Negative dynamic Drude conductivity in pumped graphene

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We theoretically reveal a new amplification mechanism of terahertz and far-infrared radiation in graphene under interband pumping. It is enabled by indirect interband electron transitions, where in the photon emission is preceded or followed by disorder scattering. The emerging contribution to the optical conductivity, which we call the interband Drude conductivity, is negative for photon energies below the double quasi-Fermi energy of the pumped carriers. Moreover, for Gaussian correlated disorder, the real part of the net Drude conductivity becomes negative, whereas the radiation amplification by a single graphene sheet substantially exceeds 2.3%. This effect enables terahertz lasing in disordered graphene.

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graphene under interband pumping has attracted considerable interest recently.1) Studies of carrier interaction with intense electromagnetic fields2–4) are ultimately aimed at the creation of novel graphene-based optoelectronic devices, such as ultrafast photodetectors, modulators, and radiation sources.5–7) The emergence of population inversion8–11) and negative interband dynamic conductivity in pumped graphene12) enables the coherent amplification of radiation, particularly in the terahertz (THz) range. Experimental observations of such amplification13,14) support the concept of graphene-based THz lasers.15,16) Their full-scale realization faces a number of challenges, however. First, the coefficient of the interband amplification by a clean layer of pumped graphene cannot exceed 2.3%,12) which is inseparably linked with the universal optical conductivity of graphene.17,18) Second, the radiation amplification associated with the direct interband electron transitions competes with the intraband Drude absorption.19) The problem of the Drude absorption is crucial for most semiconductor lasers operating at low frequencies ω < 10^6 Hz because its rate scales as 1/ω^2.

Considering the optical conductivity of direct-gap semiconductors, one typically accounts only for direct interband and indirect (scattering-assisted) intraband electron transitions (Fig. 1). However, there also exist indirect interband transitions, in which photon absorption (or emission) is followed (or preceded) by disorder (impurity or phonon) scattering (Fig. 1). In direct-gap materials, they are generally not considered, because they are next-order processes compared to the direct interband transitions. In semiconductors possessing a band gap EG, indirect interband transitions are also less probable than indirect intraband ones. The former can appear only at frequencies ω > EG/ℏ, at which the Drude factor decreases as 1/ω^2. This is not the case for gapless graphene, where it is reasonable to give equal consideration to indirect intra- and interband transitions.

In this letter, we show that certain sources of scattering (namely, Gaussian correlated disorder) can result in the dynamic interband Drude conductivity exceeding the intraband one. Thus, this type of disorder can raise the optical gain of a single sheet of pumped graphene far above the “clean” limit of 2.3%. This resolves the problem of the Drude absorption in graphene-based THz lasers.

We consider electrons in a graphene layer interacting with an in-plane electromagnetic field described by a vector potential A = A_0 cos ω t and random scattering potential V_S(r). The Hamiltonian of the system is

$$\hat{H} = \hat{H}_0 + \hat{V}_F + \hat{V}_S(r),$$  \hspace{1cm} (1)

$$\hat{H}_0 = v_0 (\sigma \cdot \hat{p}),$$ \hspace{1cm} (2)

$$\hat{V}_F = -\frac{e v_0}{c} \sigma A_0 \cos \omega t,$$ \hspace{1cm} (3)

$$\hat{V}_S(r) = \sum_{\mathbf{r}_i} V_0 (\mathbf{r} - \mathbf{r}_i).$$ \hspace{1cm} (4)

Here $\sigma = \{\sigma_x, \sigma_y\}$ is the set of Pauli matrices, $v_0 \approx 10^6$ m/s is the characteristic velocity of quasiparticles in graphene, $\hat{p}$ is the two-dimensional momentum operator, and $V_0 (\mathbf{r} - \mathbf{r}_i)$ is the potential of a single scattering center located at $\mathbf{r}_i$. The eigenstates of $\hat{H}_0$ represent quasiparticles belonging to the conduction ($\lambda = +1$) and valence ($\lambda = -1$) bands, $|\mathbf{p} \lambda \rangle = e^{i\mathbf{p} \cdot \mathbf{r}} / \sqrt{2}$, where $\theta_p$ is the angle between the momentum $\mathbf{p}$ and the x-axis.

The electron-field interaction causes the direct interband transitions allowed in the first-order perturbation theory. The corresponding real part of the interband conductivity, Re[\sigma_{int}], can be obtained using, for example, the Fermi golden rule, with the following result:

$$\operatorname{Re}[\sigma_{int}] = \sigma_0 \left[ f_s \left( -\frac{\hbar \omega}{2} \right) - f_c \left( \frac{\hbar \omega}{2} \right) \right].$$  \hspace{1cm} (5)

Here $\sigma_0 = e^2/4\hbar$ is the universal optical conductivity of clean graphene, and $f_s(\epsilon)$ and $f_c(\epsilon)$ are the electron distribution functions in the valence and conduction bands, respectively. Below we assume them to be quasi-equilibrium Fermi...
functions, \( f_\lambda(e) = \{1 + \exp[e - \mu_\lambda/T]\}^{-1} \). In equilibrium, \( \mu_c = \mu_v = e\nu \geq 0 \), whereas for symmetrical pumping \( \mu_c = -\mu_v = e\nu \), where \( e\nu \) is the (quasi) Fermi energy. Then, one readily notes that the conductivity of the pumped graphene due to direct interband transitions is negative at frequencies \( \hbar \omega < 2e\nu \). \(^{(2)}\)

The presence of the scattering potential \( \tilde{V}_S(r) \) leads to indirect electron transitions and, thus, intraband Drude absorption. The corresponding electron transition amplitude \( \Delta V^{\text{pp}}_{\text{pp}} \) is readily obtained from the second-order perturbation theory considering (3) and (4) as perturbations:

\[
\Delta V^{\text{pp}}_{\text{pp}} = \frac{e}{2c} \frac{(v_p - v_p', \mathbf{A}_0)}{\hbar \omega} (\mathbf{p}' \cdot \tilde{V}_S \mathbf{p}). \tag{6}
\]

A similar expression can be written for the electron transitions in the valence band.

However, one can also consider a second-order process in which the photon absorption/emission is followed or preceded by interband electron scattering. Its amplitude is similarly found from the second-order perturbation theory, which yields

\[
\Delta V^{\text{pp}}_{\text{pp}} = \frac{e}{2c} \frac{(v_p + v_p', \mathbf{A}_0)}{\hbar \omega} (\mathbf{p}' \cdot \tilde{V}_S \mathbf{p}). \tag{7}
\]

Applying the Fermi golden rule for these indirect inter- and intraband transitions, one can find the real part of the dynamic conductivity. We denote it by the superscript “D” (Drude) to distinguish it from the interband conductivity due to the direct transitions (5):

\[
\text{Re}[\sigma^{\text{D}}_{\text{intra}}] = \frac{2\pi e^2}{\hbar^2} \sum_{p,p',\lambda} \left[ f_\lambda(e_p) - f_\lambda(e_{p'}) \right] \delta(e_p + \hbar \omega - e_{p'})[\mathbf{p}' \cdot \tilde{V}_S \mathbf{p}]^2 (v_p - v_{p'})^2, \tag{8}
\]

\[
\text{Re}[\sigma^{\text{D}}_{\text{inter}}] = \frac{2\pi e^2}{\hbar^2} \sum_{p,p'} \left[ f_\lambda(e_p) - f_\lambda(e_{p'}) \right] \delta(h\omega - e_p - e_{p'})[\mathbf{p}' \cdot \tilde{V}_S \mathbf{p}]^2 (v_p + v_{p'})^2.
\]

Here \( v_p = v_p p / p \) is the quasiparticle velocity, and \( g = 4 \) is the spin-valley degeneracy factor. In the classical limit \( \hbar \omega \ll (k_B T, e\nu) \), Eq. (8) naturally reduces to the conductivity obtained using the Boltzmann theory. On the other hand, Eq. (8) is valid until \( \omega \gg \tau_p^{-1} \), where \( \tau_p \) is the momentum relaxation time. We will focus on this frequency range. The expression (9) cannot be obtained from the simple kinetic equation because it involves interband transitions. This is the central equation of this letter. Further, we will analyze it and compare the magnitudes of \( \text{Re}[\sigma^{\text{D}}_{\text{intra}}] \) and \( \text{Re}[\sigma^{\text{D}}_{\text{inter}}] \).

A key difference between the inter- and intraband Drude conductivities can be seen from the energy conservation law. For the interband energy conservation in Eq. (9) to be fulfilled, the transmitted momentum \( \mathbf{q} = \mathbf{p}' - \mathbf{p} \) should be small; namely, \( q \ll \hbar \omega \). In the intraband process (8), in contrast, \( q \gg \hbar \omega \) is required. Therefore, indirect interband transitions are favored by scattering potentials with large Fourier components \( V_\mathbf{q} \) at small \( q \). For indirect interband transitions to dominate over intraband ones, the Fourier components of the scattering potential should either be singular at \( q \to 0 \) or decrease abruptly as \( q \) increases.

Consider scattering by random defects with Gaussian correlations, such that the potential energy correlator is \( \left< V(\mathbf{r}) V(\mathbf{r}') \right> = V^2 \exp[-|\mathbf{r} - \mathbf{r}'|^2 / \ell_c^2] \), where \( \ell_c \) is the correlation length, and \( V^2 \) is the average square of the scattering potential. The squared modulus of the scattering matrix element appearing in the Fermi golden rule is calculated as\(^{(24)}\)

\[
||\langle \mathbf{p}' \lambda'| \tilde{V}_S |\mathbf{p}, \lambda \rangle ||^2 = \pi \hbar^2 V^2 e^{-q l_c^2/2} \frac{1 + \lambda' l_c \cos \theta_{\text{pp}}}{2}. \tag{10}
\]

Passing to the elliptic coordinates in Eq. (9) and evaluating the integrals, one obtains the following relation for the interband Drude conductivity:

\[
\text{Re} \left[ \sigma^{\text{D}}_{\text{inter}} \right] = \frac{V^2 \ell_c^2}{\hbar^2 g v_0} \left[ f_\lambda \left( -\frac{\hbar \omega}{2} \right) - f_\lambda \left( -\frac{\hbar \omega}{2} \right) \right] J_1 \left( \frac{\omega}{2 v_0} \right), \tag{11}
\]

where \( J_1(x) \) is the dimensionless integral

\[
J_1(x) = \int_0^1 dt e^{-\alpha^2 t^2} \sqrt{1 - t^2} \left( 1 - \sqrt{1 - t^2} \right). \tag{12}
\]

with the following asymptotic values

\[
J_1(x) \approx \begin{cases} 
1/6 & x \ll 1, \\
1 - x^2/2 & x \gg 1.
\end{cases} \tag{13}
\]

Considering the intraband Drude conductivity associated with the correlated disorder, one can show that the energy–momentum restriction \( q v_0 > \hbar \omega \) yields a small factor \( \exp[-(\omega v_c / 2v_0)^2] \) in the expression for \( \text{Re}[\sigma^{\text{D}}_{\text{intra}}] \). This results in an abrupt drop beyond \( 1/\alpha^2 \) in the intraband Drude absorption with increasing frequency. There is no such small factor in the expression for the interband Drude absorption. As a result, the net Drude conductivity \( \text{Re}[\sigma^{\text{D}}_{\text{intra}} + \sigma^{\text{D}}_{\text{inter}}] \) can become negative in some frequency range below \( 2e\nu / \hbar \). This is illustrated in Fig. 2, where we plot the contributions of the inter- and intraband scattering processes to the Drude conductivity separately.

The minimum of the net Drude conductivity shifts to lower values with increasing correlation length \( \ell_c \). This is mainly due to an abrupt drop in the “normal” Drude absorption \( \text{Re}[\sigma^{\text{D}}_{\text{intra}}] \propto e^{-q v_0 / \ell_c} \). The minimum is also shifted toward smaller frequencies as \( \ell_c \) increases (roughly, it is achieved at \( \omega \sim v_0 / \ell_c \)). These two trends are illustrated in Fig. 3, where we plot the net Drude conductivity vs the frequency at different correlation lengths. The value of \( \overline{V^2} \) does not affect the presence of negative Drude conductivity (until \( \omega \gg \tau_p^{-1} \)) and determines only its magnitude. More-
over, the dip in the frequency dependence of the conductivity becomes deeper as the strength of the disorder potential increase. Thus, a sample with Gaussian correlated disorder amplifies the radiation better as it becomes “dirtier”.

One can also find the contributions to the interband Drude conductivity associated with scattering by random uncorrelated charged impurities with an average density $n_i$. The result is as follows:

$$\text{Re} \left[ \frac{\sigma^D_{\text{inter}}}{\sigma^D} \right] \approx \frac{2 \pi c^2}{3 q_S^3} \left[ f_v \left( \frac{-\hbar \omega}{v} \right) - f_c \left( \frac{-\hbar \omega}{v} \right) \right] \times \left[ 1 - \frac{15\pi - 32}{10} \frac{\hbar \omega}{q_S v_0} \right]$$

where $q_S = 8\alpha_e (k_B T/v_0) \ln(1 + e^{\epsilon_{F}/k_B T})$ is the Thomas–Fermi momentum, $\alpha_e = e^2/(\hbar v_0 k_B)$ is the coupling constant, and $\kappa_0$ is the background dielectric constant.

A direct numerical comparison of $\text{Re} [\sigma^D_{\text{inter}}]$ and $\text{Re} [\sigma^D_{\text{extra}}]$ for the uncorrelated impurities shows that the presence of the interband processes reduces the value of Drude absorption by a maximum of ~5% in pumped graphene for the typical values $\epsilon_{F} = 50$ meV and $\kappa_0 = 5$ at room temperature. Hence, one should not expect sufficient renormalization of the radiation absorption due to the interband transitions for such scattering potentials. This conclusion is valid for any scattering potential weakly depending on $q$ (acoustic phonon scattering is another example). In those cases, the phase space restriction $q < \hbar \omega/v_0$ results in the relative smallness of the interband Drude conductivity.

From a practical viewpoint, it is important to reveal the contribution of the considered interband Drude conductivity to the net dynamic conductivity of pumped graphene. The optical conductivity of clean graphene incorporates the direct interband conductivity (5) and Drude conductivity due to carrier–phonon scattering and carrier–carrier scattering. We denote the latter by $\sigma_{\text{ph}}$ and $\sigma_{\text{cc}}$, respectively. It was shown that in the high-frequency range $\omega \gg \tau_0^{-1}$, one typically has $\sigma_{\text{cc}} \gg \sigma_{\text{ph}}$. In samples with correlated disorder, the contributions (9) and (8) are added to the conductivity of clean graphene.

Figure 4 shows examples of the net dynamic conductivity in such samples calculated for different correlation lengths $\ell_c$. Even in the presence of strong carrier–carrier scattering, the interband limit $-\sigma_0$ on the negative dynamic conductivity can be surpassed because of indirect interband transitions.

In Fig. 5, we compare the dynamic conductivities of a clean pumped graphene layer and a graphene layer with Gaussian correlated impurities at different frequencies and quasi-Fermi energies. The presence of the interband Drude transitions significantly broadens the domain of negative conductivity, particularly toward lower frequencies. The negative dynamic conductivity below $-\sigma_0$ indicates the possibility of optical amplification above 2.3% by a single layer of pumped graphene.
In conclusion, we studied the photon emission/absorption processes in graphene enabled by indirect interband transitions. Scattering by Gaussian correlated disorder results in the interband Drude conductivity exceeding the “normal” intraband conductivity. Under population inversion, this leads to a negative net Drude conductivity and radiation amplification above 2.3% by a single graphene sheet. The fabrication of artificial correlated disorder, for example, by selective absorption of atoms \(^{26}\) or nanoperforation,\(^{27}\) opens a path to novel graphene-based lasing structures.

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