2D Materials

PAPER

Terahertz wave generation and detection in double-graphene layered van der Waals heterostructures

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Abstract

We report on the first experimental observation of terahertz emission and detection in a double graphene layered (GL) heterostructure which comprises a thin hexagonal-boron nitride tunnel-barrier layer sandwiched between two separately contacted GLs. Inter-GL population inversion is induced by electrically biasing the structure. Resonant tunneling and negative differential resistance is expected when the two graphene band structures are perfectly aligned. However, in the case of small misalignments we demonstrate that the photon-absorption/emission-assisted non-resonant- and resonant-tunneling causes all excess charges in the n-type GL to recombine with the holes in the p-type GL giving rise to an increased measured dc current. This work highlights a novel strategy for the realization of efficient voltage-tunable terahertz emitters and detectors.

1. Introduction

Graphene, because of its well-known unique characteristics, is a captivating material for many applications ranging from flexible displays [1, 2] photodetectors (PD) [3], ultrafast lasers [4], plasmonics oscillators [5] as well as optical modulators [6]. In particular, applications of graphene field effect transistors (GFET) operating in the terahertz (THz) range are appealing as it is one of the least explored frequency region and holds potential to revolutionize the fields of security, medical imaging, chemical sensing and high-speed wireless communication [7]. Recent research on vertically stacked heterostructures of graphene with hexagonal-boron nitride (h-BN) and other two-dimensional materials has opened a new realm for intriguing device physics making them ideal candidates for future high-frequency technology [8–12].

Here, we report on the fabrication and first experimental observation of THZ emission and detection in the DGL heterostructures based on a thin h-BN tunnel-barrier sandwiched between two monolayers of graphene. The measured relative intensity of the emission spectra shows a clear gate bias dependence with an estimated emission power around 7 pW μm⁻². In the detection regime our device yielded measured responsivity of 1.55 A W⁻¹ at 1 THz and 300 K. These results reasonably agree with some important aspects of the theoretical model reported in the recent work by Ryzhii et al. [19–22] allowing us to attribute the observed THZ emission (detection) to the photon-assisted resonant (non-resonant) radiative inter-GL transitions. These results have shed light on one of the path that might lead to the realization of new, compact, tunable, coherent, and room-temperature operating THz sources and detectors.

2. Device description and principles of operation

The fabrication of the proposed DGL heterostructure involves micromechanical cleavage and transfer of tiny
flakes of graphene and h-BN on a Si/SiO₂ substrate. The fabrication process includes first transfer of a relatively thick h-BN layer (∼50 nm) on top of an oxidized Si wafer used as the back gate. This first layer of h-BN acts as an atomically flat substrate on which the active part of the device is mounted and ensures high mobility in the GLs. The h-BN flakes used in this work were exfoliated from ultra-pure h-BN single bulk crystal. On this high-quality, atomically flat h-BN buffer layer, an exfoliated monolayer of graphene was transferred carefully using the standard dry transfer process and patterned using electron-beam lithography and oxygen plasma etching to define the contact bars. Next, a relatively thin layer of h-BN (around 2–7 atomic layers) was transferred. This thin layer served as the tunnel barrier. The second GL was then transferred on top of the thin h-BN layer. Lastly, Ti/Au contacts were fabricated on both GLs by using e-beam lithography, evaporation, and lift-off processes. The schematic of the device is shown in figure 1(a) while figures 1(b) and (c) depict the SEM images of the fabricated device. One can see the position of source and drain electrodes (light gray), the active area of the device (dark gray) and the SiO₂ substrate (black) in figure 1(b). The line-like structure in between ‘source’ and ‘drain’ contacts, is the active area of the device containing overlapped graphene monolayers. This SEM image shows a top view of the device consisting of Si/SiO₂/h-BN/graphene/h-BN/Graphene. Being a top view the stacked structure cannot be seen in this image. A zoomed image in 1(c) shows the active area of the device to be around 1.4–1.5 μm wide.

Relative crystallographic orientation between the two GLs is vital to get resonant transitions from the device. Appearance of negative differential conductance (NDC) in the current–voltage (I–V) characteristics is one indication of how well the two GLs are aligned [23, 24]. In order to observe the NDC at low enough energies (or gate voltages) the two layers have to be aligned so that their crystallographic axes are parallel. Since most of the flakes are exfoliated along the crystallographic directions a straight-line edge of the 2nd GL flake was aligned to meet that of the 1st GL flake during the transfer process [25]. For aligning the edge of a flake in our devices, the substrate stage was rotated to match the line of the edge. With microzoom-scope eye and assistance from the computer, the best relative alignment possible is around ∼1° [23]. There are two possibilities of the straight-line edges; one is ‘zig-zag’ and the other is ‘armchair,’ but they cannot be distinguished well via optical microzoom-scope observation [26]. When both the top and bottom GLs are aligned with the zig-zag edges or armchair edges, the misalignment could be very small (∼1° or less), but the cases of one side with the zig-zag and the other with the armchair give rise to large misalignment (∼30°). In this work, two devices were fabricated independently by using the aforementioned procedure. Both the devices have slight mis-alignment (∼4°) between the crystallographic axes of the two GLs, while differing in terms of thickness of h-BN tunnel barrier layer, ∼3 nm in device 1 and ∼2 nm in the case of device 2. The device 1 is used for emission studies and device 2 for the THz detection studies. Some of the other fabricated samples have small mis-alignment ∼1° (see supplementary information for details). However, a recent report by Kim et al [27] showcases another technique to have highly accurate layer alignment by carefully picking the monolayers by a hemispherical epoxy handle, which should be introduced in future study.

The physical mechanism behind the device operation is the photon assisted tunneling transitions. The

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**Figure 1.** (a) The schematic of the fabricated DGL device. (b) SEM image of the device showing the position of source and drain electrodes (light gray), the active area of the device (dark gray) and the SiO₂ substrate (black) when viewed from top. (c) A zoomed SEM image showing active area of the device to be 1.4–1.5 μm wide.
bias voltage ($V$) applied between the GL’s contacts induces the electron gas in one GL and the hole gas in the other GL, forming a DGL capacitor, displacing the chemical potentials between the GLs. The electron and hole densities as well as the chemical potentials in the GLs are also modulated by the gate voltage ($V_g$).

Figures 2(a) and (b) schematize the device operation mechanism and show the inter-GL valence band to valence band ($V$–$V$) and conduction band to conduction band ($C$–$C$) resonant-tunneling.

When the Dirac points in the two GLs are aligned by applying $V$ and $V_g$ pertinently, the system is in resonance and the energy and momentum conservations are satisfied during the tunneling. This pronouncedly increases and maximizes the probability of quantum mechanical resonant tunneling (QM-RT) between the two GLs. Further application of bias $V$ leads to bands misalignment and the reduction of tunneling current, producing the NDC shape in the $I$–$V$ characteristics. Nevertheless, when the system is slightly off resonance with a bands-offset energy $\Delta$ in THz and IR range, TM polarized photons with energy $\hbar \omega \sim \Delta$ can mediate the inter-GL resonant tunneling between the p-type and the n-type GLs (see figures 2(a) and (b)). Therefore, in this case the device operation principle is associated with the absorption/emission of the TM polarized photon with energy $\hbar \omega \sim \Delta$ accompanied by the electron tunneling transitions between the GLs and causing the electric terminal current. The inter-GL radiative transitions with the absorption (figure 2(a)) or emission (figure 2(b)) of photons conserve the electron lateral momentum and are referred to as photon assisted quantum mechanical resonant tunneling (PA-RT).

The energies of the photons emitted or absorbed by the structure in the inter-GL resonant-tunneling (RT) transitions is determined by the voltage dependent band-offset energy $\Delta$ in THz and IR range. The calculation result for the bias voltage dependent band-offset energy $\Delta$ of the DGL device having a 3 nm thick $\text{h-BN}$ tunnel barrier layer. $\kappa = 4$ is the permittivity of the h-BN layer.

**Figure 2.** Band diagrams of the DGL emitter/detector structures with (a) photo-absorption-assisted inter-GL, and (b) photo-emission-assisted inter-GL radiative transitions. Wavy arrows indicate the inter-GL radiative $V$–$V$ and $C$–$C$ transitions both in devices (a) and (b). The inter-GL transitions work for the TM-mode THz photon radiations in our structures. (c) The calculation result for the bias voltage dependent band-offset energy $\Delta$ of the DGL device having a 3 nm thick h-BN tunnel barrier layer. $\kappa = 4$ is the permittivity of the h-BN layer.
tunneling inter-GL transition is lower than the initial state ($\Delta > 0$), the tunneling is associated with emission of the photons (see also figure 2(b)) whereas when $\Delta < 0$, the tunneling is associated with absorption of the photons (see also figure 2(a)). The resonant tunneling causes all excess charges in the n-type GL to recombine with the holes in the p-type GL. Our recent study reveals the possibility of plasmon-assisted interlayer resonant/non-resonant tunneling as another mechanism of producing net gain in the THz range [15].

3. Experimental methods

The detection experiments were carried out with our dedicated setup. It involves a uni-traveling-carrier photodiode (UTC-PD) module as a THz source. The UTC-PD works as a THz photomixer providing tunable output in the THz range (300 GHz–1 THz) in response to two continuous-wave (CW) laser beam inputs whose difference in frequencies are tuned to be equal to the output frequency. In case of 1 THz with 1 μW output power, the wavelengths of the two CW laser beams were set to be 1540.000 nm and 1547.946 nm, respectively, and their intensities were optically amplified to a level of 35 mW. Then the beams were coupled and fed to the UTC-PD. The THz photons were guided onto the active area of the DGL device through a set of parabolic and indium tin oxide mirrors. The gate and drain terminals were connected to a semiconductor parameter analyzer (Keysight: Model B1500A) while the source terminal was grounded. The dc current–voltage ($I$–$V$) characteristics of the DGL device were recorded for different $V_g$ values. The dc photocurrent–voltage characteristics were obtained by subtracting the measured $I$–$V$ curves without the THz irradiation from the ones with the THz irradiation. The experiment was performed at room temperature in oblique incidence geometry as shown in figure 3(a) to yield the THz electric field perpendicular to the GLs (TM polarized field) and thus the tunneling transitions associated with the absorption of the THz photons [28].

The emission experiments were carried out using Fourier-transformed far-infrared spectroscopy at 100 K and 300 K. The sample was placed in the vacuum chamber and the biases were controlled using voltage/current source-meters (Keithley 2400) while the radiation intensity was measured using a 4.2 K-cooled silicon bolometer. To carry out the experiments, we first measured the background spectrum, i.e. the spectrum with unbiased sample. Then we measured bias dependent spectra with current flowing through the sample and normalized it to the background spectra to eliminate the undesired effects arising due to static blackbody emission and any spectral artifacts emitted from the elements inside the spectrometer. Since the emitted TM polarized photons exit the sample from the side edges (see figure 3(b)), a set of metal prisms was used to collect and direct the emitted radiation towards the detector as shown in figure 3(c).

4. Results and discussions

Figure 4 depicts the measured $I$–$V$ curves for device 1 exhibiting no traces of the NDC at room temperature (figure 4(a)) but weak NDC traces at 100 K (figure 4(b)). Under the conditions that the weak NDCs are obtained, the bias voltage conditions allow the inter-GL population inversion and photon-assisted RT radiative transitions between the conduction bands and the valence bands of the two GLs. To verify such an expected emission of THz radiation, we measured emission spectra of the device by using the FT-IR setup as described in section 3 for different gate bias conditions at a fixed drain voltage ($V_d = 0$ V). Figure 5 depicts emission spectra of device 1 at 300 K (a) and at 100 K (b). Figure 5(a) shows no signs of increased THz emission beyond the background blackbody radiation as expected given the absence of NDCs in $I$–$V$ curves at 300 K (see figure 4(a)). On the contrary, when the sample is cooled down to 100 K, one can see broadband THz emission from the DGL structure under specific gate bias conditions as depicted in figure 5(b). The measured spectra show increasing emission intensity with increasing negative gate bias voltages while no emission is observed for positive gate voltages. This reasonably agrees with the calculations shown in figure 2(c) based on the theoretical prediction by Ryzhii V et al [21].
The device exhibits relatively broad emission ranging from 2 to 6 THz, peaking around 3.1 THz, likely due to a weak and rather broad NDC in this sample at 100 K as shown in figure 4(b). The linewidth of the emission spectra reflects the resonant tunneling quality factor $Q$ which correlates with the carrier momentum relaxation time, rotational misalignment and non-uniform tunneling due to inhomogeneities present in GLs [23, 29]. The peak position is determined by $\Delta$ which is a function of $V$ and $V_g$. This means that the emission frequencies from the DGL device is expected to be voltage tunable when the sample shows pronounced gate tunable NDC peaks in the $I$–$V$. The integrated emission intensity of device 1 in the 2–6 THz frequency range was estimated to be $7 \text{ pW} \mu\text{m}^{-2}$ at $V_g = -4 \text{ V}$. This estimation is based on the calculation of the black body radiated power at 100 K which serves as reference and multiplied by the peak values of intensity of normalized emission curve at a particular gate voltage. Recent paper has reported thermal emission from GFET of the order of 2.1 nW in 1–3 THz range by using a double-patch antenna along with a silicon lens attachment [30]. Our DGL source hold potentialities to emit THz with much higher power given a pronounced and stronger NDC (figure S1(b) in supplementary information) and coupling device structure. The observed results might include in part the effect of plasmon-assisted resonant tunneling, which could be performed under inter-GL misaligned conditions [13].

For biasing conditions when $\Delta < 0$, the RT is preceded by an absorption of photons. When the NDC cannot be obtained but only thermionic positive conductance is obtained as seen in $I$–$V$ curves of device 2 at 300 K (figure 6(a)), the tunneling process takes place in a non-resonant manner so that the photon-assisted tunneling may take place in a broadband nature with the incident photon energy. The detection experimental results are shown in figure 6(b), depicting the incoming radiation induced tunneling photocurrent versus DGL bias voltages $V$ for different $V_g$ levels under 1 THz photon irradiation at 300 K. The photocurrent increases with increasing $V$ both positively and negatively. The increase of the DGL photocurrent with the positive gate biases is more likely due to the non-resonant tunneling associated with absorption of THz photons. This nonlinear monotonic increase comes from the quadratic rectification whose responsivity...
corresponds to the nonlinearity of the I–V curve in figure 6(a). The prior works of Vasko [31] and Ryzhii et al [14, 32] give theoretical support for the photo-absorption-assisted resonant/non-resonant tunneling and the resultant tunneling current in the DGL. They also provide thorough discussion on effects of applied gate bias on sensitivity of these devices. The several peaks superposed onto the monotonic nonlinear gate bias on sensitivity of these devices. The several peaks are not essentially arising from the device itself and not resonant peaks neither but most likely fluctuations caused by fluctuation in power of the THz emitter used for detection experiments and environmental noise. Since the photo-current is of the order of pico-amperes, the sources of noise mentioned above affects very much the experiments and results.

The absorption of incident radiation is generally limited to the fermi surface charge carriers in standard semiconductors. For instance, in the case of lateral GFET [35], only the electrons in top surface of graphene have freedom to receive the incident energy and contribute to the photo-generated current, thus limiting the sensitivity of such devices. However, in our case, current flows vertically down from one GL to the other counterpart. Not only the electrons present at fermi surface but all the excess charge carriers in top GL can absorb the incoming radiation and thermionically tunnel to the other GL leading to very high photocurrents. We quantify the photocurrent generated by the 1 μW incoming THz irradiation in terms of photoresponsivity. Intrinsic responsivity for a detector can be defined as:

$$R_j = \frac{S_j \Delta j}{S_{P\mu} \sin \theta J^2},$$

where $S_j$ is THz beam spot size ($S_j = 2.83 \text{ mm}^2$), $S_{P\mu}$ is DGL active area ($S_{P\mu} = 54.75 \text{ \mu m}^2$), $\Delta j$ is maximum induced photocurrent ($\Delta j = 15 \text{ pA}$, see figure 6(b)), $\theta$ is THz incident angle ($\theta = 45^\circ$, see figure 3(a)), and $P_{\text{in}}$ is THz input power ($P_{\text{in}} = 1 \mu\text{W}$). It is assumed in the above definition that all the incoming THz power is coupled effectively to the device. However, due to absence of any antenna structure in this case, only a small fraction of incident radiation is channeled onto the device. Hence, this value of responsivity shall be considered as a lower limit.

Using equation (1), we obtained $R_j \sim 1.55 \text{ A W}^{-1}$ for non-resonant detection at room temperature. In the case of resonant tunneling, the current flowing through the device is about $10^3$ times larger than the non-resonant case (see $I–V$ characteristics with NDCs in figure S1(b) in supplementary information, compare to figure S1(a)). Since the incoming THz field is expected to induce resonant tunneling in a well aligned sample, three orders larger $\Delta j$ can be expect in the resonant case. Responsivity is directly proportional to photocurrent $\Delta j$, its considerably larger value in the resonant case will likely result in drastic enhancement of responsivity. Indeed, a theoretical work by Rodriguez [34] also predict five orders higher responsivity ($3 \times 10^5 \text{ V W}^{-1}$ in samples showing NDCs) in resonant detection case compared to the non-resonant (almost zero for samples without NDCs).

Our detectors hold great potentialities compared to other graphene based THz photodetectors. Indeed, graphene FET based THz detectors operating: on photothermoelectric effects; over-damped plasma waves excitation; and on both thermoelectric/plasmonic effects were recently reported with responsivity $\sim 5 \text{ nA W}^{-1}$ [35], 4.7 V W$^{-1}$ [30] and 10 V W$^{-1}$ [36]; $\sim 1.3 \text{ mA W}^{-1}$ [37]; 0.25 V W$^{-1}$ [38] respectively. Our detector shows higher responsivity even in the non-resonant detection regime and is promised to about three orders of magnitude higher sensitivity in the resonant mode. The next step of this work to observe the resonant THz detection in the DGL device with a small crystallographic misalignment between the GLs (figure S1(a) in supplementary information) will be a subject of future publication. Introduction of an antenna structure such as a grating gate structure proposed by Ryzhii et al [14] or patch antenna and Si lens attached to the device would greatly
improve the responsivity as experimentally reported by Tong et al [30].

5. Conclusion

We fabricated a DGL van der Waals heterostructure device and experimentally observed its response to the THz radiation via non-resonant tunneling between the GLs. Since the device holds an undesired crystallographic rotational misalignment between the GLs, no NDCs was observed at room temperature. Correspondingly, non-resonant photoresponse was observed at room temperature in response to 1 THz photon irradiation, demonstrating a high responsivity of 1.55 A W⁻¹. In one of our devices we observed weak NDCs at 100 K, giving a broadband emission (2–6 THz) with an intensity 7 pW μm⁻². The emission was observed only when the band-offset levels between the GLs were set in negative so that the photoemission-assisted resonant tunneling can take place. These results are the first demonstration proving experimentally that the gated DGL devices based on the photon-assisted resonant radiative inter-GL transitions can be exploited to create highly efficient THz sources and THz photo detectors.

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