## Stimulated Terahertz Emission from Optically Pumped Graphene and Its Threshold Behavior

Akira Satou<sup>1,3</sup>, Stephane Boubanga-Tombet<sup>1,3</sup>, Taiichi Otsuji<sup>1,3</sup>, and Victor Ryzhii<sup>2,3</sup>

<sup>1</sup>Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan <sup>2</sup>University of Aizu, Tsuruga, Ikki-machi, Aizu-Wakamatsu 965-8580, Japan <sup>3</sup>Japan Science and Technology Agency, Tokyo 107-0075, Japan <u>a-satou@riec.tohoku.ac.jp</u>

Graphene has attracted much attention for wide variety of device applications due to its exceptional electronic and optical properties. Recently, we have proposed THz lasers using optically pumped graphene [1-3]. The negative dynamic conductivity can occur at THz frequency due to the population inversion and hence the lasing is possible (see Fig. 1(a)). In previous work, we measured the carrier relaxation and recombination dynamics in optically pumped epitaxial graphene on silicon [4] and exfoliated graphene [5] using THz time-domain spectroscopy based on an optical pump/THz&optical probe technique. A pulsed optical beam was impinged to a graphene sample and a CdTe crystal placed on it. The THz probe pulse generated from CdTe stimulates the THz emission from the sample. The comparison of emission spectra for sample spots with/without graphene indicated that the coherent THz probe pulse passing through graphene is amplified by the stimulated emission from optically pumped graphene. In this paper, we study both experimentally and theoretically the threshold behavior of the THz stimulated emission from graphene for pumping intensity.

An exfoliated monolayer-graphene/SiO2/Si sample is prepared. 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. An 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, and one for optical probing. The pumping laser is simultaneously focused at normal incidence from the back surface onto the graphene (for optical pumping) and to the CdTe (for generating THz prove pulse marked with "1" in Fig. 1(b)). This THz proves beam reflecting back in part stimulates the THz emission in graphene, which is electrooptically detected as a THz photon echo signal (the secondary pulse marked with "2" in Fig. 1(b)). We changed the spot size of the pumping beam by mechanically shifting a focal lens. Fig. 1 (c) shows the emission spectra and measured gain as a function of the pumping intensity. The gain was calculated by dividing the peak intensity of the stimulated emission from the sample spot with graphene by that of the emission from the spot without graphene. It can be seen in Fig. 1(c) that the threshold intensity is about 10<sup>7</sup> W/cm<sup>2</sup> and the gain increases linearly with the pulse intensity.

We also conducted numerical calculation based on the rate equations derived from the quasiclassical Boltzmann equation [3]. The carrier distribution (equivalent electron and hole distributions) is governed by the following equations for the total energy and concentration of carriers:

$$\begin{split} \frac{d\Sigma}{dt} &= \frac{1}{\pi^2} \sum_{i=\Gamma, \mathbf{K}} \int d\mathbf{k} \Big[ (1 - f_{h\omega_i - v_w \hbar k}) (1 - f_{v_w \hbar k}) / \tau_{i\mathrm{O,inter}}^{(+)} - f_{v_w \hbar k} f_{h\omega_i - v_w \hbar k} / \tau_{i\mathrm{O,inter}}^{(-)} \Big], \\ \frac{d\mathbf{E}}{dt} &= \frac{1}{\pi^2} \sum_{i=\Gamma, \mathbf{K}} \int d\mathbf{k} v_w \hbar k \Big[ (1 - f_{h\omega_i - v_w \hbar k}) (1 - f_{v_w \hbar k}) / \tau_{i\mathrm{O,inter}}^{(+)} - f_{v_w \hbar k} f_{h\omega_i - v_w \hbar k} / \tau_{i\mathrm{O,inter}}^{(-)} \Big] \\ &+ \frac{1}{\pi^2} \sum_{i=\Gamma, \mathbf{K}} \int d\mathbf{k} h\omega_i \Big[ f_{v_w \hbar k} (1 - f_{v_w \hbar k + h\omega_i}) / \tau_{i\mathrm{O,intra}}^{(+)} - f_{v_w \hbar k} (1 - f_{v_w \hbar k - h\omega_i}) / \tau_{i\mathrm{O,intra}}^{(-)} \Big], \end{split}$$

where  $\Sigma$  and E are the carrier concentration and energy density,  $f_{\varepsilon}$  is the quasi-Fermi distribution,  $\tau_{iO,inter}^{(\pm)}$  and  $\tau_{iO,intra}^{(\pm)}$  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  $\omega_{K} = 161$  meV, + for

absorption, and – for emission). We calculated the time evolution of the quasi-Fermi energy  $\varepsilon_F$  and carrier temperature  $T_c$ , which characterize the carrier distribution, after the pulse excitation. We observed that the population inversion, i.e., positive quasi-Fermi energy, occurs after around 1 ps of the excitation with some threshold pulse intensity. Fig. 2 shows the normalized dynamic conductivity of optically pumped graphene after 3.5 ps of the pulse excitation, as a function of pumping intensity with signal frequency 2 THz and with different momentum relaxation time. We took t = 3.5 ps because it approximately corresponds to the duration of the THz emission from the CdTe coming to the graphene after the pulse excitation in our experiment. Comparing Figs. 1(b) and (c), one can see the qualitative agreement with experimental and theoretical results.

## References

[1] V. Ryzhii, M. Ryzhii, and T. Otsuji, J. Appl. Phys. 101 (2007) 083114.

[2] V. Ryzhii, M. Ryzhii, A. Satou, T. Otsuji, A. A. Dubinov, and V. Ya Aleshkin, J. Appl. Phys. **106** (2009) 084507.

[3] A. Satou, T. Otsuji, and V. Ryzhii, Technical Digest of 2nd International Symposium on Graphene Devices: Technology, Physics, and Modeling, Sendai, Japan, 27-29 Oct. 2010.

[4] H. Karasawa, T. Komori, T. Watanabe, H. Fukidome, M. Suemitsu, V. Ryzhii, and T. Otsuji, J. Infrared Milli. Thrz. Waves onlin 2010. doi: 10.1007/s10762-010-9677-1.

[5] S. Boubanga-Tombet, S. Chan, A. Satou, T. Watanabe, V. Ryzhii, and T. Otsuji, European Optical Society Annual Meeting 2010, Paris, France, 26-29 Oct. 2010.

## **Figures**



Figure 1: (a) Schematic view of the THz amplified stimulated emission from optically pumped graphene, (b) emission spectra and (c) pumping intensity dependence of measured gain of THz emission from optically pumped graphene.



Figure 2: Normalized negative dynamic conductivity calculated theoretically.