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First success in stimulated emission of terahertz radiation from graphene a new material
First big step ahead to a new principle terahertz laser

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SUMMARY

Terahertz frequency band staying between electric microwaves and lightwaves had been unexplored for a long time, but now is emerging in an aspect of industrial applications to terahertz imaging in such fields as security/safety of material identification and/or living body measurement owing to rapid advancement of device technology for broadband light sources and highly sensitive detectors. All those technology, however, needs at least table-top size for system implementation. In order to furthermore utilize this potential frequency band into future ICT world including ultrahigh-speed communications like 100-Gbit/s personal wireless network, room-temperature operating integrated terahertz device technology is indispensable so that development of innovative terahertz microelectronic devices has been urgently required. Although a quantum cascade laser (QCL) is undoubtedly one of the candidates for that purpose, it suffers from room temperature operation due to phonon scattering. A new principle of terahertz laser should be appreciated.

In such a situation, research group of Prof. Taiichi Otsuji at RIEC, Tohoku University, Japan, in collaboration with research group of Prof. Ryzhii Victor at University of Aizu has succeeded in stimulated emission of terahertz electromagnetic radiation at room temperature from a new material called "graphene" under optical pumping with a femtosecond-pulse laser operating in the optical communication band. This result will be the first big step ahead to a new type of terahertz laser.

Graphene, a monolayer carbon-atomic honeycomb lattice crystal, has attracted attention due to its peculiar carrier transport and optical properties holding massless and gapless energy spectrum [1-3]. Linear dispersion relations for the electron and hole energy spectra with zero energy band gap in graphene provide nontrivial features like negative-dynamic conductivity in far infrared and terahertz spectral ranges [4]. Research and development of graphene materials/devices have been exploding over the world since the first success in fabrication of graphene done by the group of Prof. A. Geim at Manchester University, UK in 2004 [1].

Prof. Victor Ryzhii and his group member at University of Aizu, in collaboration with Prof. Otsuji's group at RIEC Tohoku Univ., had investigated the extraordinary optical property of graphene from the early time and proposed a new principle of terahertz laser operation in publication [4] in 2007. When graphene is optically pumped, the population inversion can be achieved. Due to very fast optical phonon emission $\tau_0 \approx 10^{-12}$s and relatively slow recombination τ_R □ 10^{-12} ps, as shown in Fig. 1, the photogeneration of electrons and holes leads to the emission of a cascade of optical phonons, so that the photoelectrons and photoholes occupy the states with the energies close to but not equal to the Dirac point: $\varepsilon_N = \hbar(\Omega/2 - N\omega_0) < \hbar\omega_0$, where $\hbar\Omega$ the pumping photon energy, $\hbar\omega_0$ the optical phonon energy, and N the number of emitted optical phonons. As a consequence, photoelectrons/holes recombine to radiate the photons with the energy $\hbar\omega = 2\varepsilon_N$ [4]. Due to large optical phonon energy of ~200 meV and gapless/symmetric band structure, stimulated emission in a wide terahertz frequency range is expected by tuning the pumping photon frequency. In publication [4], we have analytically revealed that when the pumping energy exceeds a threshold level, the dynamic conductivity at the radiating terahertz photon frequency ω_0 becomes negative, giving rise to stimulated emission of terahertz radiation. This leads to the laser operation when the active gain media of graphene is installed into a laser cavity.

In order to verify the proposed new insight, we conducted an experimental observation of electromagnetic radiation from optically pumped graphene. The sample used in the experiment is our original "heteroepitaxial graphene on silicon" which has been developed by Prof. Maki Suemitsu and his group members at RIEC, Tohoku Univ. in the framework of a JST-CREST project (see Fig. 2). To obtain the terahertz photon radiation as the consequence of the above mentioned carrier relaxation/recombination dynamics, the pumping photon energy/wavelength was carefully selected at around 1550 nm. To perform an intense pumping beyond its threshold, a femtosecond pulsed fiber laser was used as the pumping source. Time-resolved field emission property was measured by an electrooptic sampling method (see Fig. 3). Its Fourier spectrum reflects the pumping laser spectrum, which is the clear evidence of the occurrence of stimulated emission (not the spontaneous emission) (see Fig. 4). The next step is to accommodate the graphene into a laser cavity to realize continuous-wave laser operation in the terahertz range. This is the first big step ahead to a new type of terahertz lasers that enable the future ultrahigh-speed wireless communications as well as ubiquitous smart sensor network innovation.

The result obtained for the first time was presented at the European Conference on Lasers and Electrooptics (CLEO-Europe) on June 19th in Munich, Germany [5]. This work has been finantially supported by the JST CREST program "Development of Graphen-on-Silicon Device/Process Technology" and the JSPS Grant-in-Aid for Basic Research (S) program

“Exploring Novel Electromagnetic Circuit Based on Management of Low-Dimensional Plasmonic Dispersion.”

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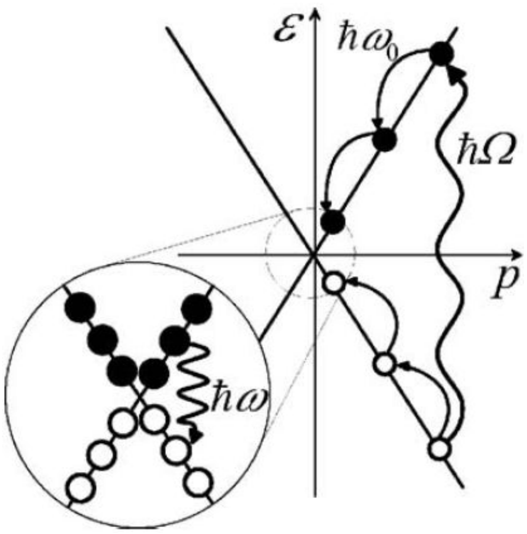


Fig. 1. Carrier relaxation and recombination process for optically pumped graphene.

Valence electrons are optically excited to the conduction band. Photogenerated electrons (black circle) and photoholes (white circle) will emit a cascade of optical phonons to lose their energy on the respective band. Finally no-more optical phonons are able to be emitted but still stay at excited states beyond the Dirac point (crossover point, corresponding to the equilibrium Fermi level) so that they will recombine to emit a photon.

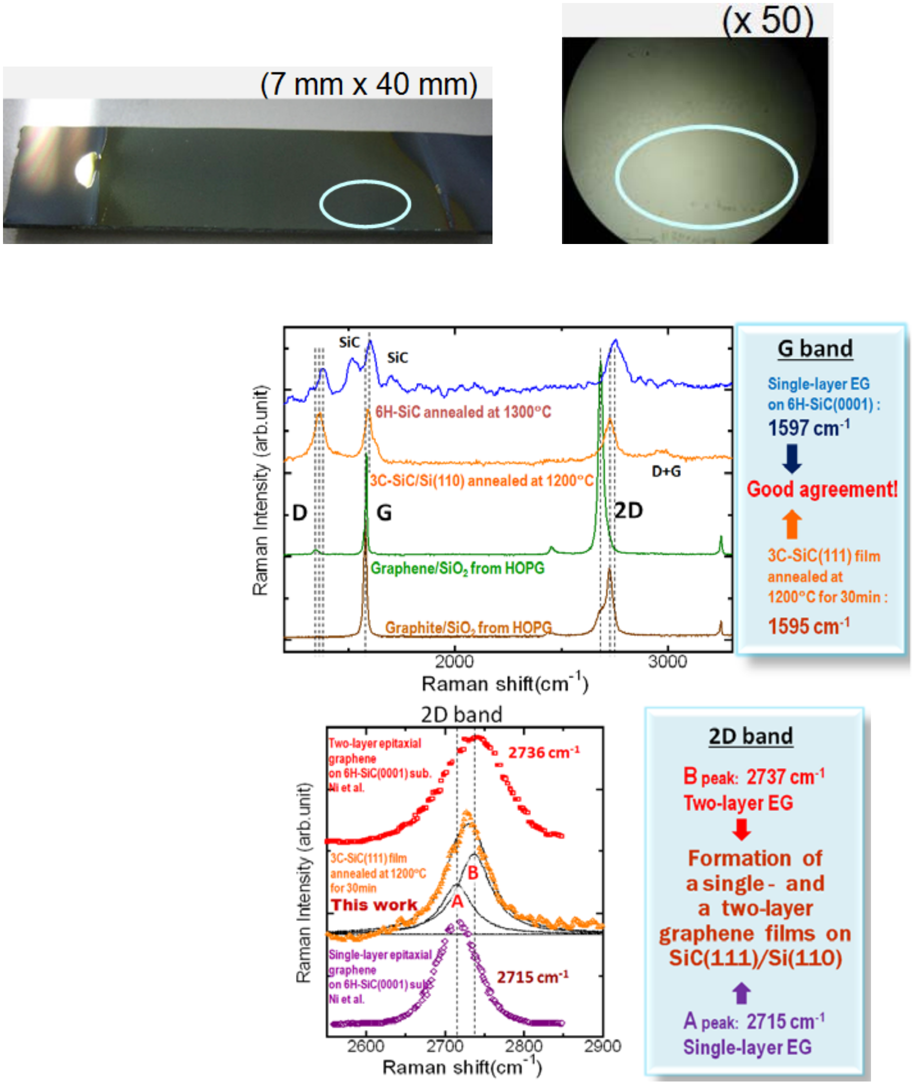


Fig. 2. Heteroepitaxial graphene on silicon. Upper: photo and microzoom images, middle: Raman spactra at D, G, and 2D band in comparison with other samples made with different technology, lower: Raman spactra at 2D band.

The G band spactra directly reflect the crystal lattice structure of “graphite” and corresponding optical phonon energy is defined to be 198 meV. The D band is a defect-oriented spectrum, and the 2D band includes the information of the number of the layers. The sample utilized in this work includes mono- and bi-layer of graphene. The sample is an original of Prof. Maki Suemitsu and his group members at RIEC Tohoku Univ.

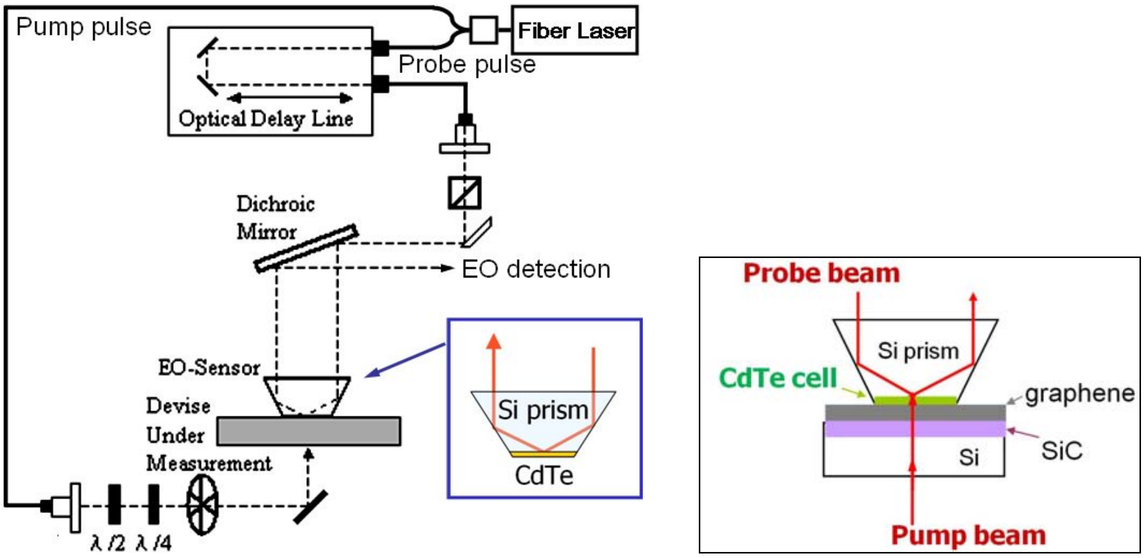
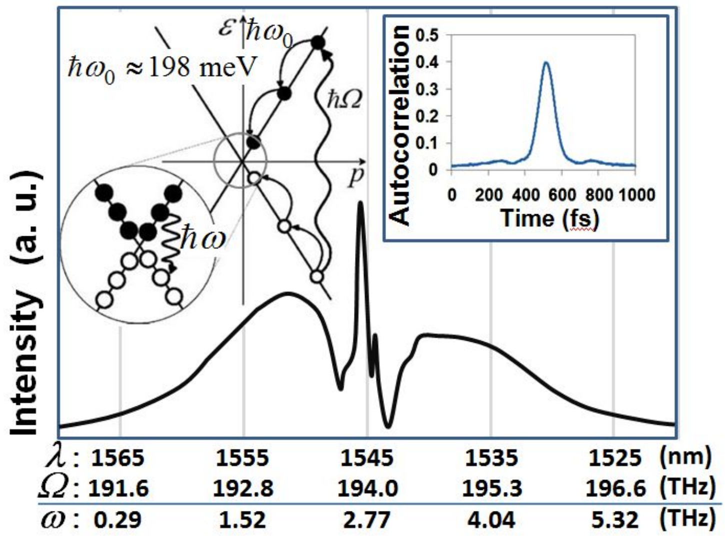


Fig. 3. Electrooptic sampling system.

A single femtosecond fiber laser is divided in two beams one for pumping and the other for probing. The pumping laser is illuminated from the backside of the sample in a linear polarization while the probing laser is illuminated from the topside. A electrooptic sensor crystal, CdTe, is placed on top of the sample surface. Throughout the Si prism attached onto the sensor, the probing beam is reflected back to the detection block. When the beam is reflected at the CdTe sensor surface, the polarization is modulated depending on the electric field intensity arisen from the sample. By sweeping the probing timing by means of optical delay line, whole the temporal profile of the field emission property is sampled.



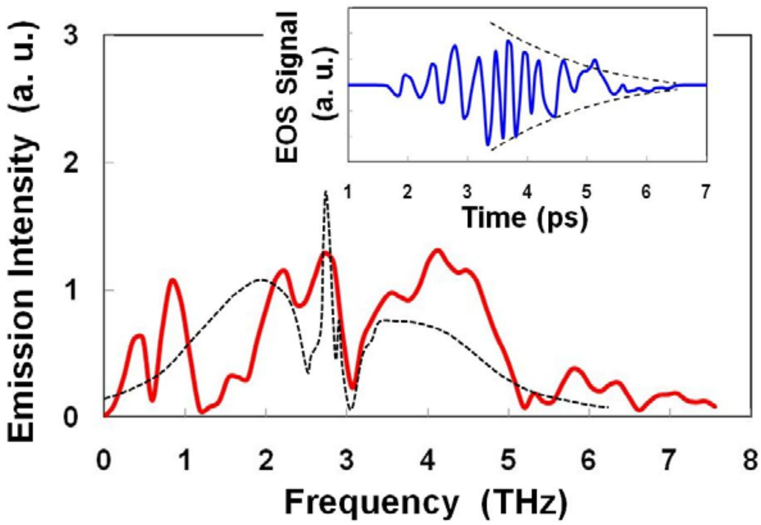


Fig. 4. Upper: temporal and spectral profiles of the pumping laser beam used in this experiment, lower: measured field emission property (inset: temporal response, main plot: Fourier spectrum).

80-fs pulse width, 20-MHz repetition fiber laser is utilized. Horizontal axis in upper figure indicates its wavelength and frequency together with the estimated terahertz photon frequency radiated from the sample. The dashed line in lower figure is a replica expected from the pumping laser spectrum. The solid red line is the Fourier spectrum of the measured temporal response which is fairly corresponding to the pumping one. The lower cutoff around 2 THz may be caused by small bandgap opening due to the existence of bilayer graphene. Since the measurement was done in an averaging fashion, the obtained response is undoubtedly a coherent process which is never obtained in spontaneous emission but only done in stimulated emission.