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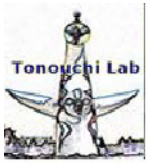

Osaka University

Molecules Absorption to Graphene Studied by Laser Terahertz Emission Microscope

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Osaka University, Osaka, Japan

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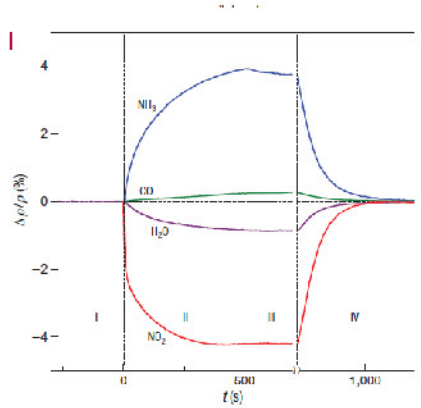
Rice University, Houston, U.S.A.

"Imaging molecular adsorption and desorption dynamics on graphene using terahertz emission spectroscopy," Scientific Reports 4, 6046(2014)

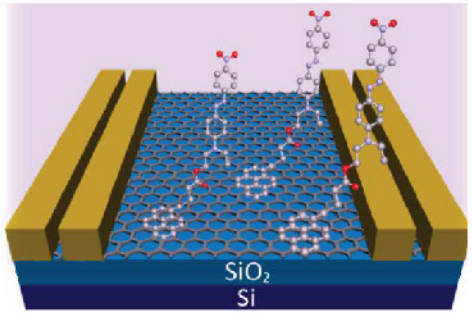
This work was supported by Grant-in-Aid 25630149 and 26107524, Core-to-Core program, MEXT/JSPS, the Program for promoting international joint research, Osaka University, the Murata Science Foundation, the Robert A. Welch Foundation through Grant No. C-1509, and the National Science Foundation through Grant No. OISE-0968405.

Background

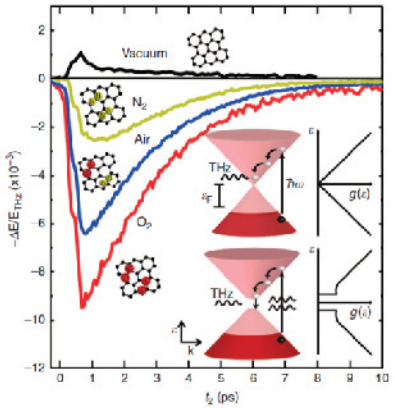
Resent researches on Gas Sensing with Graphene



Nat. Mater. **6**, 652 (2007)



*Nano Lett.***12**, 182(2012)



Nat. Comm. **3**, 1228 (2012)

Carrier doping induced by adsorbed gas molecules changes conductivity of graphene.

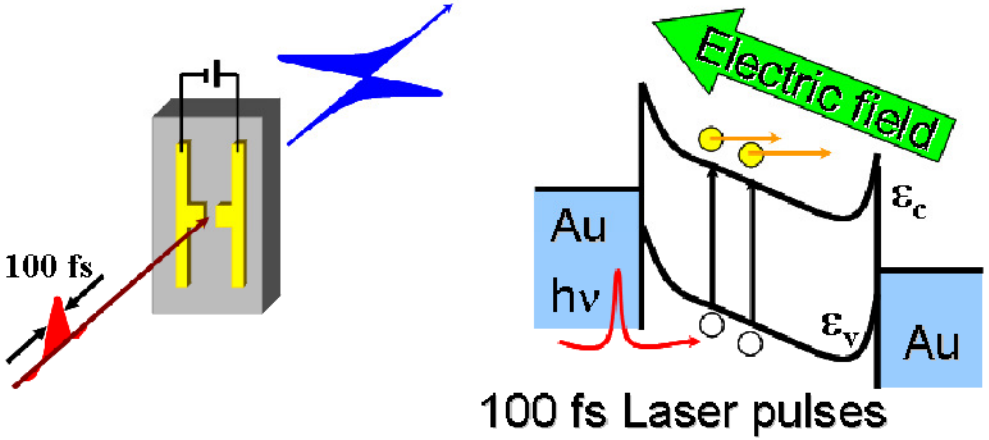
For advanced graphene device it is valuable to characterize local condition of graphene films.

In this study, we propose a new approach (THz emission) to evaluate 2D distribution of gas molecules on graphene.

How to generate THz pulses

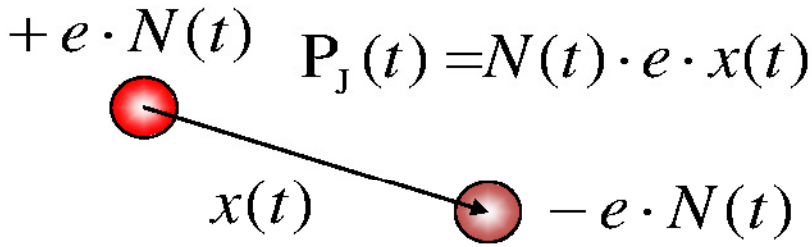
$$\nabla^2 E - \mu\varepsilon\frac{\partial^2 E}{\partial t^2} = \mu\frac{\partial j}{\partial t} \qquad j = \frac{\partial P(t)}{\partial t} \qquad \frac{\partial j}{\partial t} = \frac{\partial^2 (P_J + P_{NL})}{\partial t^2}$$

Photocurrent generation



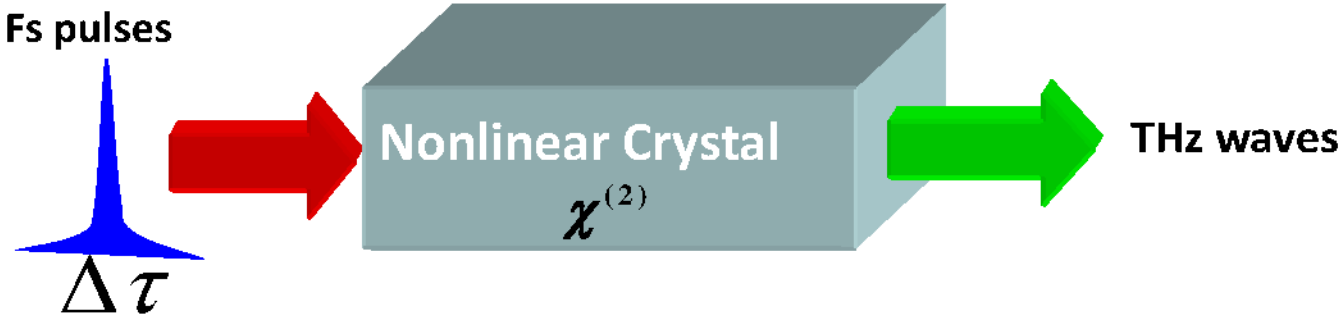
100 fs Laser pulses

Electromagnetic source



$$E_{far}(z,t) \propto \frac{\partial^2 P(t - z/c)}{\partial t^2}$$

Optical Rectification

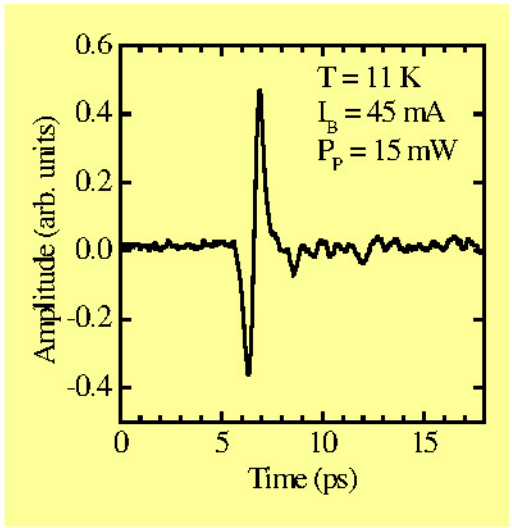


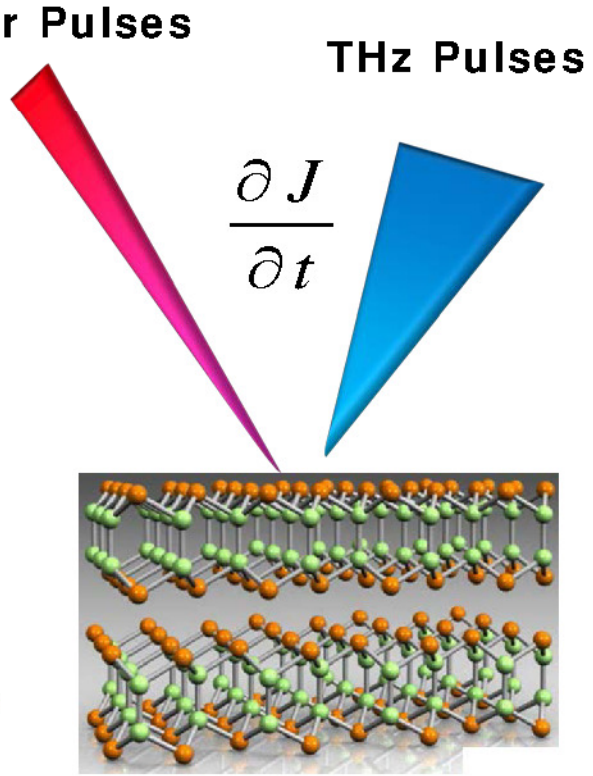
$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E_1(t) E_2(t)$$

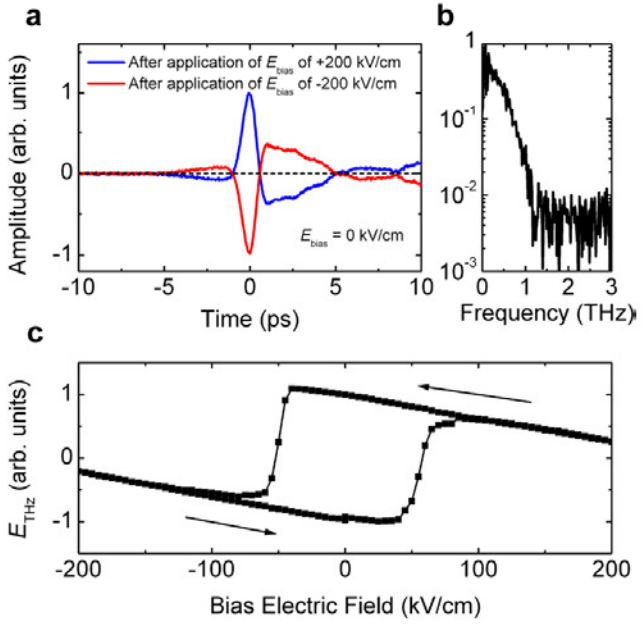
THz Emission from Various Materials excited with a femtosecond laser

Osaka University

One can observe terahertz (THz) radiation from various kinds of materials, when excited with a femtosecond laser, owing to ultrafast current modulation. THz waves reflect various kinds of properties such as local electric field, particularly ultrafast transient phenomena, in their waveforms







High Tc Superconductor

Jpn. J. Appl. Phys, 35(1996)2624.

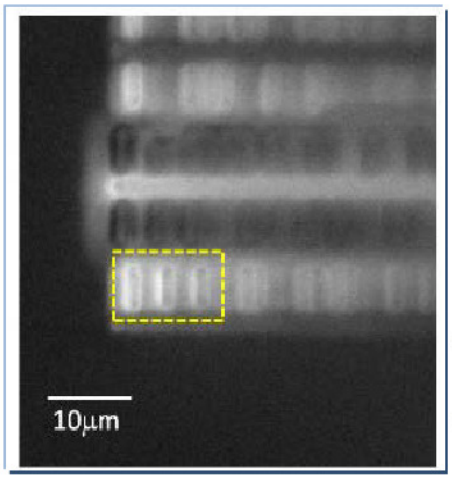
Ferroelectrics $BiFeO_3$

Phys. Rev. Lett. 96, 117402 (2006).

Scanning Laser THz Emission Microscope

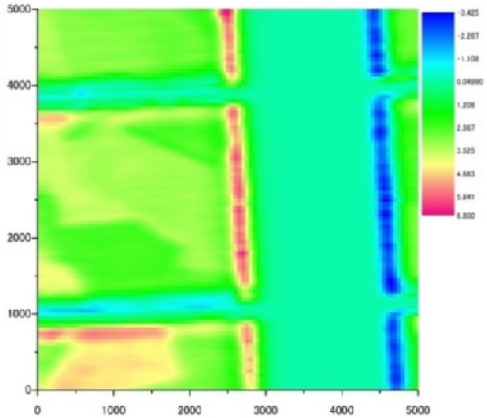
Osaka University

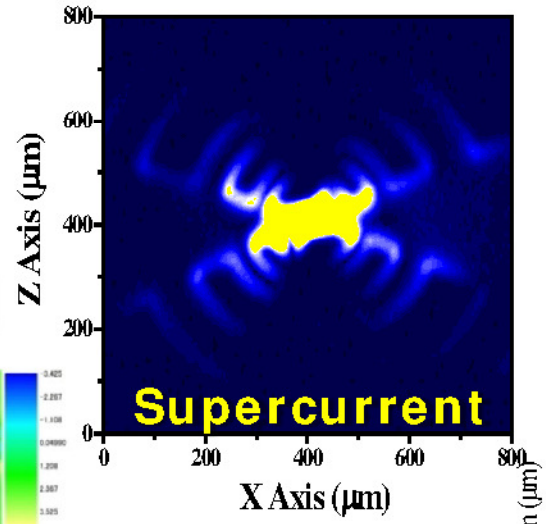
One can visualize the emission image by scanning the laser beam on it, the resolution of the image is limited by the laser beam diameter rather than THz wavelength. Thus construction of a laser THz emission microscope (LTEM) would provide a new tool for material science and application.



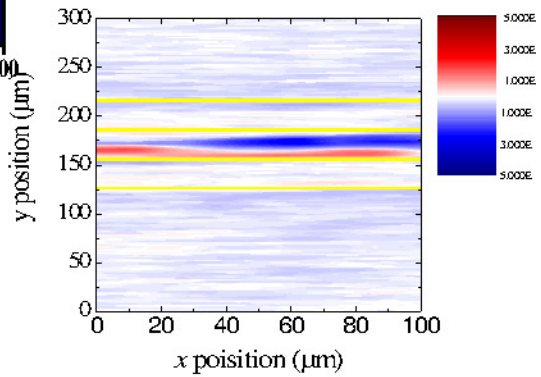
LSI

Solar Cell

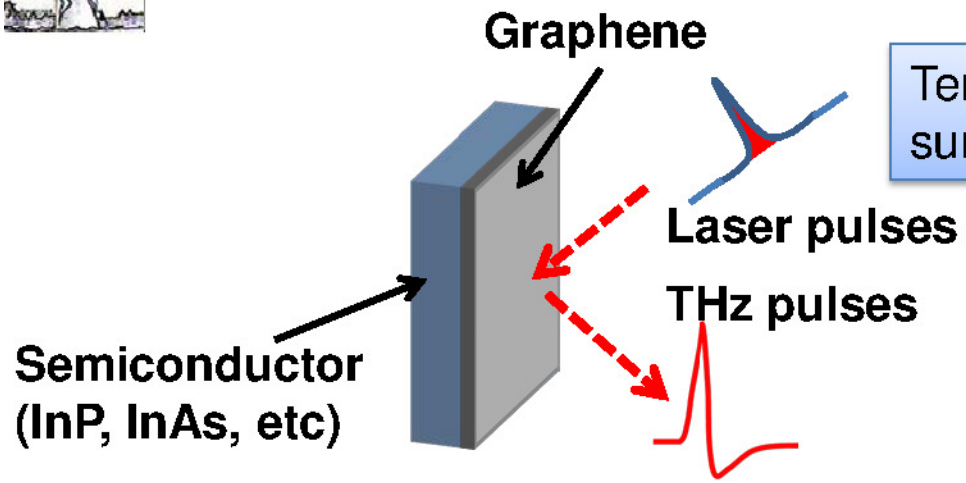




Ferroelectric Domain



Approach

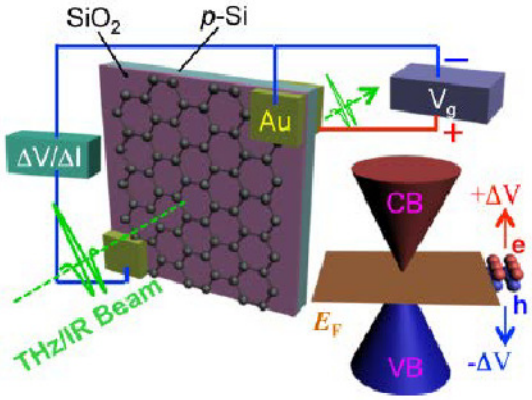


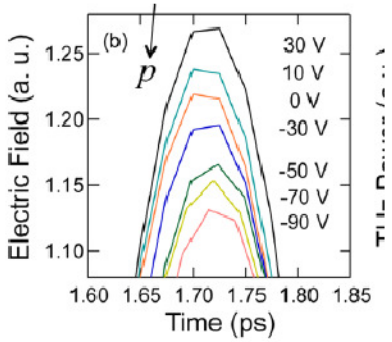
Terahertz emission from semiconductor surface due to photoexcited surge current.

- **Fermi energy shift**
- **Change of Band bending**

Change of terahertz radiation waveform and intensity

Fermi energy shift

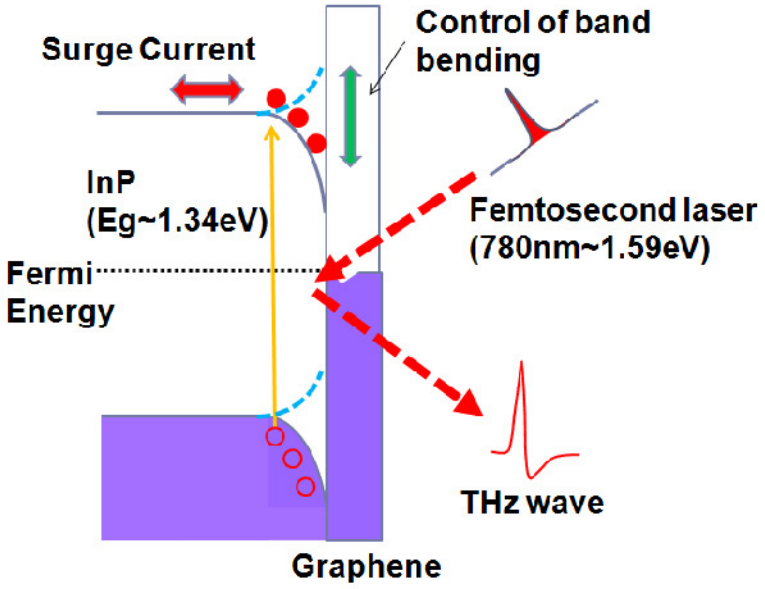




Nano Lett. **12**, 3711 (2012)

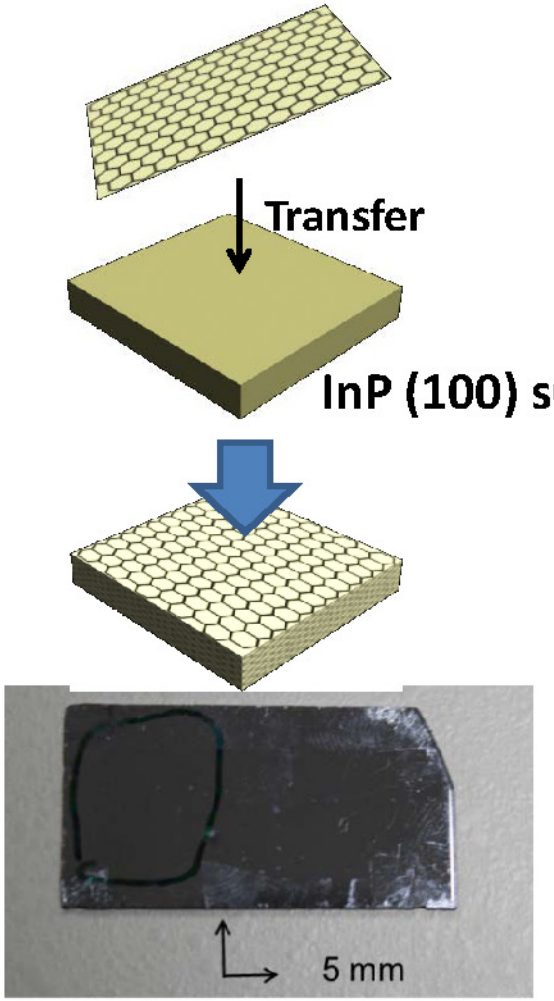
Change in transmittance of THz waves

Modulation of band structure



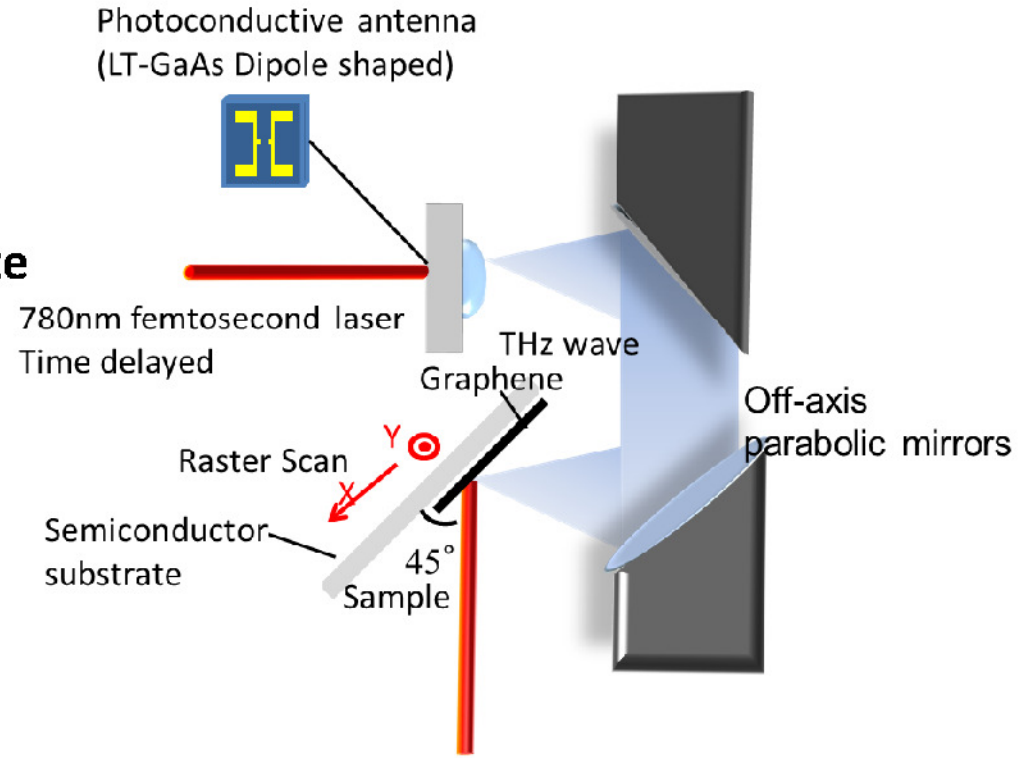
Experimental Setup

CVD Graphene



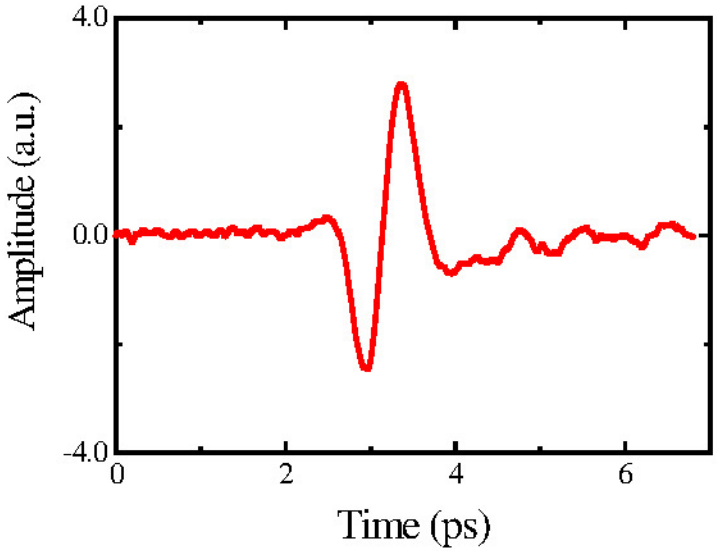
Graphene on various kinds of substrates is available.

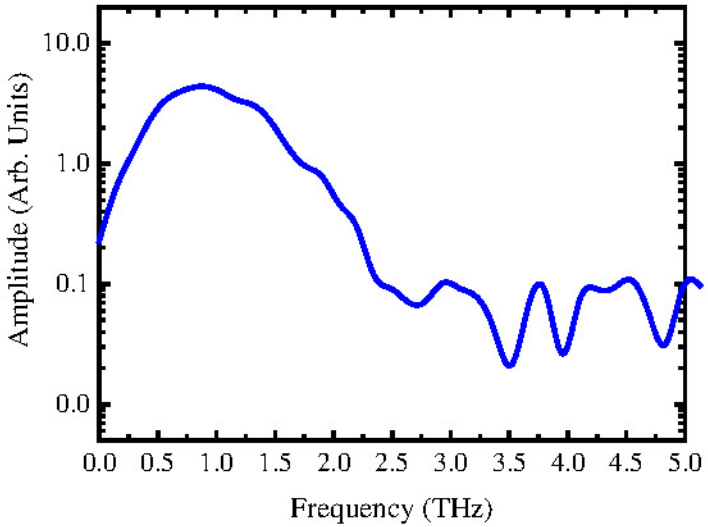
Laser THz Emission Spectroscopy/Microscopy



- Laser power: ~ 20 mW
- Spot size: ~200 μmϕ
- Fluence: ~400nJ/cm^2

Distribution of THz Wave Amplitude





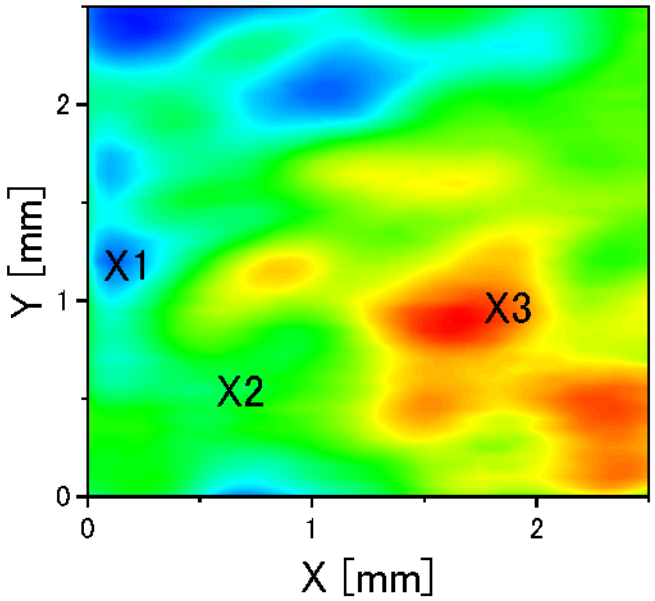
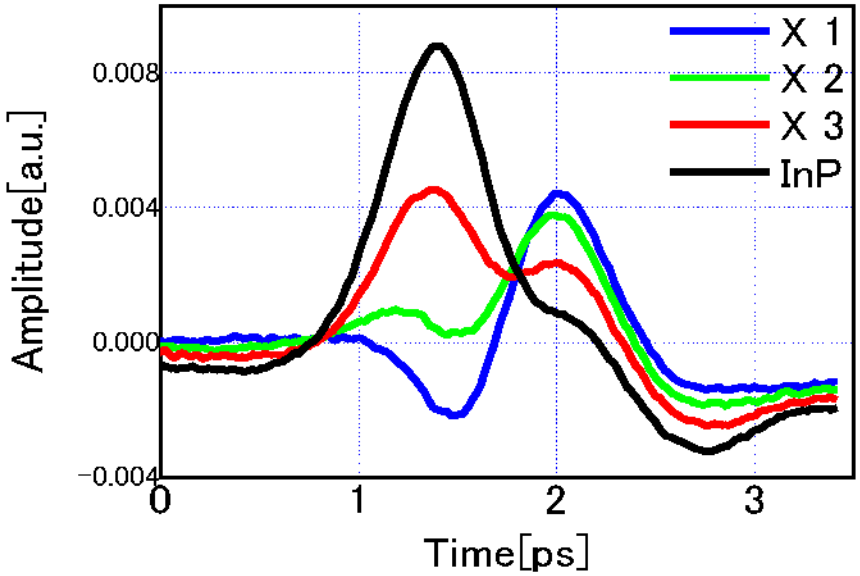
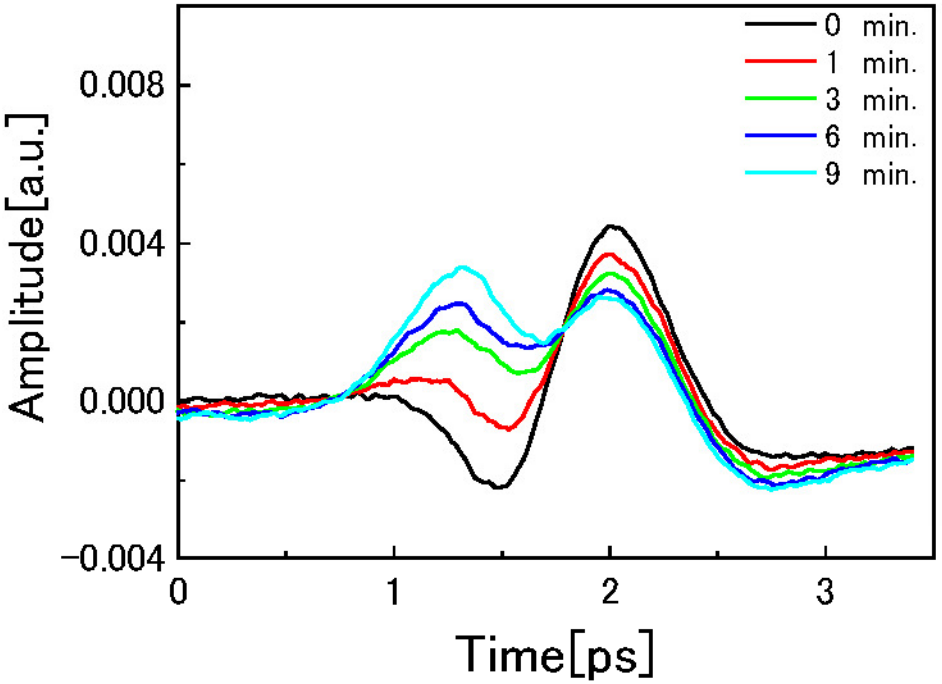


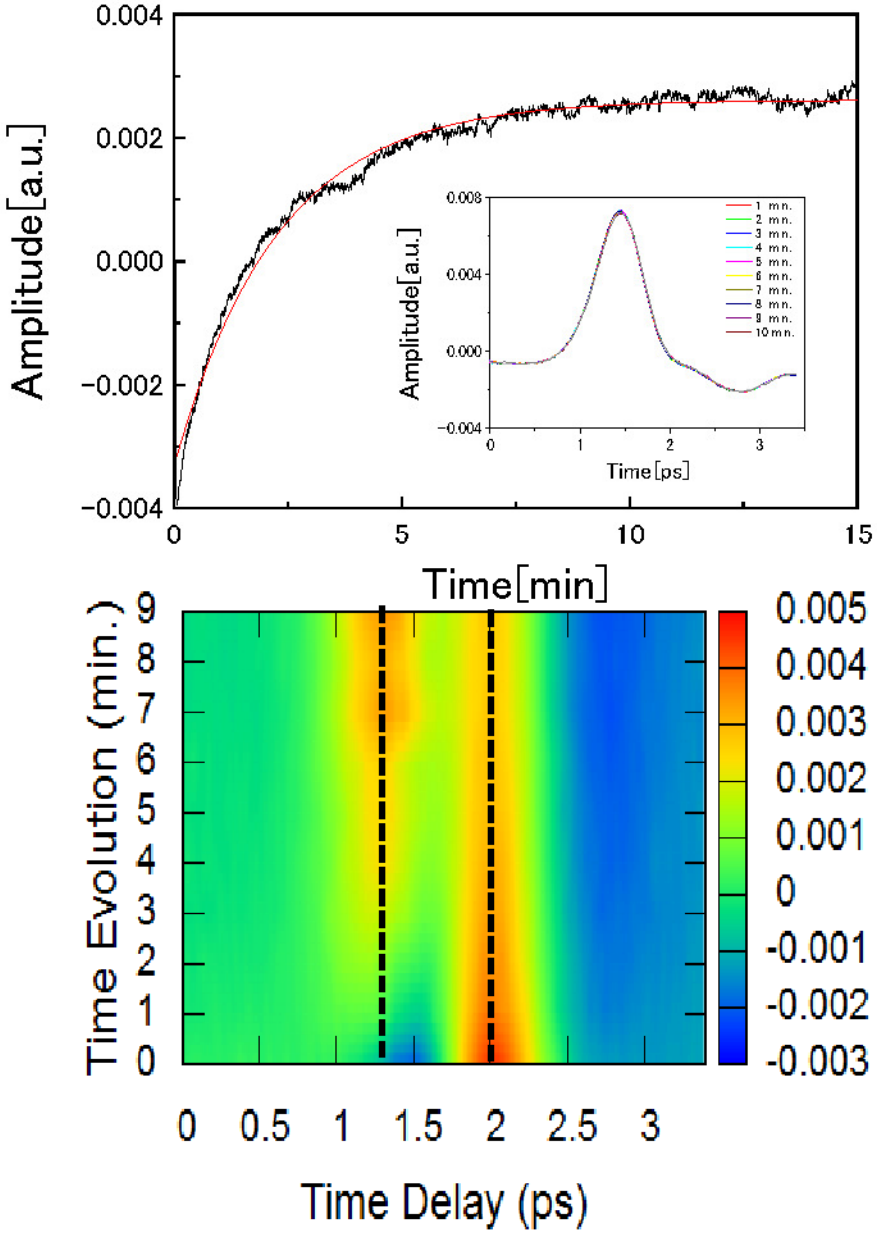
Image of THz wave amplitude



THz waveforms at the points, X1,X2,X3

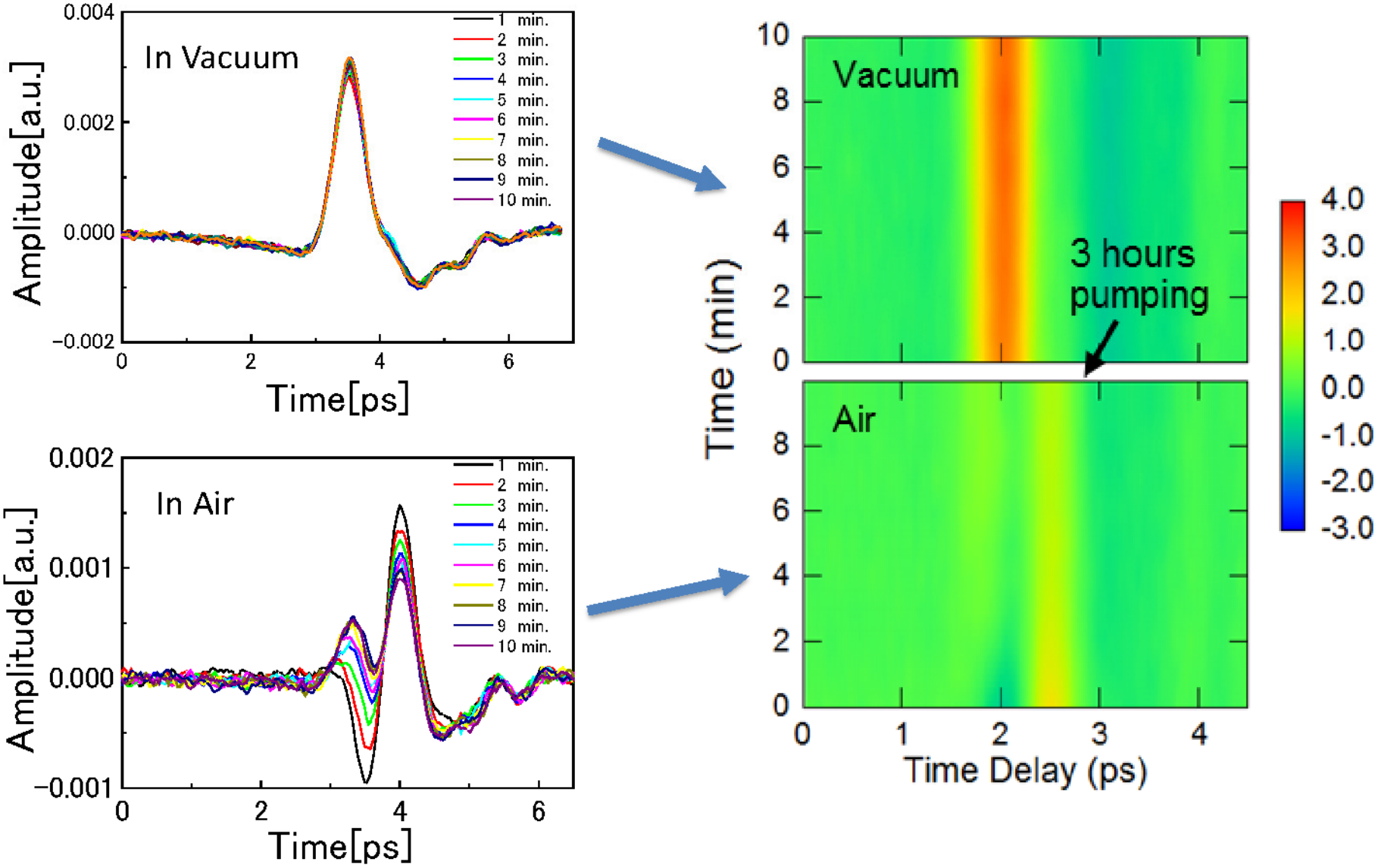
Slow Time Dependence



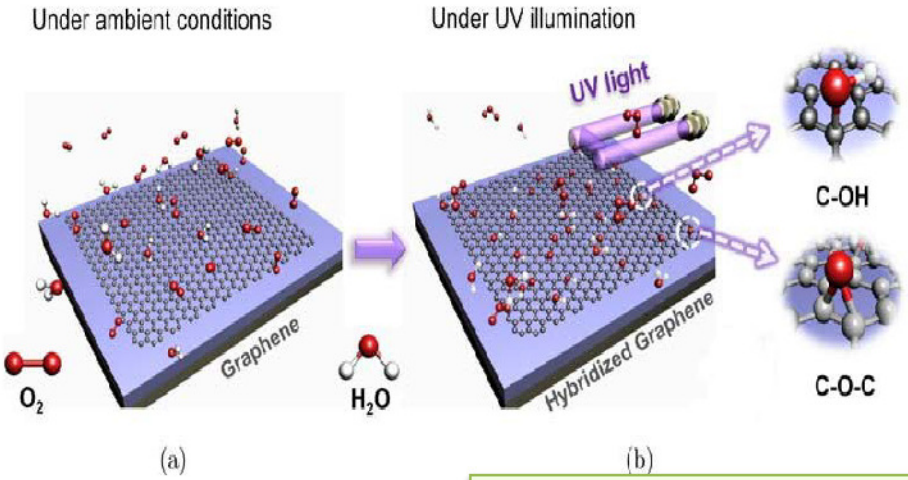


Two peaks appear in the terahertz waveform from Graphene/InP.

Terahertz Waveforms in Air and Vacuum



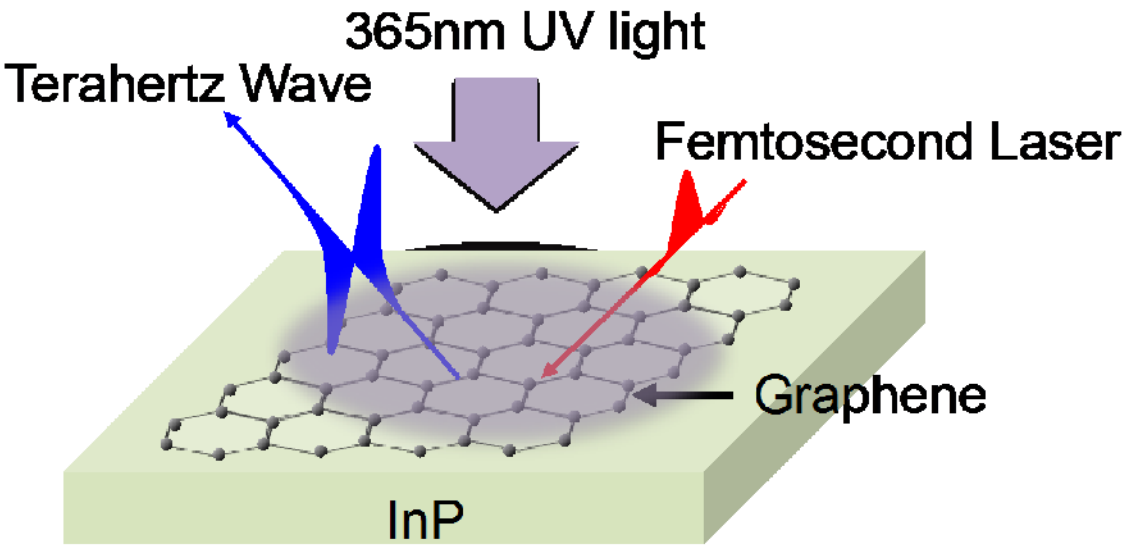
UV light illumination effect



UV illumination assists oxidation of graphene.

NANO: Brief Reports and Reviews **6**, 409 (2011)

Experimental procedure



Evacuation of air

↓

Gas introduction

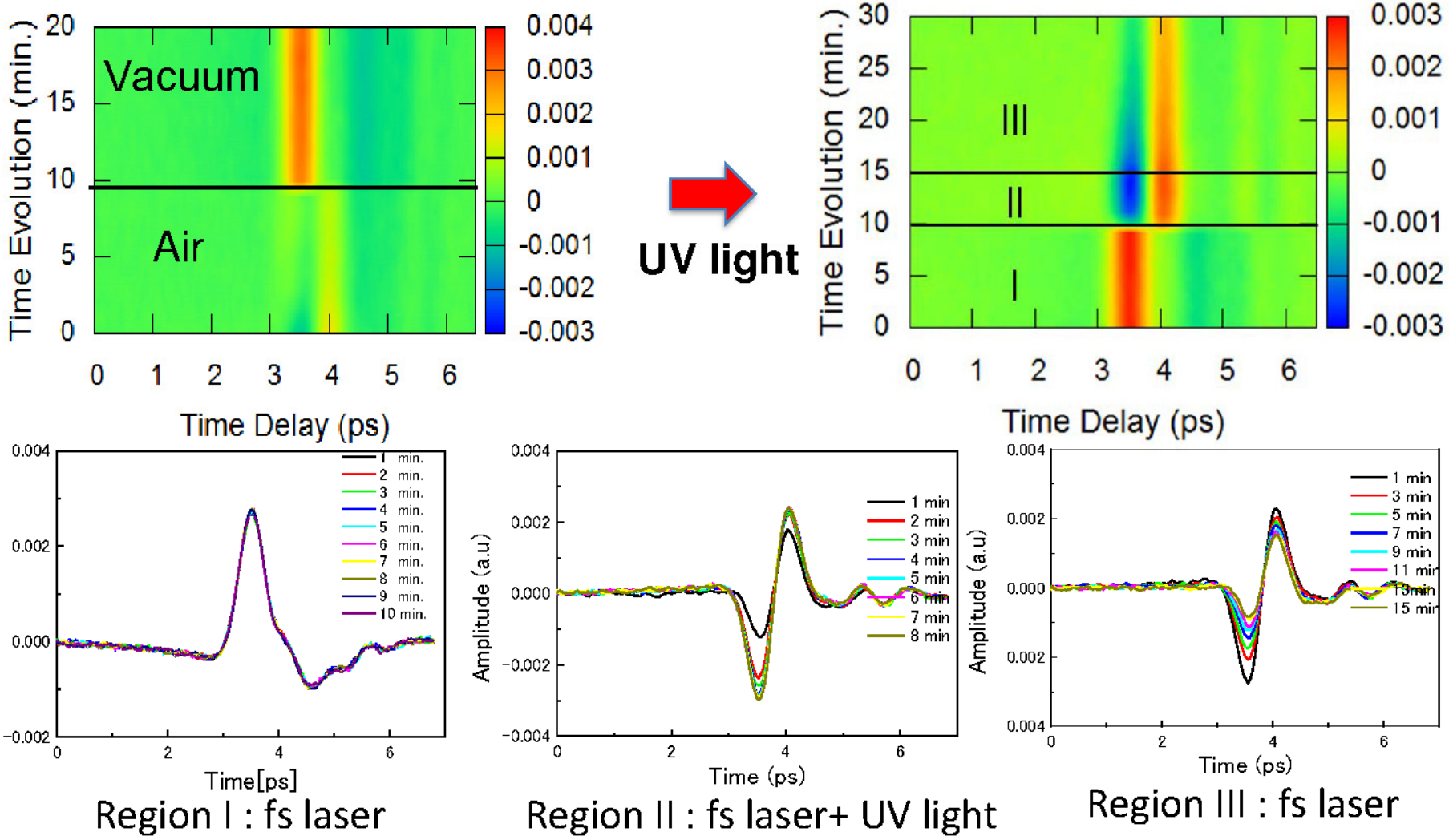
↓

UV light illumination

↓

Switch off the UV light

UV Light Effect in Air



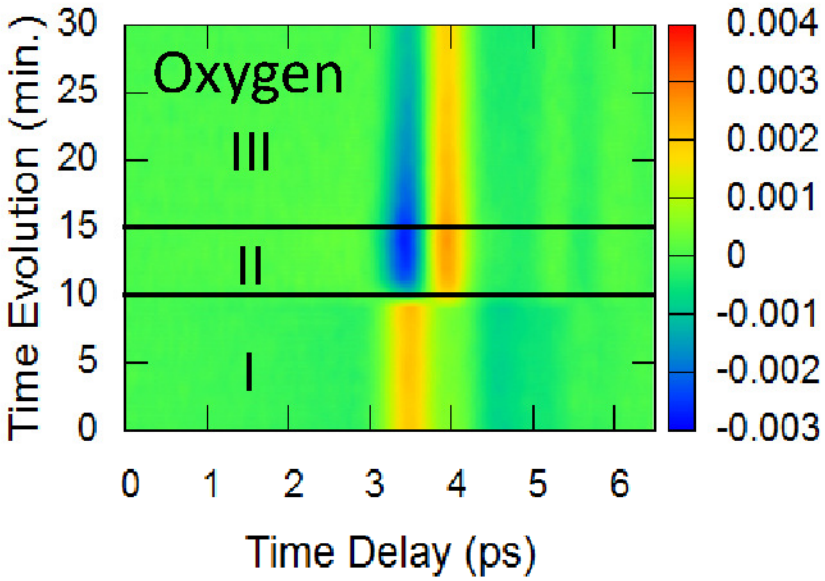
Region I : fs laser

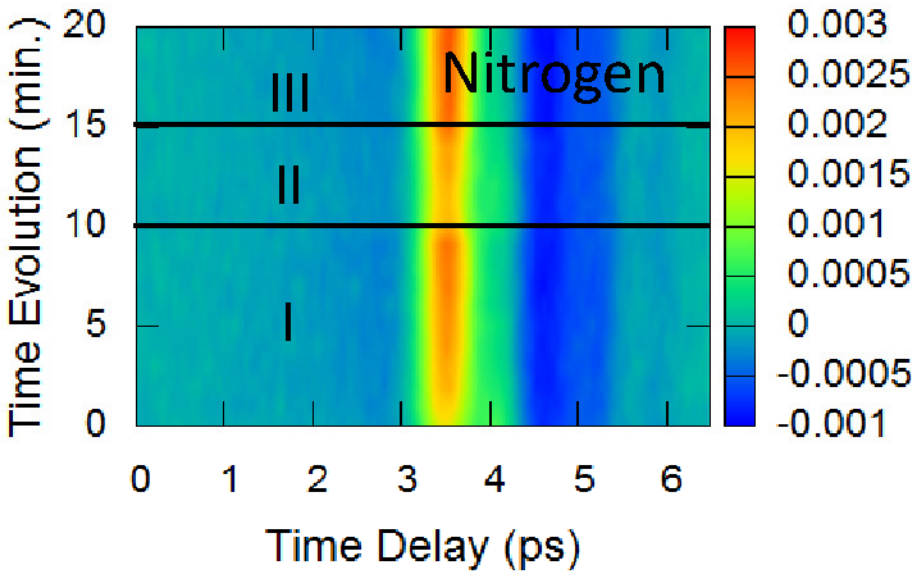
Region II : fs laser+ UV light

Region III : fs laser

Drastic change in the terahertz waveforms under UV light

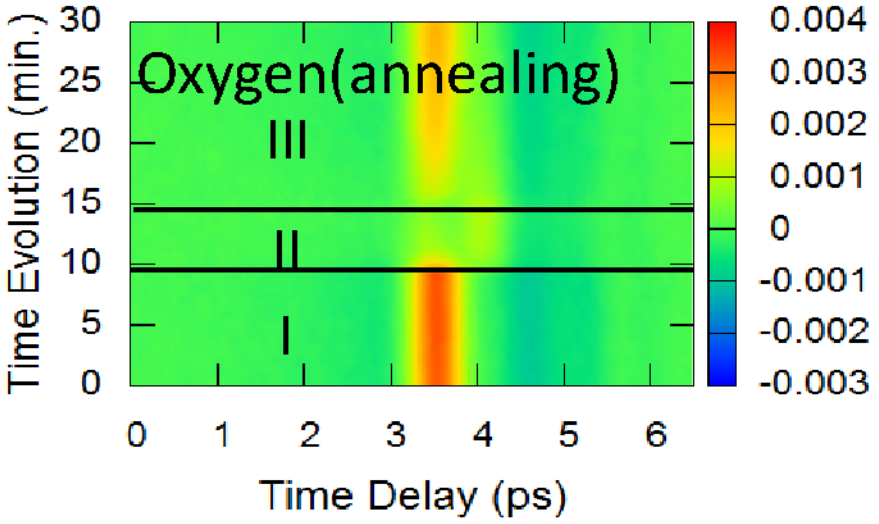
Terahertz Emission Under UV light



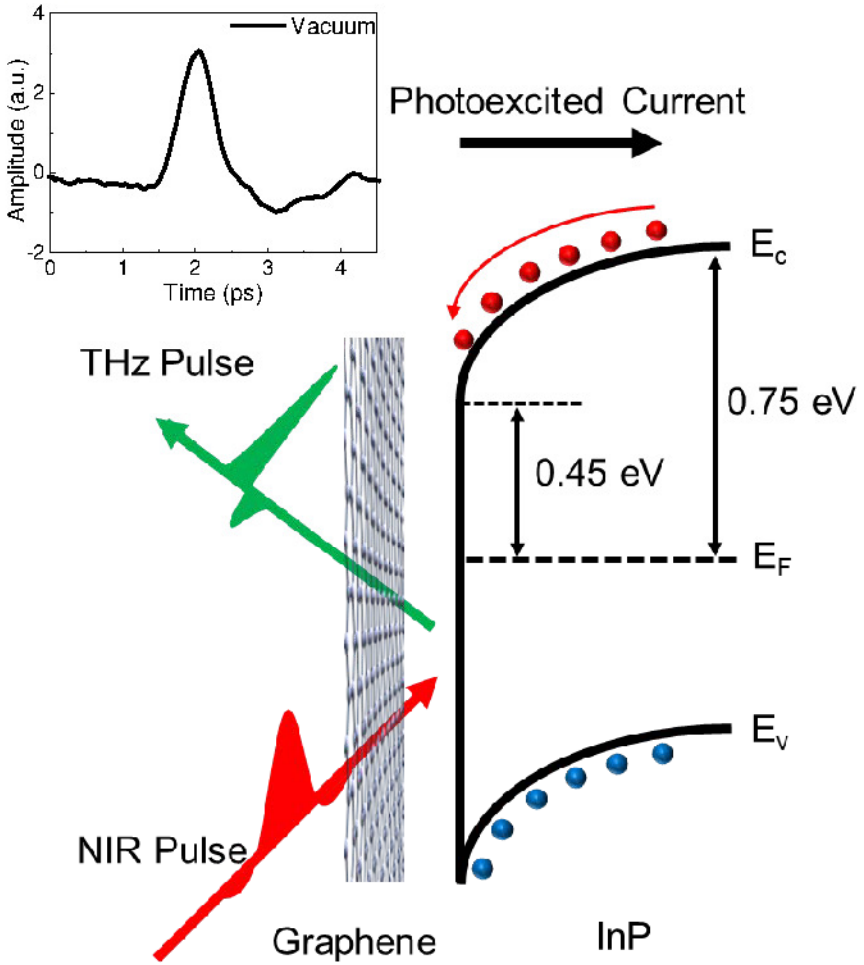


Pre-annealing:
120 ° C, 2hour

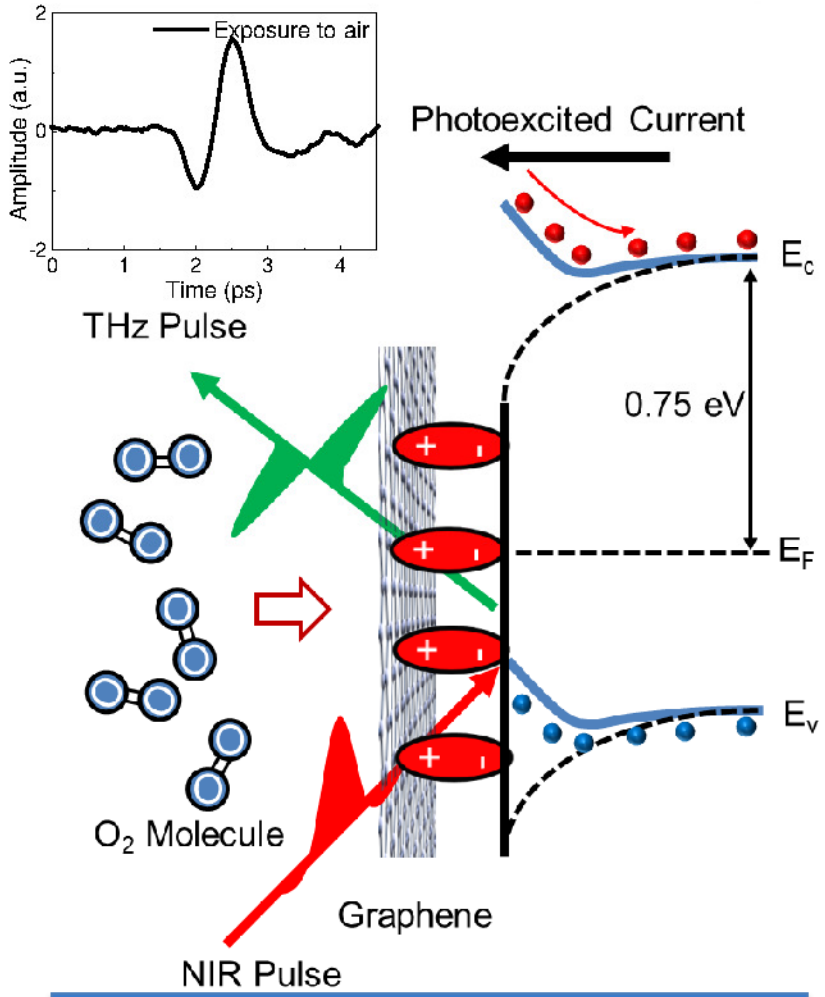
Smaller change in THz waveform



Discussion



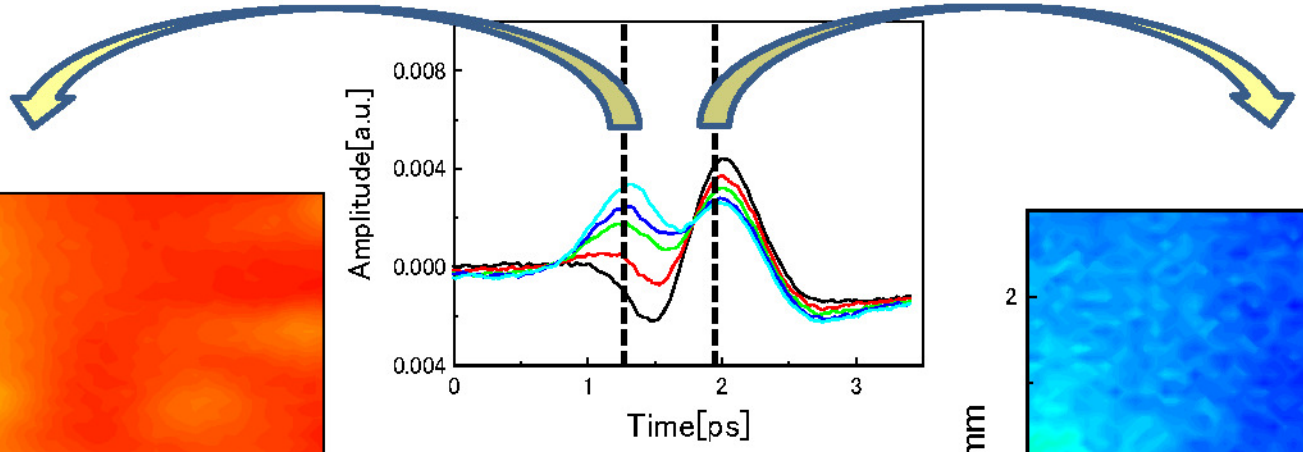
Desorption of oxygen

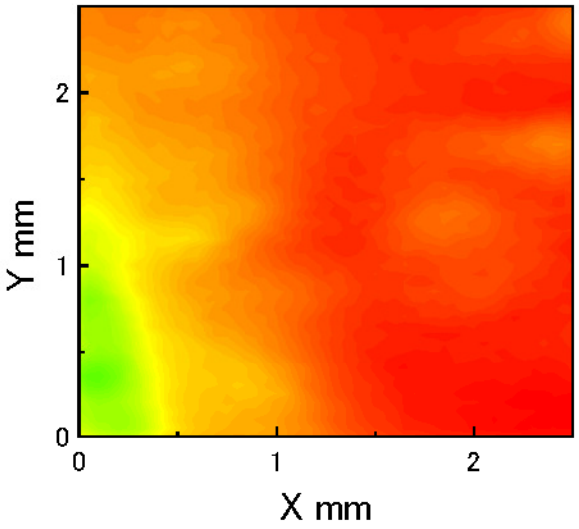


Adsorption of oxygen

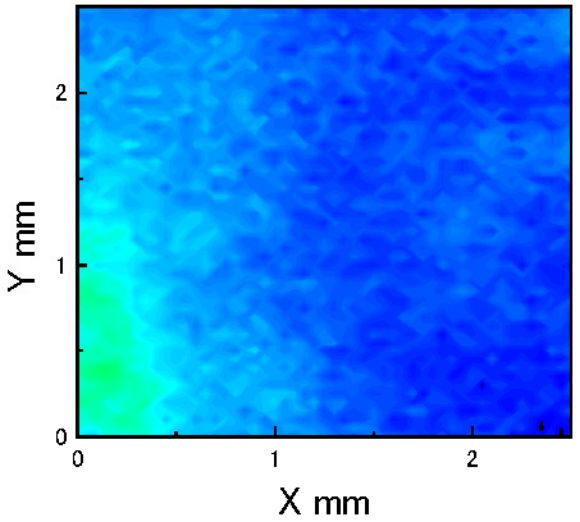
Changes in THz waveforms are caused by the existence of electric dipoles induced by adsorbed oxygen molecules.

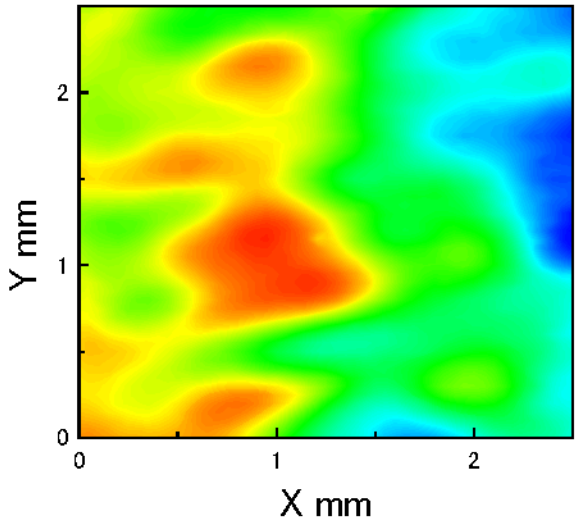
THz imaging



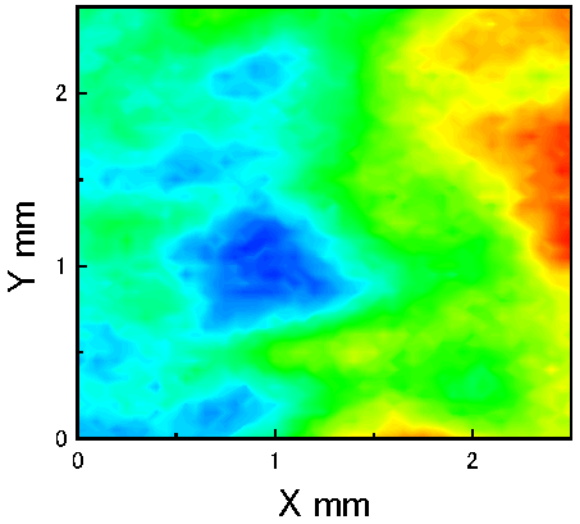


InP





Gr+InP



Summary

- THz waveforms from graphene-coated InP changed with time under pulsed laser excitation.
  - # The first peak in the THz waveform increased and the second peak decreased under the illumination of fs laser.
  - # No changes in Vacuum.
  - # UV light illumination enhanced the change of the waveforms illumination.
- Large intensity distribution of THz radiation indicates the feasibility of 2D mapping of adsorbed gas molecules.
- The change of the THz waveforms can be explained by modification of the surface depletion-layer potential of InP due to the adsorbed gas molecules.

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