Terahertz modulated optical sideband generation in graphene

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(Dated: November 7, 2018)

Exploration of optical non-linear response of graphene predominantly relies on ultra-short time domain measurements. Here we propose an alternate technique that uses frequency modulated continuous wavefront optical fields, thereby probing graphene’s steady state non-linear response. We predict frequency sideband generation in the reflected field that originates from coherent electron dynamics of the photo-excited carriers. The corresponding threshold in input intensity for optimal sideband generation provides a direct measure of the third order optical non-linearity in graphene. Our formulation yields analytic forms for the generated sideband intensity, is applicable to generic two-band systems and suggests a range of applications that include switching of frequency sidebands using non-linear phase shifts and generation of frequency combs.

Owing to its gate tunable electronic, optical and opto-electronic properties, the exploration of non-linear optical effects in graphene has attracted significant interest in experiments [1–10] as well as in theory [11–20]. Experimentally, several non-linear optical effects such as higher harmonic generation [1, 2], third order non-linearity [3, 4, 6–10] and four wave mixing [5] have been demonstrated in graphene. There are also predictions of ultra-broad-band wave mixing at low powers, with generation of several side-bands at terahertz (THz) frequencies in bilayer graphene [21]. Such measurements and estimates offer fundamental insights in optical nonlinear interactions and relaxation mechanisms in lower dimensional systems [2, 3, 15, 21, 22] along with a promise of applications including compact and useful THz sources and gate tunable opto-electronic devices [23–25]. However, experiments till date have been primarily limited to ultra-short time-domain spectroscopy [26, 27] which are technologically involved and intricate, and physical interpretations generally rely on large scale computation.

Here we propose an alternate technique that uses frequency modulated continuous wavefront (CW) optical fields, probing optical non-linearity in the ‘steady state’. In particular we focus on the non-linear optical sideband generation in graphene due to inter-band polarization generation combined with optical Bloch oscillations [28, 29]. In presence of a CW pump (frequency ωp) and a frequency modulated probe beam (ωp + ωs) the optically pumped population inversion and the inter-band coherence oscillate at the modulation frequency. Such coherent ‘slushing’ of the inter-band quasiparticles excitations leads to a new sideband generation at frequencies ωp − ωs = 2ωp − (ωp + ωs), as shown in Fig. 1. This results in distinct signatures in reflectivity along with non-linear polarization rotation at the new sideband frequency. Our formulation based on the dynamics of density matrix for a generic two band systems, can be easily applied to other materials as well.

The predicted sideband generation is a direct consequence of non-degenerate four-wave mixing due to third-order non-linearity in graphene [28, 30]. Estimation of the corresponding intensity threshold and polarization rotation offers an alternative technique for probing non-linear optical effects and relaxation rates with CW fields in graphene [30]. Furthermore, the formulation is applicable from THz to optical domain with applications including switching with controlled non-linear phase shifts approaching π/2 with reasonable incident CW power and cascaded generation of frequency combs [31].

Our formulation starts with Hamiltonian of an electronic system interacting with an electro-magnetic field. It can be described using the dipole approximation [32], i.e., \( H = H_0 + e \mathbf{E} \cdot \mathbf{r} \). Here \( H_0 \) is the bare Hamiltonian, \( e \) is the electronic charge, and \( \mathbf{E} \) is the electric field. For simplicity we focus on a generic two-band electronic system with two spin states, \( |e\rangle \) and \( |g\rangle \), and two energy levels, \( \epsilon_g \) and \( \epsilon_e \), separated by a Zeeman energy, \( \epsilon_z \), in an external magnetic field \( \mathbf{B} \). The Hamiltonian \( H_0 \) can be written as:

\[
H_0 = \epsilon_e |e\rangle \langle e| + \epsilon_g |g\rangle \langle g| + \frac{\epsilon_z}{2} (|e\rangle \langle g| + |g\rangle \langle e|) - \frac{\hbar}{2} \mathbf{B} \cdot \mathbf{S}
\]

where \( \mathbf{S} \) is the spin operator.

For a CW pump at frequency \( \omega_p \), the time dependence is given by:

\[
\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0(t) e^{i(\omega_p t + \mathbf{k}_p \cdot \mathbf{r})}
\]

where \( \mathbf{k}_p \) is the wave vector of the pump.

The polarization of the pump and probe fields can be written as:

\[
\mathbf{P}(\mathbf{r}, t) = \mathbf{P}_0(t) e^{i(\omega_p t + \mathbf{k}_p \cdot \mathbf{r})}
\]

where \( \mathbf{P}_0(t) \) is the pump polarization.

The total polarization of the system is given by:

\[
\mathbf{P}(\mathbf{r}, t) = \mathbf{P}_0(t) + \mathbf{P}_1(t)
\]

where \( \mathbf{P}_1(t) \) is the polarization generated due to the interaction with the pump and probe fields.

The polarization \( \mathbf{P}_1(t) \) can be written as a function of the electron density matrix and the Hamiltonian \( H_0 \). The polarization \( \mathbf{P}_1(t) \) can be obtained by solving the time-dependent Schrödinger equation for the density matrix and the Hamiltonian \( H_0 \).}

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band system [33–35], with its quasiparticle dispersion described by the Hamiltonian, $H_0 = \sum_k h_k \cdot \sigma$, where $h_k = (h_{1k}, h_{2k}, h_{3k})$ is a vector composed of real scalar elements and $\sigma = (1, \sigma_x, \sigma_y, \sigma_z)$ is a vector composed of the $2 \times 2$ identity and the three Pauli matrices. The eigenvalues for conduction/valence band are, $\varepsilon_{1k} = h_{0k} \pm g_k$, where $g_k \equiv \sqrt{h_{1k}^2 + h_{2k}^2 + h_{3k}^2}$. Accordingly, the state vectors are given by, $\rho = |\psi_{1k}^{c/v}\rangle = \varepsilon_{1k}^{c/v} |\psi_{1k}^{c/v}\rangle$.

The frequency modulated electromagnetic field is, $E(t) = Re\left[|\rho|^2 e^{-i\omega_{1k}t} + |\rho|^2 e^{i(\omega_p t + \omega_s t)}\right]$, composed of a primary pump beam (of amplitude $|\rho|^2$ and frequency $\omega_p$) and a probe beam (of amplitude $|\rho|^2$ and frequency $\omega_p + \omega_s$, where $\omega_s \ll \omega_p$). In general the pump and the probe fields can have different polarization angles, $\Theta_p$ and $\Theta_s$, respectively, so that $E_p = |\rho|^2 (\cos \Theta_p, \sin \Theta_p)$ and $E_s = |\rho|^2 (\cos \Theta_s, \sin \Theta_s)$ for vertical incidence.

The dynamics of the two band system described above is obtained by analytically solving the equation of motion (EOM) for the density matrix $(\rho_k^{c/v})$. The diagonal elements of $\rho_k^{c/v}$ comprise of the carrier distribution in the conduction $(\rho_k^{c})$ and valence $(\rho_k^{v})$ bands, while the off-diagonal elements $\rho_k^{cv} = (\rho_k^{cv})^* \equiv p_k$ capture the inter-band coherence. The incident optical field ‘pumps’ the carriers from the valence band to the conduction band via vertical transitions. This optical pumping of carriers is countered by damping terms originating from the vacuum fluctuations, electron-electron interactions, electron-phonon interactions, and disorder, leading to a finite population inversion $(n_k = \rho_k^{c} - \rho_k^{v})$ – shown in Fig. 1. Including the damping terms phenomenologically in the EOM of the density matrix leads to the following set of coupled optical Bloch equation (OBE) [33–36],

$$\begin{align*}
\partial_t n_k &= 4 \text{Im} \langle \Omega_k^{c} n_k \rangle - \gamma_1 (n_k - n_k^{eq}), \\
\partial_t p_k &= i\omega_k p_k - i\Omega_k^{cv} n_k - \gamma_2 p_k.
\end{align*}$$

The inter-band Rabi frequency can be expressed in terms of the inter-band optical matrix element $M_k^{cv} = \langle \psi_k^{c} | \vec{\nabla} H_0 | \psi_k^{v}\rangle$ as: $\hbar \Omega_k^{cv}(t) = i\langle \vec{E}(t) \cdot M_k^{cv} / \omega_k \rangle$, where $\omega_k = (\varepsilon_{1k} + \varepsilon_{1k})/\hbar$ is the vertical transition frequency. In Eqs. (1)-(2), $\gamma_1$ and $\gamma_2$ are the phenomenological damping rates of the inter-band population inversion and coherence, respectively. For simplicity, we assume these rates to be constants.

For incident CW field, competition between optical pumping and decay rates, results in an eventual steady state. In this regime, analytical solutions can be obtained by making the following ansatz for the population inversion and inter-band coherence [28]:

$$\begin{align*}
n_k &= n_k^{(0)} + n_k^{(1)} e^{-i\omega_p t} + n_k^{(-1)} e^{i\omega_p t}, \\
p_k &= \left[ p_k^{(0)} + p_k^{(1)} e^{-i\omega_p t} + p_k^{(-1)} e^{i\omega_p t} \right] e^{i\omega_p t}.
\end{align*}$$

Here, the superscript $(0)$ is used to denote the steady state solution of the OBEs in presence of CW pump field leading to optical response at frequency $\omega_p$ [33–36]. Presence of a probe field leads to slowly oscillating sidebands which are denoted by the superscript $(1)$ and results in response at frequencies $\omega_1 = \omega_p + \omega_s$. Since the population inversion is a real quantity, one is forced to add a new term with the superscript $(-1)$ in the ansatz for the population inversion, such that $n_k^{(-1)} = |n_k^{(-1)}|^*$.

This leads to an additional contribution in the inter-band coherence, and accordingly, these new terms (with superscript $(-1)$) leads to optical response at frequency $\omega_1 = 2\omega_p - \omega_s$. This non-linear term in the inter-band coherence combines with the optical Bloch oscillations leading to the $\omega_1$ sideband response in the current. Its consequence in reflectivity and polarization rotation is the primary focus for this work.

Using the ansatz of Eq. (3) in Eqs. (1)-(2), and taking long time average for the steady state, we obtain

$$\begin{align*}
n_k^{(0)} &= \left( 1 + \frac{C^2 \gamma_2 \omega^2}{2} \right) \frac{|\rho|^2 \cdot M_k^{cv2}}{(\omega_k - \omega_p)^2 + \gamma_2^2} \cdot n_k^{(-1)}.
\end{align*}$$

Here, $\rho = |\rho|^2/|\rho|^2$, $M_k^{cv} = M_k^{cv}/\epsilon_{pF}$ and we have defined the dimensionless parameter $\zeta = e|\vec{E}| \epsilon_{pF}/(\hbar \omega \sqrt{2m})$ - which uniquely characterizes the non-linearity in the system [33–36] See supplementary material (SM) [37] for details of the calculation. Equation (5) can be systematically expanded in powers of $\zeta$ with $\zeta \to 0$ denoting the equilibrium distribution (Fermi function denoted by $n_k^{eq}$) in absence of optical interactions, while the $\zeta^2$ terms denote the $|\rho|^2$ correction to the modified distribution function. The $\zeta \to \infty$ limit is the saturation limit of maximum population inversion with $n_k^{(0)} \to 0$. The sideband population inversion corresponding to the probe frequency component $(\omega_p + \omega_s)$ is [37] can be expressed as,

$$n_k^{(1)} = n_k^{(0)} \langle E_s \cdot M_k^{cv} \rangle \langle E_p \cdot M_k^{cv} \rangle \xi_k.$$

Here, we have defined,

$$\xi_k = \frac{P_k}{2\hbar^2 \omega_k^2 (\omega_s + i\gamma_1) + |E_p| \cdot |M_k^{cv}|^2 \cdot Q_k},$$

$$P_k = \frac{-(\omega_k - 2i\gamma_2)}{[\omega_k - (\omega_p + \omega_s - i\gamma_2)]},$$

$$Q_k = \frac{2(\omega_s + i\gamma_2)}{[\omega_k - (\omega_p + \omega_s - i\gamma_2)]},$$

It can easily be checked that $|n_k^{(1)}| \to 0$ in both the limiting cases of vanishing intensity of the pump beam ($\zeta \to 0$) as well as in the saturation limit ($\zeta \to \infty$), as expected. Recall that $n_k^{(-1)} = |n_k^{(-1)}|^*$ and the analytical expressions for the components of the inter-band coherence are presented in Sec. S1 of the SM [37]. These $\omega_1$ sideband components of the density matrix generate a new optical sideband whose amplitude and polarization depend
TABLE I. The reflection coefficients in two dimensional materials in terms of optical conductivities [35, 38]. Here we have defined, \( \sigma_{ij} = \sigma_{ij}/\sigma_0 \), \( \sigma_0 = e^2/(4h) \), and \( \tilde{Z} = 2/(\pi \alpha_F \tilde{\sigma}_d) \) where \( \alpha_F \approx 1/137 \) is the fine structure constant, and \( \tilde{\sigma}_d = (2/\pi \alpha_F + \tilde{\sigma}_{xx}) (2/\pi \alpha_F + \tilde{\sigma}_{yy}) - \tilde{\sigma}^2_{xy} \) [39].

| Coefficient | Exact expression | \( O(\alpha_F) \) |
|-------------|-----------------|-----------------|
| \( r_{ss} \) | \[ \tilde{Z} (\tilde{Z} \sigma_d + \tilde{\sigma}_{yy}) - 1 \] | - |
| \( r_{pp} \) | \[ 1 - \tilde{Z} (\tilde{Z} \sigma_d + \tilde{\sigma}_{xx}) \] | - |
| \( r_{sp} \) | \[ -\tilde{Z} \tilde{\sigma}_{xy} \] | - |
| \( \chi_{sk} \) | \[ -r_{ps}/r_{ss} \sigma_{yy}/\sigma_{xx} \] | - |
| \( \chi_{pK} \) | \[ r_{sp}/r_{pp} - \sigma_{yy}/\sigma_{yy} \] | - |

Equation (11) clearly emphasizes the gain in the optical response at the probe frequency \( \omega_p + \omega_s \). As stated earlier, the response at frequencies \( \omega_p \) and \( \omega_p + \omega_s \) interfere leading to a sideband generation at the frequency \( \omega_{-1} = \omega_p - \omega_s \). The details of the calculations are presented in Sec. S1 and S2 of the SM [37].

The moment resolved optical conductivity due to the newly generated sideband is given by,

\[
\sigma_k^{(1)} = \sigma_k^{(0)} \left[ 1 + \frac{|E_p \cdot M_k^{cv}|^2 \xi_k}{|\omega_k - (\omega_p + \omega_s)| [\omega_k - \omega_p']^{-1}} \right].
\]

Equation (12) highlights the optical response generated at the new sideband frequency \( \omega_p - \omega_s \), and is one of the significant finding of this work. This new sideband response originates from the third order non-linearity in graphene. The dependence of the longitudinal optical conductivities on the non-linearity parameter \( \xi \propto |E_p| \) and the pump polarization angle \( \Theta_p \) is shown in Fig. 2. The transverse component of the optical conductivity are presented in Fig. S3 of the SM [37]. As expected, both \( \sigma_{xx}^{(1)} \) and \( \sigma_{xx}^{(0)} \) reduce to the universal optical conductivity of graphene, \( \sigma_0 = e^2/(4h) \), in the linear regime response of \( \xi \rightarrow 0 \). However, the new sideband contribution \( \sigma_{xx}^{(1)} \) is finite only in the non-linear regime of \( \xi \approx 1 \), and vanishes in the linear response as well as in the saturation regime (\( \xi \gg 1 \)).

The generated sideband would leave its signature in a range of optical and photo-conductivity measurements [3]. Here, we focus on its impact in optical reflectivity. In particular, we explore the pump power and polarization angle dependence of the reflectivity (amplitude and phase in \( r = \sqrt{\Re e^{i\Phi}} \)), and the Kerr angle \( \Theta_{Kerr} \) [35, 38]. For graphene, though small, the reflectivity is routinely measured [33, 35, 40, 41], while the phase of the reflection coefficient can be measured using a generic interference

FIG. 2. (a) Real part of the momentum resolved conductivity kernel corresponding to the pump (\( \sigma_{xx}^{(0)} \) - in red), probe (\( \sigma_{xx}^{(1)} \) - in green) and the newly generated sideband (\( \sigma_{xx}^{(-1)} \) - in blue) as a function of \( \log_{10} \zeta \) for \( \Theta_p = 0 \) (solid curve) and \( \Theta_p = \pi/4 \) (dotted curve). Here \( \omega_k = 0.8 \omega_p \) and \( k = (0, 1) \). The corresponding integrated optical conductivities (in units of \( \sigma_0 = e^2/(4h) \)) as a function of \( \log_{10} \zeta \). As expected, both \( \sigma_{xx}^{(0)} \) and \( \sigma_{xx}^{(1)} \) display linear response behaviour (\( \rightarrow \sigma_0 \)) as \( \zeta \rightarrow 0 \). However, the newly generated \( \sigma_{xx}^{(-1)} \) is finite only after the onset of the nonlinear response regime (\( \xi \approx 1 \)). (c) The polarization angle dependence of longitudinal conductivities for \( \xi = 1 \). Other parameters are: \( \omega_p = 5 \times 10^{14}s^{-1}, \gamma_1 = 1 \times 10^{12}s^{-1}, \gamma_2 = 5 \times 10^{13}s^{-1} \) and \( \mu = 0 \).

on the amplitude and polarization of the incident pump beam.

The optical conductivity at different frequencies can now be obtained via the calculation of the charge current response: \( J = \sum_k \text{Trace}(\rho_k M_k) \), where \( M_k/e \) is the effective velocity operator. The corresponding optical conductivity matrix can be expressed as a Brillouin zone sum of the momentum resolved conductivity matrix:

\[
\sigma^{(i)} = (2\pi)^{-d} \int_{BZ} \frac{dk}{4\pi} \sigma^{(i)}_k, \quad \text{with } i = 0, \pm 1 \text{ and } d \text{ denoting the dimensionality of the system.}
\]

The momentum resolved optical conductivity matrix corresponding to the pump frequency \( \omega_p \) is given by

\[
\sigma_k^{(0)} = \frac{i\gamma_2}{\hbar \omega_p} M_k^{cc} \otimes M_k^{cv} \omega_k - \omega_p \]

Here, \( \omega' = \omega_p + i\gamma_2 \) and \( \otimes \) denotes the outer product of the optical matrix element vectors. The momentum resolved optical conductivity corresponding to the probe frequency (\( \omega_p + \omega_s \)) sideband is

\[
\sigma_k^{(1)} = \sigma_k^{(0)} \left[ 1 + \frac{|E_p \cdot M_k^{cc}|^2 \xi_k}{|\omega_k - (\omega_p + \omega_s)| [\omega_k - \omega_p']^{-1}} \right].
\]
setup. Thus $R$, $\Phi$ and $\Theta_{\text{Kerr}}$ can be probed as a function of the probe laser power and polarization angle (see Fig. 1). The dependence of the $s$ and $p$ components of the reflection coefficients on the respective optical conductivities in graphene are tabulated in Table I [39].

To compare the reflection amplitude and phase of the sideband [42] with that of the pump beam, we define the following:

$$
\frac{R_{ss}^{(\lambda)}}{R_{ss}^{(0)}} = \frac{|r_{ss}^{(\lambda)}|^2}{|r_{ss}^{(0)}|^2}, \quad \tan \Phi^{(\lambda)} = \frac{\text{Im}[r_{ss}^{(\lambda)}]}{\text{Re}[r_{ss}^{(\lambda)}]},
$$

where $\lambda = \pm 1$ for reflectance measured at the sideband frequencies $\omega_p \pm \omega_s$. The dependence of the ratios of the reflectance and $\Phi$ defined in Eq. 13, is shown in Fig. 3 as a function of the pump beam intensity ($\propto \zeta^2$). Clearly the $\omega_{\pm 1}$ sideband response at manifests only in the non-linear regime of $\zeta \approx 1$. In the optical regime (say $\omega_p = 5 \times 10^4 \text{s}^{-1}$), the estimated damping constants in graphene [30] are $\gamma_1 \approx 10^{12} \text{s}^{-1}$, $\gamma_2 \approx 5 \times 10^{13} \text{s}^{-1}$. Using these values, the $\zeta \approx 1$ condition in graphene corresponds to a CW laser intensity $\approx 10^9 \text{Wcm}^{-2}$, which is reasonable [3]. Furthermore, at reasonable CW powers we also observe non-linear phase shifts in excess of $\pi/2$ for the new sideband. Such large non-linear phase shifts are of great interest in the switching applications in THz and optical domains. The polarization angle $\Theta_p$ dependence of the reflection probability and its phase is shown in Fig. 4(a)-(b).

Non-linear optical response in graphene also generates a finite $\sigma_{xy}$, which in turn leads to Kerr rotation (polarization rotation of the reflected beam) [35, 38]. The Kerr rotation angle for $s$- and $p$- polarized incident pump beam is given by [35, 38],

$$
\theta^{(\lambda)}_{s/p\text{Kerr}} = \frac{1}{2} \tan^{-1} \left( \frac{2\text{Re}[\chi^{(\lambda)}_{s/p\text{Kerr}}]}{1 - |\chi^{(\lambda)}_{s/p\text{Kerr}}|^2} \right),
$$

where $\chi_{s/p\text{Kerr}}$ can be expressed in terms of the reflection coefficients (see Table I). The variation of the Kerr angle for the $s$ and $p$ components of pump, probe and the new sideband beam as a function of $\Theta_p$ is shown in Fig. 4(c)-(d). The polarization rotation of the $\omega_{\pm 1}$ sideband seems to be significantly large and different from that corresponding to the pump and probe frequencies.

In summary, we predict generation of a new modulated optical sideband in graphene in presence of a CW frequency modulated pump-probe setup. Physically, the ‘shuffling’ of the inter-band coherence due to interference of the pump and the probe results in the generated sideband that carries unique signature of the third order non-linear response in graphene. Experimentally, this manifests in the polarization, reflectivity, and in the phase of the reflection coefficient (see Fig. 3) at the sideband frequencies. In particular, the peak of the sideband gain occurs at a threshold, characterized by a single parameter $\zeta$ set by system decay rates and the pump power. A careful characterization of generated sideband gain can thereby provide a direct method of characterizing non-linear response of two-band systems with CW fields, in contrast to traditional, technologically involved time domain measurements. It also suggests a range of applications that include switching of frequency sidebands using non-linear phase shifts and generation of frequency combs.

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