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DC current induced second order optical nonlinearity in graphene

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Abstract: We calculate the dc current induced second harmonic generation in doped graphene using the semiconductor Bloch equations under relaxation time approximations. We find that the maximum value of the effective second order susceptibility appears when the fundamental photon energy matches the chemical potential. For a surface current density $1.1 \times 10^3$ A/m and a relaxation time at optical frequencies of 13 fs, the effective second order susceptibility $\chi^{(2)\text{eff}}$ can be as large as $10^{-7}$ m/V for $\hbar\omega = 0.2$ eV or $10^{-8}$ m/V for $\hbar\omega = 0.53$ eV.

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References and links

1. Introduction

Graphene has attracted intense interest in photonics, electronics, and optoelectronics due to its unique electronic and optical properties [1, 2]. The linear optical conductivity takes the universal value [3] \( \sigma_0 = e^2/4\hbar \) over a very broad energy range, due to the linear dispersion at low energy excitations. In the dipole approximation, second order nonlinearities are absent in pristine graphene due to the inversion symmetry of the honeycomb lattice, but third order nonlinearities are observed to be several orders of magnitude larger than in conventional semiconductors and metals [2, 4–10]. Various third-order processes have been measured, including four wave mixing [4], third harmonic generation [5, 6], self-phase modulation [2, 7, 8], two-photon absorption [9], and two-color coherent current injection [10]. The electrical properties of graphene are also striking, especially at low carrier densities. The mobility [11] can be as high as \( 2 \times 10^6 \) cm\(^2\)V\(^{-1}\)S\(^{-1}\) for carrier densities below \( 5 \times 10^9 \) cm\(^{-2}\); in that density regime a minimum conductivity [12–14] on the order of \( 4e^2/\pi\hbar \) has been observed experimentally. At higher carrier densities, the electron transport is diffusive, with the dc resistance [15] mainly due to electron scattering from local and remote impurities, and electron-phonon scattering; this is well described by Boltzmann transport theory [16, 17]. Experimentally, the density and hence the conductivity can be tuned by chemical doping [18] or by applying an external gate voltage [19].

In the development of integrated graphene-based photonic devices, the presence of a second order nonlinearity is important for realizing various essential building blocks, such as fast and compact electro-optic modulators [20, 21], optical switches [22–24], and optical transistors [25, 26]. In many of these device implementations the second-order process being exploited is second harmonic generation (SHG). For instance, several commonly proposed schemes to obtain all-optical switching and transistor action rely on employing a SHG-signal as seed or output signal [22, 23, 25, 26]. Another example is the use of SHG for enhancing thermo-optical switching in ring resonators [24]. SHG in graphene-based photonic devices can arise from breaking the inversion symmetry of graphene, by growing graphene on a substrate to utilize the surface effect [27, 28], by utilizing the weak variation of the optical electric field over the unit cell and its associated magnetic field [29–33], or by applying a dc electric field to generate an asymmetric steady state [34, 35].

This work focuses on the last scenario, for which the effective second order susceptibility \( \chi^{(2)}_{\text{eff}}(\omega_1, \omega_2) \) can be described by a third order response \( \chi^{(3)}_{\text{dabc}}(\omega_1, \omega_2, 0)E_{\text{dc}}^2 \) with \( E_{\text{dc}} \) the dc field strength. In describing the dc limit \( \omega_1 \to 0 \) of \( \chi^{(3)}_{\text{dabc}}(\omega_1, \omega_2, \omega_0) \) for any material, one of two following regimes is usually appropriate: (i) in an insulator or undoped semiconductor, a dc field induces a static electric polarization, which gives rise to a nonzero second order optical response; this is usually called electric field induced second harmonic generation (EFISH); (ii) in a doped semiconductor or graphene, besides inducing a static electric polarization the dc field drives free electrons to generate a dc current, which can result in current induced second
We start with the SBE for graphene, which we treat in the usual two-band tight binding model.

2. Model

Results with those of other materials. Estimate the CSHG coefficients at different frequencies and relaxation times, and compare the performance of the monolayer material MoS$_2$ [40]. In monolayer graphene the CSHG has already been experimentally observed [35], and intraband contributions to it have been theoretically studied [33]. However, an evaluation of the contribution from interband transitions, which are usually important in the optical frequency regime, is still lacking. A full theoretical treatment is of interest as monolayer graphene could be a very suitable material for generating strong CSHG, since the maximum surface current density in monolayer graphene can be up to 3000 A/m [41], which is several order of magnitude higher than that in regular semiconductors.

In this paper, we theoretically consider the dc current induced second order nonlinearity in doped monolayer graphene while including the interband transitions. We first introduce the semiconductor Bloch equations (SBE) to describe the dc and optical response of graphene under relaxation time approximations. We calculate the shift of the carrier distribution in the Fermi-Dirac distribution at temperature $T$.

Nonetheless, even with the use of the two phenomenological constants we employ below, $\tau_1$ for the response to the dc field and $\tau_2$ for the response to the optical field, we are able to obtain an analytic result for the perturbation calculation within the linear dispersion approximations around the Dirac cones.

Before the optical field is turned on, we assume the dc field drives the system to a steady state $\rho_k^{dc}$. Even here, in a full microscopic theory it would be important to take into account
carrier-carrier scattering, carrier-phonon scattering, and carrier-impurity scattering, leading to a very complicated calculation [48,49]. But within the framework of Eq. (1), for a weak dc field the shifted populations $n_{sk+\delta k}$, with $\delta k \approx eE_{dc}t_1/\hbar$ constitute a good approximation to steady state; they follow from a perturbative solution of the steady state equation

$$0 = -\frac{i}{\hbar} [\delta k, eE_{dc} \cdot \xi_{sk} + \rho_{sk}^{dc}] - \frac{e}{\hbar} E_{dc} \cdot \nabla k \rho_{sk}^{dc} - \frac{\rho_{sk}^{dc} - \rho_{sk}^0}{\tau_1},$$

(2)

In general, we write $\rho_{sk}^{dc} = \rho_{sk}^0 + \delta \rho_{sk}^D + \delta \rho_{sk}^N$, collecting terms which are divergent with $\tau_1^{-1} \rightarrow 0$ in $\delta \rho_{sk}^D$, and all other terms in $\delta \rho_{sk}^N$. Up to the linear term of $E_{dc}$, Eq. (2) can be solved perturbatively: the $\delta \rho_{sk}^D$ only have diagonal terms

$$\delta \rho_{sk}^D = -\frac{e}{\hbar} E_{dc} \tau_1 \frac{\partial n_{sk}}{\partial k_c},$$

(3)

which arise from the first order of the expansion of $n_{sk+\delta k}$; the $\delta \rho_{sk}^N$ have only off-diagonal terms

$$\delta \rho_{sk}^N = -\frac{e}{\hbar} E_{dc} \frac{\varepsilon_{sk}^c}{\omega_{sk}^{c+sk}} \left( n_{sk} - n_{sk+\delta k} \right),$$

(4)

which are associated with the interband polarization. Hereafter Roman scripts indicate Cartesian components, to be summed over if repeated. Considering the large interband energy difference relevant to doped samples, it is safe to set $\tau_1^{-1} \rightarrow 0$ in $\delta \rho_{sk}^N$. The dc current is then calculated as $J_{dc} = e \sum_{sk} \delta \rho_{sk}^D + e \sum_{sk} \delta \rho_{sk}^N$. In the clean limit $\tau_1^{-1} \rightarrow 0$, it is easy to check that the contribution from $\delta \rho_{sk}^N$ to $J_{dc}$ vanishes by using Eq. (4), while the contribution from $\delta \rho_{sk}^D$ diverges by using Eq. (3). Both $\delta \rho_{sk}^D$ and $\delta \rho_{sk}^N$ can induce nonzero SHG, and correspond to the CSHG and EFISH respectively. Later we show the ratio between the conductivity from the intraband and interband contribution is about $|\mu|/\tau_1/\hbar$, hence CSHG will dominate if it is larger than unity. For characteristic values in doped graphene, $\mu$ is below but on the order of $1$ eV, and $\tau_1$ varies from picosecond to subpicosecond [50]. Thus the ratio is larger than unity, and in doped graphene we expect CSHG to be the much larger effect, and the one we study here. Hence we put $\delta \rho_{sk}^N \rightarrow 0$.

With the optical field applied, the system responds as $\rho_{sk}(t) = \rho_{sk}^{dc} + \rho_{sk}^{op}(t)$, with the contribution to the density matrix due to the optical excitation, $\rho_{sk}^{op}(t)$, satisfying the equation

$$\frac{\partial \rho_{sk}^{op}}{\partial t} = -\frac{i}{\hbar} [\delta k, \rho_{sk}^{op}] + \frac{ie}{\hbar} [E_{dc} + E(t)] \cdot (\xi_{sk} \rho_{sk}^{op}) + \frac{ie}{\hbar} [E_{dc} + E(t)] \cdot (\xi_{sk} \rho_{sk}^{dc}) + \frac{ie}{\hbar} [E_{dc} + E(t)] \cdot (\xi_{sk} \rho_{sk}^{op})$$

$$+ \frac{ie}{\hbar} [E_{dc} + E(t)] \cdot (\xi_{sk} \rho_{sk}^{dc}) + \frac{ie}{\hbar} [E(t) \cdot (\xi_{sk} \rho_{sk}^{op}) + \frac{i\nabla k \rho_{sk}^{dc}}{\tau_2} - \frac{\rho_{sk}^{op}(t)}{\tau_2},$$

(5)

We note that in this equation the dc field $E_{dc}$ does not lead to any divergence, even as $\tau_2^{-1} \rightarrow 0$, because of the nonzero frequency of $E(t)$. In fact, the $E_{dc}$ term in Eq. (5) is related to the asymmetric optical transition matrix elements in the presence of a dc field, which are usually ignored in CSHG calculations in ordinary semiconductor systems. Following the calculations [34, 37], we ignore them for a weak dc field. However, due to the unique band structure in graphene, an explicit verification of the validity of this assumption is worthwhile; we return to it in a future communication. When $E_{dc}$ is neglected in Eq. (5), the equation for $\rho_{sk}^{op}$ can be approximated as

$$\frac{\partial \rho_{sk}^{op}}{\partial t} = -\frac{i}{\hbar} [\delta k, \rho_{sk}^{op}] + \frac{ie}{\hbar} [E(t)] \cdot (\xi_{sk} \rho_{sk}^{op}) + \frac{ie}{\hbar} [E(t)] \cdot (\xi_{sk} \rho_{sk}^{op})$$

$$+ \frac{ie}{\hbar} [E(t)] \cdot (\xi_{sk} \rho_{sk}^{op}) + \frac{i\nabla k \rho_{sk}^{dc}}{\tau_2} - \frac{\rho_{sk}^{op}(t)}{\tau_2},$$

(6)
Up to the second order in $E(t)$, we have

$$\rho_k^{\text{op}}(t) = \rho_k^{\text{op}(1)}(t) + \rho_k^{\text{op}(2)}(t) + \cdots,$$

(7)

with

$$\frac{\partial \rho_k^{\text{op}(1)}}{\partial t} = -\frac{i}{\hbar}[\epsilon_k, \rho_k^{\text{op}(1)}] + \frac{ie}{\hbar}E(t) \cdot (X_{kkk} - \delta \rho_k^D) - \frac{\rho_k^{\text{op}(1)}}{\tau_2},$$

(8)

$$\frac{\partial \rho_k^{\text{op}(2)}}{\partial t} = -\frac{i}{\hbar}[\epsilon_k, \rho_k^{\text{op}(2)}] + \frac{ie}{\hbar}E(t) \cdot (X_{kkk} - \delta \rho_k^D) - \frac{\rho_k^{\text{op}(2)}}{\tau_2}.$$

(9)

Putting $E(t) = \int \frac{d\omega}{2\pi} e^{-i\omega t} E\omega$, we can solve these equations to find

$$\rho_k^{\text{op}(1)}(t) = -\int \frac{d\omega}{2\pi} e^\omega \rho_k^{\text{op}(1)}(\omega),$$

(10)

and

$$\rho_k^{\text{op}(2)}(t) = \int \frac{d\omega_1}{2\pi} \frac{d\omega_2}{2\pi} \int \frac{d\omega}{2\pi} E^a \omega_1 e^{-i\omega \omega^\prime} \rho_k^{\text{op}(2)}(\omega_1, \omega_2).$$

(11)

with $\omega_0 = \omega_1 + \omega_2$, where

$$\rho_k^{\text{op}(1)}(\omega_2) = \int_{0}^\omega \left( [\epsilon_k, \delta \rho_k^D] + i \frac{\partial \delta \rho_k^D}{\partial k_b} \right),$$

(12)

and

$$\rho_k^{\text{op}(2)}(\omega_1, \omega_2) = \int_{0}^\omega \left( [\epsilon_k, \rho_k^{\text{op}(1)}(\omega_2)] + i \frac{\partial \rho_k^{\text{op}(1)}(\omega_2)}{\partial k_a} \right).$$

(13)

Here $\omega_k^\pm = \omega_2 + i\tau_2^{-1}$ and $\omega_0^\pm = \omega_1 + \omega_2 + i\tau_2^{-1}$; $I^\omega(X_k)$ is a matrix function of a $k$-dependent matrix $X_k$, for which the components are defined as $I_{\omega_j \omega_k}(X_k) = X_{\omega_j \omega_k}(\omega - \omega_j \omega_k)$, with $X_{\omega_j \omega_k} = \epsilon_{\omega_j \omega_k} - \epsilon_{\omega_j \omega_k}$. The dc current induced second order optical current $J^{(2,d)}(t)$ and second order optical conductivity $\sigma^{(2,d)ab}(\omega_1, \omega_2)$ are given as

$$J^{(2,d)}(t) = \epsilon \sum_k \text{Tr}[\delta \rho_k^{\text{op}(2)}(t)]$$

$$\sigma^{(2,d)ab}(\omega_1, \omega_2) = \frac{1}{2} \left[ \sigma^{(2,d)ab}_{\text{eff},j}(\omega_1, \omega_2) + \sigma^{(2,d)ab}_{\text{eff},j}(\omega_2, \omega_1) \right],$$

$$\sigma^{(2,d)ab}_{\text{eff},j}(\omega_1, \omega_2) = \epsilon^3 \sum_k \text{Tr}[\delta \rho_k^{\text{op}(2)}(\omega_1, \omega_2)].$$

(14)

(15)

3. Results

We now use these equations to calculate the current induced second order nonlinearities in doped graphene with optical transitions around the Dirac cones $K$ and $K'$. Each of these cones provides the same contribution, so we only consider the conductivity induced by transitions around the $K$ point explicitly and double it to get the total result, including also a factor of 2 to for the spin degeneracy. As in our previous work [36], we use a tight binding model to describe the electron states in the $\pi (s = -)$ and $\pi^+ (s = +)$ bands, and use the standard approximations.

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by using plots of them, we take the zero temperature limit. Here the column vector are \( \sigma \) of nonzero components, and with \( B \) we find that the current induced second order conductivity can be written as between these two terms is \( \frac{A}{w} \). In the expressions for the response coefficients given below, and in the plots of them, we take the zero temperature limit.

To derive the correct linear response in the undoped limit, we reinstate \( \Delta \) and, within our approximations, Eq. (3) and (4) are given by

\[
\sigma_{\text{dc}}^{\text{xx}} = \sigma_0 \frac{\tau_1 |\mu|}{h} + \frac{2}{\pi} \sigma_0 \arctan \left( \frac{h}{2|\mu| \tau_1} \right)
\]

The first term is the Drude term, while the second term corresponds to the dc limit of the optical conductivity, which is zero for doped graphene in the clean limit \( \tau_1^{-1} \rightarrow 0 \), and returns to the universal optical conductivity \( \sigma_0 \) as \( \mu \rightarrow 0 \) with \( \tau_1 \) kept finite. The lower bound of the ratio between these two terms is \( |\mu| \tau_1 / h \).

Turning to the nonlinear response in doped graphene, with the neglect of EFISH (\( \Delta \rightarrow 0 \)) we find that the current induced second order conductivity can be written as \( \sigma_{\text{eff},j}^{(2),dabc}(\omega_1, \omega_2) = \sigma_j^{dabc}(\omega_1, \omega_2) \sigma_{\text{dc}} \) with \( \sigma_j^{dabc}(\omega_1, \omega_2) = -\frac{\hbar \epsilon}{8 \epsilon_0} \). Here the dimensionless quantity \( S_{dabc} \) is a fourth order tensor. Considering that the point group of the graphene monolayer is \( D_{6h} \), there are only three independent components \( S^{xyy}, S^{yyx}, \) and \( S^{xyz} \) [51], which are given as

\[
S(w_1, w_2, \gamma) = 4 \left( \frac{w_1^2 - w_2^2}{w_1^2 - w_2^2} \right) + 4 \left( \frac{w_1 - w_2}{w_1^2 - w_2^2} \right) + \frac{4}{w_1^2 - w_2^2} \left( \frac{w_1 - w_2}{w_1^2 - w_2^2} \right)
\]

Here the column vector \( S = [S^{xyy}, S^{yyx}, S^{xyz}]^T \) is used to represent these three independent nonzero components, and \( \sigma_1 = [1, -3, 1]^T, \sigma_2 = [1, -3, 1]^T, \sigma_3 = [1, 1, -3]^T, \sigma_0 = -(\sigma_1 + \sigma_2 + \sigma_3) \). The other nonzero components can be obtained by using \( S_{dabc} = S^{xyy} + S^{yyx} + S^{xyz} \), and by interchanging index \( x \) and \( y \). For CSHG with \( w_1 = w_2 = w \), the coefficients \( S(w, w, \gamma) \) become

\[
S(w, w, \gamma) = 4 \left( \frac{w_1 - w_2}{w_1^2 - w_2^2} \right) + 4 \left( \frac{w_1^2 - w_2^2}{w_1^2 - w_2^2} \right)
\]

with \( B_0 = [-2, -2, 2]^T \). We plot the dependence of \( S^{xyy}(w, w, \gamma) \) and \( S^{xyz}(w, w, \gamma) \) on \( w \) in Fig. 1 for different values of \( \gamma \). Three resonant peaks are found in \( S(w, w, \gamma) \) at \( w = 0, w = 1, \)
and \( w = 2 \). We are only interested in the peaks at \( w = 1 \) and \( w = 2 \). These two peaks also exist in CSHG in bilayer graphene [34]: the first one is induced by the transition shown in the inset of Fig. 1(c), which also exists in monolayer graphene; while the second one is interpreted by the transition between higher bands, which is not expected in monolayer graphene. However, our calculation shows that the transition depicted in the inset of Fig. 1(d) can also induce a peak at \( w = 2 \) in monolayer graphene. The value of the peaks at \( w = 1 \) and \( w = 2 \) can be approximated by 

\[
S(1, 1, \gamma) \approx 3iB_0\gamma^{-1} \quad \text{and} \quad S(2, 2, \gamma) \approx -3iB_0\gamma^{-1}/8 \quad \text{respectively.}
\]

Within this approximation, the second order conductivities at both peaks satisfy 

\[
\sigma_{2,\text{eff}} \propto \tau_1\tau_2.
\]

From Eq. (14), the optical

\[
J^{(2)}_{\omega,\text{op}}(2\omega) = 2\sigma_1E_\omega E_\omega \cdot J_{\text{dc}} + \sigma_2J_{\text{dc}}E_\omega \cdot E_\omega.
\]

where \( \sigma_1 = \sigma_j^{\text{xxxy}} = \sigma_j^{\text{yyyy}} \) and \( \sigma_2 = \sigma_j^{\text{xyxy}} \). Around the resonant peak at \( w = 1 \), the conductivity satisfies \( \sigma_2 \approx -\sigma_1 \), and the SHG current is given by 

\[
J^{(2)}_{\omega,\text{op}}(2\omega) = \sigma_1[\cos(2\theta - \phi)\hat{e}_x + \sin(2\theta - \phi)\hat{e}_y]E_\omega.
\]
\[ \phi \hat{\epsilon}_1 E_{ao}^2 J_{dc} \] for the linear optical field \( E_{ao} = E_{ao}(\sin \theta \hat{e}_x + \cos \theta \hat{e}_y) \) and surface dc current \( J_{dc} = J_{dc}(\sin \phi \hat{e}_x + \cos \phi \hat{e}_y) \). For circularly polarized light, the optical SHG current is \( J_{ao}^{(2)}(2\omega) = 2\sigma_1 E_{ao} \cdot J_{dc} \), which has the same direction as \( E_{ao} \).

An effective SHG coefficient can be constructed according to \( \chi_{\text{eff}}^{(2);dab} = \sigma_j^{(3);dabc} J_{dc}/(2\omega \omega_0 d_{gr}) \), with \( d_{gr} = 3.3 \, \text{Å} \) the effective thickness of graphene. By taking \( \tau_1 = 1 \, \text{ps} \) and \( E_{dc} = 0.1 \, \text{kV/cm} \) yielding a \( \mathbf{k} \)-space shift \( \delta k = 1.5 \times 10^3 / \text{m} \), which is the same as in Wu et al.’s calculation [34], the estimated surface dc current density is \( J_{dc} = 1.1 \times 10^3 \, \text{A/m} \), much larger than the value in bilayer graphene due to the difference between the parabolic and linear band structures. For a comparison, we take the chemical potential \( \mu = 0.2 \, \text{eV} \) and the optical damping \( \Gamma_2 = \hbar \tau_2^{-1} = 0.05 \, \text{eV} \) from Wu et al.; the peak value at \( w = 1 \), which corresponds to a fundamental wavelength of 6.2 \, \mu m, is \( \chi_{\text{eff}}^{(2);xxx} \sim 1.2 \times 10^{-7} \, \text{m/V} \) which is comparable to Wu et al.’s result for bilayer graphene. However, for a chemical potential \( \mu = 0.53 \, \text{eV} \), which corresponds to a shorter fundamental wavelength of 2.35 \, \mu m, we find a lower peak value of \( \chi_{\text{eff}}^{(2);xxx} = 2.5 \times 10^{-9} \, \text{m/V} \) for \( \Gamma_2 = 0.05 \, \text{eV} \) and \( \chi_{\text{eff}}^{(2);xxx} = 1.24 \times 10^{-8} \, \text{m/V} \) for \( \Gamma_2 = 0.01 \, \text{eV} \). The peak CSHG coefficients at higher fundamental frequencies are smaller due to the \( |\mu|^{-4} \) dependence of \( \sigma_j^{dabc} \).

4. Discussion and conclusion

We have calculated the current induced second order response in graphene by using the semiconductor Bloch equations under relaxation time approximations. Our general equations hold for arbitrary temperatures, but we exhibited expressions for the response coefficients and plotted their values in the zero temperature limit. These depend strongly on the scattering time (both at dc and at optical frequencies), the chemical potential, and the fundamental frequency. Since the thermal energy at room temperature is about 30 meV, which is much smaller than the Fermi energies in which we are interested (about 0.5 eV), the effects of finite temperature can be expected to be negligible, except insofar as they affect the scattering times; taking the limit to zero temperature allowed us to give analytic expressions for the response coefficients, which should lead to simple estimates and an improved physical understanding. A maximum value of the \( \sigma_j^{(2);dab} \) is found when the energy of the fundamental photon matches the chemical potential. For a surface current density \( 10^3 \, \text{A/m} \) in graphene, the effective \( \chi_{\text{eff}}^{(2);xxx} \) can be as large as \( 10^{-7} \, \text{m/V} \) for \( \hbar \omega = 0.2 \, \text{eV} \) or \( 10^{-8} \, \text{m/V} \) for \( \hbar \omega = 0.53 \, \text{eV} \). For the same parameters, the coefficient of CSHG is comparable to that in bilayer graphene [34]. For the usual scenario of graphene on a substrate there is always the possibility of interface effects also leading to SHG, and there will be forbidden contributions due to deviations from the electric dipole approximation in the calculation of the optical response. But CSHG can be controlled by adjusting the dc current, and so it should be possible both to separate out any CSHG contribution from the total SHG, and as well control the total SHG, by adjusting the dc current [35]. Thus our calculations show that the use of dc current to break inversion symmetry and make graphene second-harmonic active should lead to the effective generation of second harmonic light in graphene structures.

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