Amplified Stimulated Terahertz Emission from Optically Pumped Graphene at Room Temperature

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We report on fast relaxation and relatively slow recombination dynamic of photogenerated electrons/holes in an exfoliated graphene. Under suitable pumping the carriers dynamics can lead to non trivial feature such as negative dynamic conductivity in terahertz spectral range. Therefore we conduct time domain spectroscopy studies and shows that graphene sheet amplifies an incoming terahertz field. The graphene emission spectra dependency on the laser pumping power shows a threshold like behavior, testifying the occurrence of the negative conductivity and the population inversion in terahertz range, paving the way for a new class room temperature graphene based terahertz lasers.

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Graphene is a one-atom-thick planar sheet of carbon atoms that are densely packed in a honeycomb crystal lattice.[1] This material has many peculiar properties and potential applications. For example, the prediction and observation of half-integer quantum Hall effect,[2] finite conductivity at zero charge carrier concentration,[3] perfect quantum tunneling effect,[3] ultrahigh carrier mobility,[3] including massless and gap less energy spectra. The gap less and linear energy spectra of electrons and holes lead to nontrivial features such as negative dynamic conductivity in the terahertz (THz) spectral range,[4] which may lead to the development of a new type of THz laser.[5,6] This perspective generate an intense interest due to the ongoing search for viable THz detectors and emitters.

To realize such THz graphene-based devices, understanding the non-equilibrium carrier relaxation/recombination dynamics is critical. Recently, time-resolved measurements of fast non-equilibrium carrier relaxation dynamics have been carried out for multilayer and monolayers of graphene that were epitaxially grown on SiC[7,8] and exfoliated from highly oriented pyrolytic graphite (HOPG).[9,10] Several methods for observing the relaxation processes have been reported. Dawlaty et al.[10] and Sun et al.[11] used an optical-pump/THz-probe technique and George et al.[11] used an optical-pump/optical-probe technique. The measured optical phonon lifetimes found in these studies were ~ 7 ps[11,14], 2-2.5 ps[13], and ~ 1 ps[11], respectively, some of which agreed fairly well with theoretical calculations by Bonini et al.[12]. A recent study by Breusing et al.[15] more precisely revealed ultrafast carrier dynamics with a time resolution of 10 fs for exfoliated graphene and graphite.

In this paper we report on the fast relaxation and relatively slow recombination dynamics in optically-pumped graphene. The recombination process is stimulated with an THz photon probe ~ 2 ps and ~ 3.5 ps after the intraband carrier relaxation has started. The observed results suggest the occurrence of negative dynamic conductivity in the THz spectral range.

When graphene is pumped with the infrared photon having an energy hω, electrons/holes are photogenerated via interband transitions. It has been shown that the intraband carrier first establishes separate distributions around the level εf ± hω/2 (εf : Fermi energy ) within 20-30 fs after excitation (see Fig 1a). At room temperature and/or strong pumping, collective excitations due to the cc scattering, e.g., intraband plasmons should have a strong influence on the carrier relaxation dynamics. As discussed in[11,14,15] the quasi-equilibrium distributions at around εf ± hω/2 is rapidly redistributed within 200-300 fs (Fig 1b). Then optical phonons (OPs) are emitted on the high-energy tail of the electron and hole distributions on a few picoseconds time scale(Fig 1c). This intraband relaxation process is relatively fast and accumulates the nonequilibrium carriers around the
0.8 eV (1550 nm), much higher than the optical phonon energy (wavelength) is carefully selected to be around

laxation/recombination dynamics, the pumping photon emissions from the above-mentioned carrier re-

layers and few layers of graphene. To obtain the THz cm x 2.5 cm size one have some islets of monolayers, bi-

of 300 nm of thickness. On this substrate of about 2.5

about 500-560 µm SiO

geometry [20]. The sample used is exfoliated graphene on

an electro-optic (EO) sampling method in total-reflection
time-resolved field emission properties are measured by
an optical pump/THz-and-optical-probe technique. The
conduct time-domain spectroscopy experiments based on
pumping intensity is sufficiently high.

if the pumping photon energy is suitably chosen and the
emissions over a wide THz frequency range are expected
due to the gapless symmetrical band structure, photon
states effects and Pauli blocking lead to the population
fast intraband relaxation (ps or less) and the interband
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Dirac point. The recombination process are mainly done
by interband cc and carrier-phonons (cp) scattering. The
fast intraband relaxation (ps or less) and the interband
recombination process (≫ 1 ps) slowed by the density of

states effects and Pauli blocking lead to the population
inversion [21]. In the case of the radiative recombination,
due to the gapless symmetrical band structure, photon
emissions over a wide THz frequency range are expected
if the pumping photon energy is suitably chosen and the
pumping intensity is sufficiently high.

In order to verify the above mentioned concept we
conduct time-domain spectroscopy experiments based on
an optical pump/THz-and-optical-probe technique. The
time-resolved field emission properties are measured by
an electro-optic (EO) sampling method in total-reflection
geometry [20]. The sample used is exfoliated graphene on
SiO2/Si substrate . The sample structure as presented
in figure 1 is made of one layer of Si <100> oriented of
about 500-500 µm of thickness and 10−3. 5 ∗ 10−3 Ωcm
of resistivity; following by an thermal dry layer of SiO2
of 300 nm of thickness. On this substrate of about 2.5
cm x 2.5 cm size one have some islets of monolayers, bi-
layers and few layers of graphene. To obtain the THz
photon emissions from the above-mentioned carrier re-

laxation/recombination dynamics, the pumping photon
energy (wavelength) is carefully selected to be around
0.8 eV (1550 nm), much higher than the optical phonon
energy (∼ 0.2 eV). The graphene sample is placed on
the stage and CdTe crystals of about 2 mm long and 0.5
mm large are used as THz electro-optic transceivers and
placed onto the sample. A femtosecond pulsed fiber laser
with full width at half-maximum of 80 fs, frequency of
20 MHz and average power of about 4 mW was used as
the pumping source. More precise description of the ex-
perimental set-up can be see in Refs [21, 22]. The laser
is split into two path used for pump and probe. The
pumping laser beam linearly polarized is mechanically
chopped at ∼ 1.2 KHz (for lock’in detection), and si-
multaneously focused with a beam diameter of about 40
µm onto the sample and the CdTe from the back side,
while the probing beam is cross polarized with respect to
the pump beam and focused from the top side (see Fig
1). Owing to second-order nonlinear optical effects, the
CdTe crystal can rectify the pump laser pulse to emit
THz envelope radiation. This THz pulse is used for EO
detection via pockel effect . The same THz pulse is partial-
ly reflected at the top surface of the CdTe then subject
back to the graphene, working as the THz probe pulse
(arrowed blue line in Fig 1b) to stimulate THz photon
emission via electron-hole recombination in the graphene
(see Fig 1). Therefore the original data of experimen-
tal temporal response consist of the first forward propa-
gating THz pulsation (no interaction with graphene) fol-
lowed a photon echo signal (probing the graphene). The
delay between these two pulsation is given by the total
round-trip propagation time of the THz pulse through
the CdTe. The system bandwidth is estimated to be
around 6 THz, mainly limited by the Reststrahlen band
of the CdTe sensor crystal.

The experiments were done with two CdTe crystal
transceivers (A and B): (100) and (101) oriented respec-
tively and of about 120 µm and 80 µm of thickness re-
spectively. The graphene monolayer area investigated is
of about 7000 µm².

Figure 2 shows temporal responses measured on monol-
layer graphene with the thinner (black curve) and the
Thicker crystal (red curve) for the pumping pulse inten-
sity of about 3 ∗ 10⁷ W/cm². These curves was plot with
the same origin for comparison. One can notice that,
as predicted each temporal profile is composed of two
peaks from optical rectification (OR) in CdTe and the
THz photon echo signal. The measured times delays be-
tween these two pulsations with crystal A (thinner) and
crystal B (Thicker) of around 2 ps and 3.5 ps respectively
are in a good agreement with the round trip propagation
time of THz pulse through the CdTe crystal. The refrac-
tive indexes of CdTe may be obtained from [22]. The OR
pulse is found be broader in crystal B compare to that
one of crystal A, this is the consequence of better phase-
matching conditions in thin crystal. Indeed the coherent
length in CdTe is estimated to be around 100 µm at 0.8
eV [23, 24]. This coherent length is maximum at 1050
nm [20]. The inset of figure 2 present the echo signal
peak measured with crystal B on graphene as well as an
reference curve (grey curve) measured on the area with-
out graphene. One can notice that the peak obtained on
graphene is more intense than that one obtained on
the substrate. This suggests that graphene amplifies the
THz echo (THz probe) signal. It is thought that this THz
pulse stimulate emission from graphene amplifying the
incoming echo photon by photoelectron/hole recombi-
nation in the range of the negative dynamic conductivity.

The graphene transfer function $H(\omega)$ is defined like
$H(\omega) = Y(\omega)/X(\omega)$ where $Y(\omega)$ and $X(\omega)$ are the Fourier
transform of the second peak measured on graphene and
substrate respectively.

Figure 3 shows the transfer function of monolayer
graphene for different values of pumping pulse inten-
sity. One can see from these results that decreasing
$I_{\text{pump}}$ drastically reduce the emission spectra and below 5 * 10^6 W/cm^2 the emission completely disappear and only attenuation can be seen. We also present in the inset of figure 3 (lower panel) the corresponding ratio $A_G/A_S$ where $A_G$ and $A_S$ are the amplitude of echo signal peak measured on graphene and substrate respectively. The graphene transfer function and $A_G/A_S$ shows a clear threshold like behavior testifying the occurrence of the negative conductivity and population inversion in optically pumped graphene. The threshold intensity if found to be around 10^7 W/cm^2. This is a good starting point for the realization of room temperature THz lasers based on graphene. The inset of figure 3 (upper panel) present the normalized EOS signal amplitude of the first peak (see figure 2) for different values of the pumping pulse intensities. Inset : Temporal responses of echo photon signal measured on graphene (red line) and on the area without graphene (grey line).

Figure 4 shows the Fourier transform of the photon echo signal measured on the area without graphene (reference) with the crystal A (black curve) and the crystal B (red curve). The normalized normalized dynamic conductivity for the pumping pulse intensity three time higher than the threshold pumping pulse intensity at 300 K is also presented (see [6, 22]). The photon echo pulse stimulate the recombination process and the emission of THz photon in graphene within the negative dynamics conductivity area (blue shaded area). The expected graphene emission spectra bandwidth is limited at lower frequencies by the drude mechanism of THz absorption and the higher frequency limit is given by the system bandwidth which is estimated to be around 6 THz (see the horizontal solid lines in fig 4). The black and red shaded area shows the expected graphene emission bandwidth, from ~1 THz to ~5 THz using the thicker crystal and from ~1 THz to ~6 THz using the thinner crystal. The broader spectra with thin crystal is due to the better phase-matching conditions.

The inset of figure 2 present the transfer function of monolayer graphene obtained with crystal A (black line) and crystal B (red line). The obtained spectra is in good agreement with the above mentioned expectations. It is possible to compare these spectra within the smallest

**Figure 2.** Measured temporal responses on monolayer graphene with thicker (red line) and thinner (black line) CdTe crystals for the pumping pulse intensity of about 3 * 10^7 W/cm^2. Inset : Temporal responses of echo photon signal measured on graphene (red line) and on the area without graphene (grey line).

**Figure 3.** Transfer functions of monolayer graphene for different values of pumping pulse intensities. Inset : Normalized EOS signal amplitude of the first peak (upper panel) and $A_G/A_S$ ratio where $A_G$ and $A_S$ are the amplitude of echo signal peak measured on graphene and substrate respectively (lower panel) for different values of the pumping pulse intensities.

**Figure 4.** Shows the Fourier transform of the photon echo signal measured on the area without graphene (reference) with the crystal A (black curve) and the crystal B (red curve). The normalized normalized dynamic conductivity for the pumping pulse intensity three time higher than the threshold pumping pulse intensity at 300 K is also presented. The photon echo pulse stimulate the recombination process and the emission of THz photon in graphene within the negative dynamics conductivity area (blue shaded area). The expected graphene emission spectra bandwidth is limited at lower frequencies by the drude mechanism of THz absorption and the higher frequency limit is given by the system bandwidth which is estimated to be around 6 THz (see the horizontal solid lines in fig 4). The black and red shaded area shows the expected graphene emission bandwidth, from ~1 THz to ~5 THz using the thicker crystal and from ~1 THz to ~6 THz using the thinner crystal. The broader spectra with thin crystal is due to the better phase-matching conditions.
Since the measurements are taken as an average, the observed response is undoubtedly a coherent process that cannot be obtained via spontaneous emission processes, providing clear evidence of stimulated emission. The above results and discussions confirm that the THz emission from graphene is stimulated by the coherent THz probe radiation, also that the THz emission is amplified via photoelectron/hole recombination in the range of the negative dynamic conductivity.

The occurrence of population inversion in optically pumped graphene has been theoretically reported [6, 27]. In the room temperature and/or strong pumping case, threshold pumping pulse intensity was predicted to be between $10^8$ W/cm$^2$ and $10^9$ W/cm$^2$ [27], in good agreement with our experimental results.

In conclusion, we have successfully observed coherent amplified stimulated THz emissions arising from the fast relaxation and relatively slow recombination dynamics of photogenerated electrons/holes in an exfoliated graphene. The results provide evidence of the occurrence of negative dynamic conductivity, which can be applied to a new type of THz laser.

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