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Study of Hot Carriers in Optically Pumped Graphene

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1. Introduction

Graphene, a two-dimensional solid-state material, has attracted much attention for wide variety of device applications due to its exceptional electronic and optical properties. Not only the transistor applications but optoelectronic devices such as THz lasers [1,2] and photodetectors [3,4] have been investigated. In Refs. [1,2], we demonstrated that the population inversion can occur in optically pumped graphene at THz/far-infrared range of frequency and hence the lasing at such a range is possible, utilizing the gapless energy spectrum and relatively high optical phonon (OP) energy in graphene.

In previous works [1,2], we assumed that the carrier-carrier (CC) scattering is effective only after photocarriers experience the cascade emission of OPs and fall into near the Dirac point. This assumption is clear at low temperatures and with relatively weak pumping, where the carrier concentration is sufficiently low so that the OP emission precedes the CC scattering. At room temperature, however, the thermal carrier concentration reaches 10^{11} cm^{-2} and upon strong pumping the CC scattering might be frequent enough to broaden the absorption spectrum of the pumping light or might even thermalize the carriers before the OP emissions, creating hot carriers. In such situations, conditions of the population inversion can change drastically.

In this paper, we extend the hot-carrier model developed in Ref. [2] for two limiting cases, (1) where the OP scattering is dominant and carriers take a quasi-equilibrium by the CC scattering after the cascade emission of OPs, and (2) where the CC scattering is dominant and carriers which are always in a quasi-equilibrium suffer energy relaxation and recombination via the OP scattering (latter only via interband OPs). We also take into account both the intra and interband OPs [5,6]. Using the model developed, we consider the cw pumping of intrinsic graphene in the cases (1) and (2) and investigate the possibility of population inversion. We show that sufficiently strong pumping achieves the population inversion in both cases, with the carrier cooling in (1) and with very high carrier temperature in (2).

2. Model

Under optical pumping with photon energy $\hbar\Omega$, photoelectrons and holes are generated in the intrinsic graphene. Their energy is relaxed dominantly by the intra and interband OP emission and they are recombined dominantly via the interband OP emission [6]. For both cases under consideration, the carrier distribution (equivalent electron and hole distributions) is governed by the following balance equations for the total energy and concentration of carriers:

$$\frac{2\pi}{\sqrt{\epsilon}}\frac{\alpha I_\Omega}{\hbar\Omega} = \frac{2}{\pi\hbar^2 v_W^2}\sum_{i=\Gamma,\mathrm{K}}\int_0^\infty \varepsilon d\varepsilon \times\left[\frac{f_\varepsilon f_{\hbar\omega_i-\varepsilon}}{\tau^{(i)}_{\mathrm{inter,e}}} - \frac{(1-f_{\hbar\omega_i-\varepsilon})(1-f_\varepsilon)}{\tau^{(i)}_{\mathrm{inter,a}}}\right], \quad (1)$$

$$\frac{2\pi}{\sqrt{\epsilon}}\frac{\alpha I_\Omega}{\hbar\Omega}E_{\mathrm{eff}} = \frac{2}{\pi\hbar^2 v_W^2}\sum_{i=\Gamma,\mathrm{K}}\int_0^\infty \varepsilon d\varepsilon \times\left[\varepsilon\frac{f_\varepsilon f_{\hbar\omega_i-\varepsilon}}{\tau^{(i)}_{\mathrm{inter,e}}} - \varepsilon\frac{(1-f_{\hbar\omega_i-\varepsilon})(1-f_\varepsilon)}{\tau^{(i)}_{\mathrm{inter,a}}} + \hbar\omega_i\frac{f_\varepsilon(1-f_{\varepsilon-\hbar\omega_i})}{\tau^{(i)}_{\mathrm{intra,e}}} - \hbar\omega_i\frac{f_\varepsilon(1-f_{\varepsilon+\hbar\omega_i})}{\tau^{(i)}_{\mathrm{intra,a}}}\right], \quad (2)$$

where $\alpha = e^2/\hbar c$, I_Ω is the pumping intensity, $\hbar\omega_\Gamma = 198$ meV and $\hbar\omega_{\mathrm{K}} = 161$ meV are the intra and intervalley phonon energies [5], τ is the energy-dependent scattering times of intra/interband OP emission/absorption (the type is denoted by its subscripts), f_ε is the carrier distribution function, and E_{eff} is the effective energy of a photogenerated carrier contributing to the thermalization, i.e., $E_{\mathrm{eff}} = \hbar\Omega/2$ in the case (2), and E_{eff} is the average energy of a carrier after the cascade emission of Γ and/or K OPs in the case (1).

Since carriers are thermalized by the CC scattering (after the cascade emission of intraband OPs in the case (1)), we can assume $f = 1/\{\exp[(\varepsilon-\varepsilon_F)/k_B T_e]+1\}$, where ε_F is the quasi-Fermi energy for carriers and T_e is the carrier temperature. Quantities ε_F and T_e can be found by solving Eqs. (1) and (2) numerically.

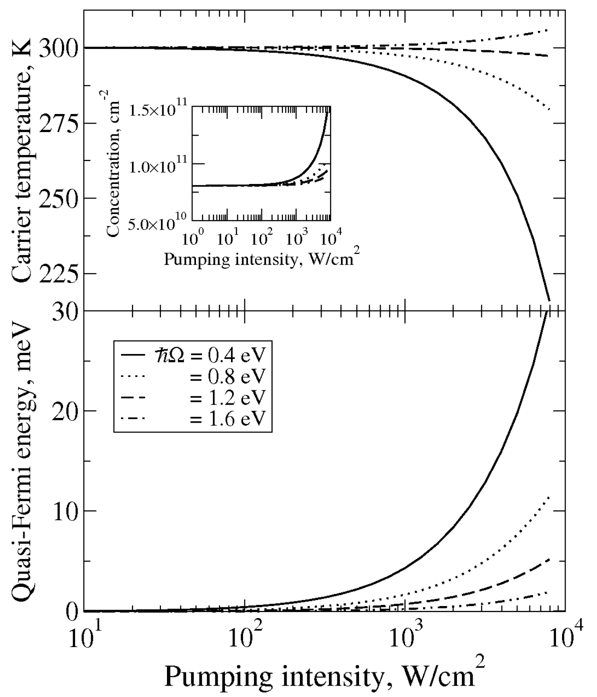

Figure 1: Carrier temperature and quasi-Fermi energy in the case (1) as a function of the pumping intensity with different pumping photon energies.

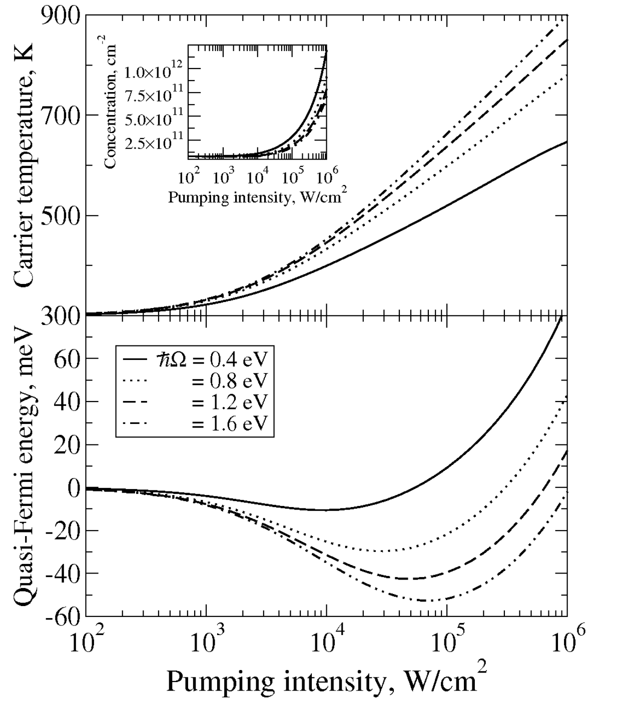

Figure 2: Carrier temperature and quasi-Fermi energy in the case (2) as a function of the pumping intensity with different pumping photon energies.

3. Results and Discussions

Figure 1 shows the carrier temperature and quasi-Fermi energy in the case (1). The range of pumping intensity is selected such that the pumping results in the total carrier concentration around 10^{11} cm^{-2} (see the inset in Fig. 1). It is clearly seen from Fig. 1 that the population inversion takes place for sufficiently strong pumping intensity as well as the carrier temperature becomes lower than the lattice temperature $T = 300$ K. The latter is achieved by imbalance between photogenerated carrier concentration and the effective carrier energy after the cascade emission of intraband OPs. It should be pointed out that the pumping intensity needed for the sufficient population inversion is higher than that estimated in the previous work [1,2] because the interband OP emission, which is taken into account in the model developed here, results in very short energy-relaxation/recombination times ($\approx$ 10 ps). Figure 2 shows the carrier temperature and quasi-Fermi energy in the case (2). Contrary to the case (1), the population inversion cannot be achieved until very strong pumping (over 10^5 W/cm^2) and the carrier temperature becomes very high.

In reality, the situation lies somewhere between the cases (1) and (2). It is determined by the interplay between the OP scattering and CC scattering. However, somewhat weak energy-transfer nature of the latter at high-energy region (where the photogeneration takes place) might slow down the thermalization process right after the photogeneration at not so high carrier concentartion (around 10^{11} cm^{-2}). In such a case, the picture of the cascade emission of OPs assumed in the case (1) might still hold, with a broadened spectrum of the pumping, and hence the population inversion is possible.

4. Conclusions

The hot-carrier distribution in optically pumped graphene was investigated with a theoretical model developed for two limitting cases (the OP-dominant and CC-dominant cases). It was shown that for both cases the population inversion is possible with sufficiently strong pumping.

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