Effects of carrier-carrier scattering on population inversion in graphene under pulse photoexcitation

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Abstract. We study the carrier relaxation dynamics in intrinsic graphene after pulse photoexcitation and reveal effects of intraband carrier-carrier scattering on population inversion in the terahertz region, by conducting simulation based on the quasi-classical Boltzmann equation. It is demonstrated that by changing the dielectric constant of the surrounding materials the rate of carrier-carrier scattering can be controlled and the relaxation dynamics differs for cases with low and high dielectric constants. It is also found that the Pauli blocking of photogeneration in case of the pulse photoexcitation causes decrease in the photocarrier concentration and thus weakening of population inversion with higher dielectric constant.

1. Introduction
Device applications of graphene utilizing its unique electronic/optical properties have been investigated extensively. In particular, We have studied terahertz (THz) lasers and active THz plasmonic devices [1-3] based on interband population inversion in graphene at room temperature, which take advantage of the high carrier mobility and gapless linear dispersion of graphene. The zero bandgap enables population inversion in very low energy region, i.e., down to the THz region. Several pumping schemes such as optical pumping and current injection have been proposed [2-5]. Experimental observations of light amplification by stimulation emission from graphene under pulse photoexcitation have already been reported in the THz [7] and near-infrared [6] regions.

For any pumping schemes, the carrier-carrier (CC) scattering is anticipated to play a crucial role in the achievement of population inversion. Very fast quasi-equilibration of carriers by the CC scattering in comparison with cooling by the optical-phonon (OP) emission hinders population inversion with optical pumping [8, 9]. The rise of the carrier temperature due to the quasi-equilibrium causes less accumulation of carriers in the the THz region. It has been shown in Ref. [8] that, if the time scale of cooling due to the OP emission is faster than the quasi-equilibrium, the threshold pumping intensity for the population inversion is much smaller than the case of the fast quasi-equilibration.

The characteristic time scale of the CC scattering can be changed, relatively to other mechanisms such as OP scattering, via the choice of surrounding dielectric materials ($\tau_{cc} \propto \epsilon^2$) and/or via the presence of gate electrodes. They can be utilized for either suppressing or enhancing the effect of the CC scattering and, in turn, for optimizing device performances. To fully reveal the role of the CC scattering on operation principles and its controllability via...
dielectric materials and/or gate electrodes, numerical simulation taking into account the CC scattering is inevitable. In this paper, we develop a kinetic transport model with the CC scattering based on a deterministic solution approach for the semi-classical Boltzmann equation and on the direct evaluation of the collision integral for the CC scattering. Using the model, we conduct transient simulation of carrier relaxation dynamics in intrinsic graphene after femtosecond photoexcitation. We demonstrate that with high dielectric constant of the surrounding materials the suppression of CC scattering induces not only cooling by OP emission but also the Pauli blocking of photogeneration of carriers, so that the threshold of pulse fluence for population inversion becomes larger with higher dielectric constant in case of pulse photoexcitation.

2. Simulation model
For a first step towards the development of a device simulation model for graphene THz devices with inclusion of the CC scattering, we start from a spatially homogeneous case (its extension to the inhomogeneous case is straightforward, although the former is computationally more expensive). Our model is based on semi-classical Boltzmann equations for electrons and holes:

$$\frac{\partial f_{cp}}{\partial t} = \sum_{c'=\epsilon,h} J_{cc'} + \sum_i J_{ci},$$  

where $f_{cp}$ is the distribution function for carrier type $c$ ($\epsilon$ and $h$ for electrons and holes, respectively). Here, $J_{cc'}$ is the collision integral for $c$-$c'$ scattering:

$$J_{cc'}(p) = \frac{4}{(2\pi\hbar)^3} \int d^3 p_1 \int d^2 p' W_{cc'}(p, p_1; p', p'_1) \times \left[ f_{cp} f_{cp'1}(1 - f_{cp}) (1 - f_{cp}p_1) - f_{cp} f_{c'p_1}(1 - f_{c'p_1})(1 - f_{c'p'_1}) \right]$$

with $p'_1 = p + p_1 - p'$ and $W_{cc'}$ being the Coulomb scattering probability. $W_{cc'}$ contains a delta function representing the energy conservation, $\delta(p + p_1 - p' - p'_1)$, so that Eq. (2) reduces to a triple integral. For simplicity, we consider the static Thomas-Fermi screening for the Coulomb potential (effects of the dynamic screening shall be briefly discussed at the end of the next section).

For other scattering mechanisms which are represented by $J_{ci}$, we consider the intraband and interband OP emission/absorption, $J_{c, op}$, as well as photogeneration, $J_{c, ph}$. Here, we assume equilibrium optical phonons. For the photogeneration, we assume a Gaussian pulse with central wavelength $\lambda$, pulse width $\Delta t$ (full-width at half-maximum), and pulse fluence $J$.

We developed a computer program that solves Eq. (1), first by evaluating $J_{cc'}$ at each point $(p_i, \theta_j)$ in parallel and then by applying the third-order TVD Runge-Kutta scheme [10] for the time-step update.

3. Results and discussions
We conducted transient simulation of carrier relaxation dynamics in intrinsic graphene after femtosecond photoexcitation. As parameters for the Gaussian pulse we fixed $\lambda = 1.55 \, \mu m$ and $\Delta t = 80 \, fs$, and we set the initial distribution functions of electrons and holes with temperature $T = 300 \, K$ and zero Fermi level (i.e., intrinsic graphene). We varied the dielectric constant of the surrounding material, $\epsilon$, to control the time scale of the CC scattering and thus to reveal its effect on population inversion.

Figure 1 shows snapshots of the electron distribution function with relatively low dielectric constant $\epsilon = (\epsilon_{air} + \epsilon_{SiO_2})/2 = 2.75$ and the pulse fluence $J = 6 \, \mu J/cm^2$. The angle $\theta$ is taken
Figure 1. Snapshots of the electron distribution function with $\epsilon = 2.75$ and $J = 6 \mu J/cm^2$ that show distinct states of relaxation dynamics: (a) initial distribution, (b) photoexcitation at the peak of the optical pulse, (c) quasi-equilibration in the energy direction, (d) quasi-equilibration in the angular direction, (e) cooling by OP emission and population inversion in the THz region ($< 40$ meV), and (f) relaxation of population inversion by recombination due to OP emission. Note that the condition of population inversion is $f > 1/2$.

with respect to the polarization of the pulse. Figure 1 clearly illustrates distinct states of carrier relaxation dynamics after the pulse photoexcitation; the photogenerated carriers together with the initial carriers are quasi-equilibrated in the energy direction in the phase space within a few hundred fs (Fig. 1(c)), followed by quasi-equilibration in the angular direction (d), then they are cooled down by OP emission after a few ps and the population inversion in the THz region is achieved (e), and the recombination takes place in a few tens of ps (f). Note that the condition of population inversion for intrinsic graphene is $f > 1/2$. The difference between time scales of quasi-equilibration in energy and angular directions originates from the small-angle nature of the Coulomb scattering.
Figure 2. Snapshots of the electron distribution function with $\epsilon = 40$ and $J = 8 \, \mu J/cm^2$: (a) photoexcitation before the peak of the optical pulse, (b) photoexcitation after the peak of the optical pulse, exhibiting a wide peak around $\epsilon = 0.2 - 0.24$ eV associated with single OP emission, (c) quasi-equilibration in the energy direction, (d) partial quasi-equilibration in the angular direction.

The time dependence of distribution in Fig. 1 after quasi-equilibration in both directions is almost identical with that calculated in Ref. [9], which is based on the assumption that the distribution is always quasi-equilibrium even during the photoexcitation. This is because the time scale of the OP emission (a few hundred fs) is slower than the completion time of quasi-equilibration, with the low value of the dielectric constant, $\epsilon = 2.75$.

The picture of carrier relaxation dynamics changes when the dielectric constant is high and the CC scattering is suppressed. Figure 2 shows snapshots of the electron distribution function for very high dielectric constant $\epsilon = 40$ and the pulse fluence $J = 8 \, \mu J/cm^2$, before the completion of quasi-equilibration. In Fig. 2(b), it is seen that a small, wide peak around $\epsilon = 0.2 - 0.24$ eV appears. The energy difference of this peak from the photogeneration peak ($\epsilon = 0.4$ eV) corresponds to the energies of optical phonons (0.16 eV for $K$ optical phonons and 0.2 eV for $\Gamma$ optical phonons), indicating that it originates from single OP emission by photogenerated carriers. It is also worth mentioning that quasi-equilibration in the angular direction is done both by CC scattering and OP emission (Fig. 2(c)), although a small deviation of the distribution function by angle is kept for a long time in the low energy region (over 1 ps, see Fig. 2(d)) due to the prohibition of OP emission by energy conservation low.

Figure 2(b) also shows another distinct feature; a very large value of the photogeneration peak even after the peak of Gaussian pulse. This means that the large fraction of photogenerated carriers stay longer at the same energy than the time scale of the photogeneration (80 fs). It causes the Pauli blocking of photogeneration and, in turn, the decrease in the photocarrier.
Concentration (see Fig. 3(a)). As shown in Fig. 3(b), the effect of Pauli blocking on population inversion is stronger than that of cooling by OP emission, so that the population inversion achieved by a fixed pulse fluence becomes weaker for higher dielectric constant.

Contrary to the case of pulse excitation studied here, however, the lowering of the threshold pumping intensity with higher dielectric constant is still expected in the case of CW pumping, as the effect of Pauli blocking is negligibly small for the pumping intensity necessary for the THz population inversion. Besides, the static screening of the Coulomb potential assumed in this work overestimates the time scale of quasi-equilibration by the CC scattering, as it ignores the dynamic screening for the high-energy photogenerated carriers. Especially, it is pointed out that the CC scattering with small-angle change is suppressed by the dynamic screening [11]. It should result in the further suppression of carrier heating and thus lowering of the threshold. For quantitative analysis of this, it is very crucial to include the dynamic screening in the simulation model.

4. Conclusion
We developed a simulation model for graphene devices taking into account the CC scattering. Based on the model, we conducted a transient simulation of carrier relaxation dynamics in intrinsic graphene after femtosecond photoexcitation. It was revealed that the relaxation dynamics can be changed by the dielectric constant of the surrounding materials. With low dielectric constant, quasi-equilibration takes place much faster than cooling by OP emission. With sufficiently high dielectric constant, on the other hand, we observed a peak in the distribution function before quasi-equilibration, which is attributed to single OP emission from the photogeneration peak. Simultaneously, it was found that the Pauli blocking of photogeneration in case of the pulse photoexcitation weakens population inversion with higher dielectric constant.

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