivity is negative in a very wide frequency range covering the entire terahertz regime. This anomaly of negative conductance is mainly due to that the third order optical transition is about an order of magnitude stronger than the linear order transition. From

FIG. 3. (Color online) The real part of total optical conductivity of graphene pn junction as a function of frequency for different electric field intensity with $\tau=10$ ps, $\epsilon_F=24$ meV, and bias voltage $V_b=40$ mV.

Fig. 3, though the electric field intensities are different, the conductance approach to the same high frequency limit.

The negative conductance means the electromagnetic field can be amplified in this type of pn junctions. The p and n regions in these junctions can serve as active regions of a laser with the pertinent cavity for terahertz radiation. This can result in spatiotemporal self-consistent oscillations of electric potential and carriers (electron and hole) densities in p and n regions.^{18,19} It has also been demonstrated that the oscillations of charges can lead to the oscillations of the dipole momentum of the system resulting in the terahertz emission,¹⁴ where the gates can play the role of an antenna converting potential oscillations into electromagnetic waves.

In conclusion, we have shown that the nonlinear response in a graphene pn junction under a forward bias leads to a strong amplification effect in the terahertz regime. This result can be useful in developing graphene-based terahertz radiation sources and other optoelectronics devices.

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