Terahertz light amplification of stimulated emission of radiation in optically pumped graphene

T. Otsuji$^{1,4}$, S.A. Boubanga Tombet$^{1,4}$, S. Chan$^{2}$, T. Watanabe$^{1}$, A. Satou$^{1,4}$, M. Ryzhii$^{3,4}$ and V. Ryzhii$^{3,4}$

1 RIEC: Research Institute of Electrical Communication, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, 980-8577, Japan
2 NSF-PIRE Nano-Japan Program, University of Pennsylvania, 3451 Walnut Street, Philadelphia, PA 19104, USA
3 Computational Nano-Electronics Laboratory, University of Aizu, Ikki-machi, Aizu-Wakamatsu, 965-8580, Japan
4 JST-CREST, 5-3 Yonbanchou, Chiyoda-ku, Tokyo, 102-8666, Japan

Abstract

The gapless and linear energy spectra of electrons and holes in graphene lead to nontrivial features such as negative dynamic conductivity in the terahertz spectral range. This paper reviews recent advances in theoretical and experimental study on terahertz light amplification by stimulated emission of radiation in optically pumped graphene.

1. Introduction

When graphene is pumped with the infrared (IR) photon having an energy $h\Omega$, electrons/holes are photogenerated via interband transitions. When the photogenerated electrons and holes are heated in case of room temperature environment and/or strong pumping, collective excitations due to the carrier-carrier (CC) scattering, e.g., intraband plasmons should have a dominant play to perform an ultrafast carrier redistribution along the energy as shown in Fig. 1 [5, 6]. Then optical phonons (OPs) are emitted by carriers on the high-energy tail of the electron and hole distributions. This energy relaxation process accumulates the nonequilibrium carriers around the Dirac points as shown in Fig. 1. Due to a fast intraband relaxation (ps or less) and relatively slow interband recombination (>>1ps) of photoelectrons/holes, one can obtain the population inversion under a sufficiently high pumping intensity [3]. Due to the gapless symmetrical band structure of graphene, photon emissions over a wide THz frequency range are expected if the pumping IR photon energy is properly chosen.

2. Theoretical Model

We consider an intrinsic graphene under the optical pulse excitation in the case where the CC scattering is dominant and carriers always take quasi-equilibrium. We take into account both the intra and interband OPs [7, 8]. The carrier distribution (equivalent electron and hole distributions) is governed by the following equations for the total energy and concentration of carriers:

Fig. 1. Carrier relaxation/recombination dynamics in optically pumped graphene.
where $\Sigma$ and $E$ are the carrier concentration and energy density, $f_\varepsilon$ is the quasi-Fermi distribution, $\tau^{\pm}_{\text{inter}}$ and $\tau^{\pm}_{\text{intra}}$ are the inverses of the scattering rates for inter and intraband OPs ($i = \Gamma$ for OPs near the $\Gamma$ point with $\omega_\Gamma = 196$ meV, $i = K$ for OPs near the zone boundary with $\omega_\Gamma = 161$ meV, $+$ for absorption, and $-$ for emission). Time-dependent quasi-Fermi energy $\varepsilon_F$ and the carrier temperature $T_c$ are determined by these equations. Figure 2 shows the typical results for fs pulsed laser pumping with photon energy 0.8 eV [9]. It is clearly seen that $\varepsilon_F$ rapidly increases with cooling the carrier and it becomes positive when the pumping intensity exceeds a certain threshold level. This result proves the occurrence of the population inversion. After that, the recombination process follows more slowly (~10 ps).

3. Experimental Results and Discussion

We observed the carrier relaxation and recombination dynamics in optically pumped graphene using THz time-domain spectroscopy based on an optical pump/THz-and-optical-probe technique [10]. An exfoliated monolayer-graphene/SiO$_2$/Si sample is placed on the stage and a 0.12-mm-thick (100)-oriented CdTe crystal is placed on the sample, acting as a THz probe pulse emitter as well as an electrooptic sensor. A single 80-fs, 1550-nm fiber laser beam having 4-mW average power and 20-MHz repetition is split into two: one for optical pumping and generating the THz probe beam in the CdTe crystal, and one for optical probing. The pumping laser, which is linearly polarized, is simultaneously focused at normal incidence from the back surface on the graphene sample to induce population inversion and the CdTe to induce optical rectification and emission of THz pulse (the primary pulse marked with “(1)" in Fig 3). This THz beam reflecting back in part at the CdTe top surface stimulates the THz emission in graphene, which is electrooptically detected as a THz photon echo signal (the secondary pulse marked with “(2)” in Fig 3).

Figure 4 shows a typical temporal response under the maximal pumping intensity. The black/red curve is the response when the pumping beam is focused onto the sample with/without graphene. The second pulse, the THz photon echo signal, obtained with graphene is more intense compared with that obtained without graphene. This indicates the
graphene act as an amplifying medium. Figure 5 shows the emission spectra from graphene after normalization to the one without graphene. The inset in Fig. 5 shows the measured gain as a function of the pumping power. A threshold like behavior can be seen testifying the occurrence of the negative conductivity and the THz light amplification by stimulated emission of radiation.

Fig. 3. Experimental setup (left) and the scheme of coherent emission from graphene by an optical-pump/THz-probe technique (right). Time-resolved electric field intensity is electrooptically sampled by the probe beam throughout the CdTe sensor crystal in total reflection geometry. The CdTe also works as a THz probe beam source.

Fig. 4. Measured temporal profile. The secondary pulse is the THz photon echo transmitted and reflected through graphene.

Fig. 5. Normalized Fourier spectra and gain profile.
4. Conclusion

We have successfully observed coherent amplified stimulated THz emissions arising from the carrier relaxation/recombination dynamics of an exfoliated graphene. The results provide evidence of the occurrence of negative dynamic conductivity even at room temperature, which can potentially be applied to a new type of THz lasers.

Acknowledgments

This work is financially supported in part by JST-CREST, Japan, JSPS-BR(S), Japan, and NSF-PIRE Nano-Japan Program, USA.

References