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# Observation of Broadband THz/Mid-IR Emission in Near-IR Laser-Pumped Multilayer Graphene

Graphene has attracted a great deal of attention as a two-dimensional electronic material having precedent peculiar properties originated from its linear and gapless dispersion relation. One of such nontrivial features of graphene is the negative-dynamic conductivity in the terahertz (THz) spectral range, which makes the creation of a new principle THz coherent light source in reality [1]. The ultrafast nonequilibrium carrier relaxation and recombination dynamics in optically pumped graphene has extensively studied in the last decade [2]. We have experimentally demonstrated THz spontaneous emission in a carrier-population-inverted exfoliated monolayer graphene under fs-IR laser pumping [3]. Independently carrier population inversion in an epitaxial graphene on a SiC substrate even 1 ps after fs-IR laser pumping was experimentally confirmed by using time- and angle-resolved X-ray photoelectron microscopy [4]. These are clear manifestations of the evidence that graphene works for a gain medium. One critical limit, however, is very weak quantum efficiency of 2.3% at maximum per monolayer graphene [1, 5]. Theoretically it is predicted that the non-Bernal-stacked multilayer graphene could multiply the gain of monolayer graphene by the number of layers [6]. In this report, we experimentally observe the THz emission from optically pumped non-Bernal-stacked multilayer graphene.

We prepared for a thermally grown multilayer epitaxial graphene on a C-face SiC substrate [3]. Raman spectroscopic measurement confirmed its non-Bernal-stacked graphene properties (Fig. 1) [5]. We conducted Fourier transform far-infrared spectroscopic measurement using a HgCdTe detector to the multilayer graphene sample under pulsed infrared optical pumping at 250, 275, and 300 K. The sample was pumped by a SOLAR OPO laser complex equipment capable of emission of coherent near-IR radiation with wavelengths from 1.94 to 2.3 μm (with a KTR OPO unit with a pulse energy of up to 5 mJ), a pulse duration of 10 ns, and a repetition rate of 10 Hz. A pulsed THz $CO_2$ laser with a wavelength of 10.6-μm, a pulse duration of 70 ns, a repetition rate of up to 50 Hz, and a pulse energy of up to 40 mJ was also prepared for an alternative source as well. The HgCdTe detector was responsible for detection of radiation in the frequency range above ~20 THz.

In the OPO laser measurement, a broadband photoluminescence was observed in the range from 20 (limited by the detectable lower cutoff) to 45 THz for the first time (Fig. 2). The left sides of these peaks are determined by the lower limit of the detectable frequency range of the HgCdTe detector, ~20 THz. Therefore, the actual gain peaks may extend further to lower frequency region. The temperature dependence of the luminescence intensity needs understanding the new physics behind. On the other hand, when the sample was excited by a pulsed $CO_2$ laser with a working wavelength of 10.6 μm, no photoluminescence was observed. It is reasonably understood that the pumping energy is less than the twice of the Fermi level in the sample under measurement so that it could not make carrier population inversion (Fig. 3).

In conclusion, we succeeded in observation of broadband THz/mid-IR spontaneous emission from non-Bernal-stacked multilayer graphene. Further experiment is on going to verify if it could provide an amplification gain being multiplied by the number of layers from that for monolayer graphene.

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## References


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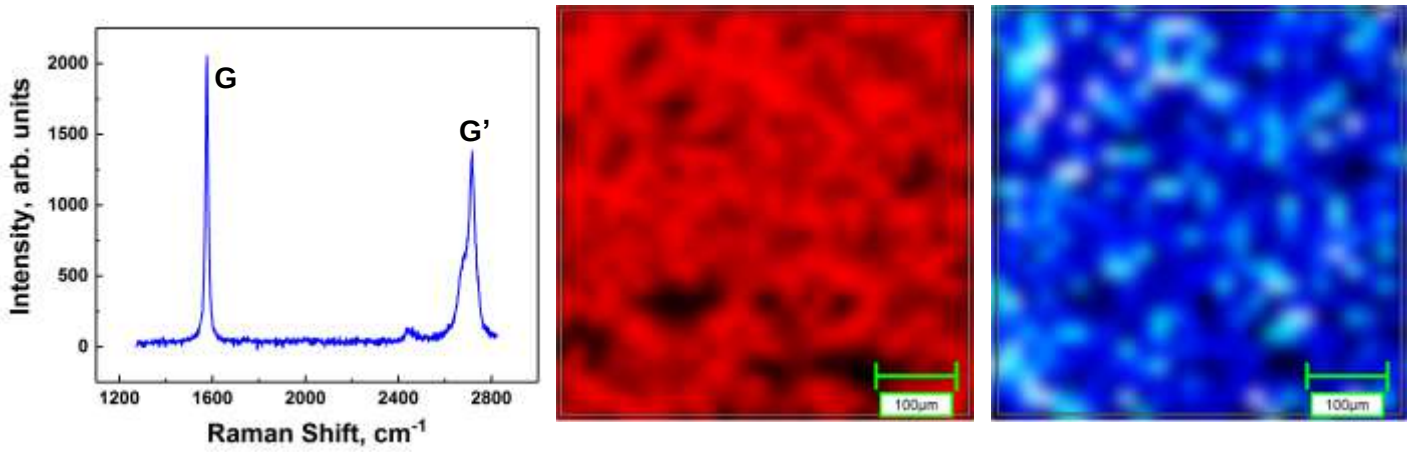
## Figures




(a) Raman spectrum of multilayer graphene (b) G' peak intensity (c) G/G' intensity

**Figure 1:** Raman spectrum of the multilayer graphene sample under measurement. (a) A Raman spectrum at a point. Intensity ratio of G' to G and the shape of G' confirm non-Bernal-stacked graphene properties [5]. (b) A mapping image of G' peak intensity. Red colored area shows existence of graphene there. (c) A mapping image of intensity ratio of G' to G. In the more blue the region is, the more layers of graphene assumes to be there.

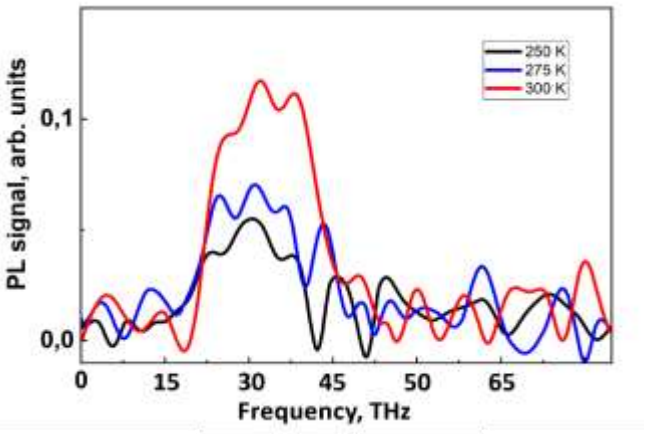



**Figure 2:** Fourier-transformed spectra of the photoluminescence at 250-300 K from epitaxial multilayer graphene pumped with 2.3-μm near-IR photons. The left sides of these peaks are determined by the lower limit of the de-tectable frequency range of the HgCdTe detector,~20 THz.

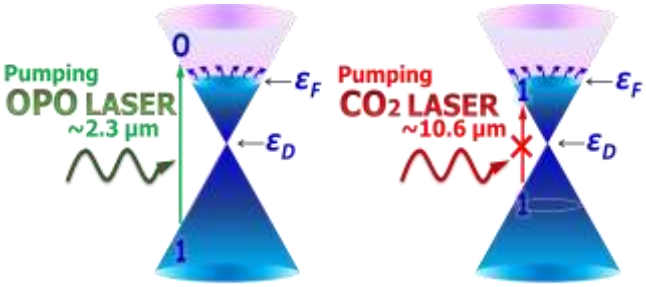



**Figure 3:** Relationship between graphene energy band and photon energy of pumping laser. OPO laser excites electron due to its high photon energy, $>2\varepsilon_F$.
On the other hand, No excitation occur with CO2 laser because its photon energy is less than $2\varepsilon_F$.