Photoelectric polarization-sensitive broadband photoresponse from interface junction states in graphene

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Keywords: graphene interface junctions, polarization sensitive photoresponse, interface junction states

Supplementary material for this article is available online

Abstract
Graphene has established itself as a promising optoelectronic material. Many details of the photoresponse (PR) mechanisms in graphene in the THz-to-visible range have been revealed, however, new intricacies continue to emerge. Interface junctions, formed at the boundaries between parts of graphene with different number of layers or different stacking orders, and making connection between electrical contacts, provide another peculiar setup to establish PR. Here, we experimentally demonstrate an enhanced polarization sensitive photoelectric PR in graphene sheets containing interface junctions as compared to homogenous graphene sheets in the visible, infrared, and THz spectral regions. Our numerical simulations show that highly localized electronic states are created at the interface junctions, and these states exhibit a unique energy spectrum and enhanced probabilities for optical transitions. The interaction of electrons from interface junction states with electromagnetic fields generates a polarization-sensitive PR that is maximal for the polarization direction perpendicular to the junction interface.

Introduction

The termination of an otherwise perfect crystal by a surface interface leads to the formation of interface electronic states. Examples of such states include Shockley or Tamm electron states [1, 2], the edge states formed in the quantum Hall regime [3, 4], the electronic states found at the surface of topological insulators [5–7], Shockley–Tamm-like states in so-called ‘photonic graphene’ [8], and Tamm states in plasmonic systems [9]. These electronic states are highly localized in the direction perpendicular to the boundary but a one-dimensional energy band is formed along the boundary. In many cases, e.g., in the quantum Hall regime [3, 4], or in topological insulators [5–7], the presence of such electronic states may result in a great enhancement of the electronic transport.

Here, we probe the existence of a different type of localized interface state; the junction states occurring at the 1D interface between heterogeneous regions of two-dimensional materials. Specifically, we present experiments on suspended graphene bilayer-trilayer and trilayer-tetralayer junctions showing a photoelectric response in the broad spectral region from the THz to the visible. Moreover, a strong polarization dependence of both the photocurrent and photo-voltage has been experimentally measured. We attribute the enhancement of the photoresponse (PR) to the interface junction electronic states. To explain our results we developed a model where the interface junction states have one-dimensional electron band structure and the transitions between these bands determine interaction of electrons with applied electromagnetic radiation. The inter-band absorption...
of linearly-polarized electromagnetic radiation crucially depends on the mutual orientation of electron wavevector \( k \) and the polarization orientation of electric field \( E \). The interface junction electron states form a one-dimensional electron localization channel aligned along the interface junction boundary. This restricts propagation of electrons in other directions and, as the result, provides the selected direction for \( k \) along the direction of interface junction. The existence of selected direction of \( k \) prevents averaging of PR contributions from different mutual configurations of \( k \) and \( E \) and, therefore, leads to the strong polarization dependence of the PR. Our explanation is supported by numerical simulations of the electronic bandstructure, the optical absorption of our interface junctions, and by analysis of polarization-sensitive photocurrent flowing along the junction [10–14]. The polarization-dependent photocurrent and photovoltage are generated in suspended devices via rectification due to the potential step at the interface between suspended and non-suspended parts of graphene flakes [14].

This work began as an attempt to explore the physics of long-wavelength infrared (IR) PR [15–22] in suspended graphene field effect transistors [23]. The previous investigations of PR mechanisms in graphene led to the development of a wide spectrum of graphene photodetectors that cover the wide range of wavelengths from GHz/THz to Visible. Some notable implementations are photovoltaic hot electron and photothermoelectric detectors [16, 18, 24–27], bolometric and cyclotron resonance quantum Hall effect graphene detectors [15], hot-electron bolometers [16], and nonlinear mixing detectors [17]. Many efforts were focused on investigations of PR in devices with p–n junctions or potential barriers created in graphene either by inhomogeneities of electrostatic potential [24–26], or by direct doping of graphene [27]. In our work, we investigated devices with homogeneous graphene flakes, and devices with graphene flakes having varying thickness resulting in interface junctions.

There have been reports of interface junction PRs in graphene. The work reported in [28] sheds light on the photothermal mechanism of PR in graphene, and demonstrated the enhanced photosensitivity of the interface junction boundary compared to the homogeneous parts of graphene flake. The authors of a more recent work [29] investigated polarization sensitivity of graphene at metal–graphene and single layer–bilayer interface junctions, and did not observe polarization sensitivity for interface junctions at the room temperature. In contrast to the [29], at temperatures below 250 K, in inhomogeneous samples we found a polarization dependence and increased broadband responsivity related to the presence of interface junctions.

### Experimental setup

In our work, few-layer graphene sheets are exfoliated, placed over pre-fabricated 2 \( \mu m \) wide 150 nm deep trenches in \( \text{SiO}_2 \) layers on Si substrates (appearing in optical images as stripes with color slightly different from that for the other parts of substrate surface) and coupled to electrodes and antennas via metal deposition through shadow masks [30]. The devices are annealed [31] in order to improve their quality by heating them with current in vacuum after mounting them in the measurement cryostat. A typical device is shown in figure 1(a). The Si substrate is used as a backgate to change the effective doping in the graphene flake, and metallic antennas (appearing white in figure 1(a)) designed to couple THz fields to the graphene flakes also act as source S and drain D electrodes. In contrast to previous works, our interface junctions were also directly attached to the source and drain electrodes.

In IR and visible frequency regimes the PR is determined by direct illumination of the graphene flake as the antenna coupling is inefficient beyond THz frequencies.

Each device is mapped by optical microscopy and Raman spectroscopy to determine the position and orientation of any interface junctions, the numbers of layers at each position, and the stacking order (see supplementary data for details). For example, for Device #1 shown in figure 1(a), the interface junction is highlighted with a red dashed line, and the flake edges are marked with blue dashed lines for better visibility. The device configurations for all the devices tested are summarized in table 1. Device #2, which has a homogeneous ABA-stacked trilayer flake without an interface, was included as a reference sample.

PR was measured as the photovoltage signal, i.e. the photo-induced change of source–drain voltage, \( V_{\text{ph}} \), for unbiased devices. The PR measurements were performed at different temperatures \( T \) in the range from 8 to 300 K, using several laser sources with different wavelengths ranging from THz to visible: a CO2-pumped far infrared (FIR) laser for 70.6 \( \mu m \) (4.25 THz) and 120 \( \mu m \) (2.5 THz), a mid-infrared (MIR) CO2 laser for 10.6 and 9.3 \( \mu m \), and a diode laser for 780 nm wavelengths. Linearly polarized light was used and careful pointing and focusing control was taken to keep the spot size and position the same (for a given wavelength) throughout the measurements to eliminate possible artifacts due to photo-thermal effects (see supplementary data for details).

The schematic of a typical device geometry and measurement setup is shown in figure 1. In the majority of experiments, the applied radiation was linearly polarized, and the direction of polarization was controlled using linear polarizers. The direction of polarization (direction of the incident electric field orientation, \( E \)) is described using the polarization...
orientation angle, $\alpha$, between the horizontal axis and the actual direction of polarization. The orientation of the interface junction relative to the horizontal axis is described via the junction orientation angle, $\Theta$.

Results and discussion

Figures 1 and 2 display the source–drain photovoltage as a function of back-gate voltage $V_{\text{gate}}$ for incident light of various polarization and frequencies. Figures 1(b)–(d) displays the results for the homogeneous graphene Device #2, and figure 2 displays the results for devices with interface junctions. Gate dependent photovoltages that are linearly dependent on the intensity of incident radiation were observed for all samples. However, the samples with interface junctions also exhibit intrinsic polarization sensitivity. For the antenna-equipped trilayer homogeneous device (no interface junctions) (figures 1(b)–(d), the photovoltage in the THz region depends on the polarization orientation with maximum amplitude at $\alpha \sim 0^\circ$ and minimal response at $\alpha \sim 90^\circ$ (figure 1(c)). However, this polarization sensitivity is not intrinsic to the material. Such dependence of the photovoltage on the polarization is expected because of the antenna coupled to the structure [17]. When illuminated by radiation in the MIR or visible region, the electromagnetic field (EF) is not controlled by the antenna. In the homogeneous Device #2, the PR in these regions does not show the polarization dependence. (see figure 1(d)).
In contrast to the homogeneous Device #2, the THz polarization-dependence of devices with interface junctions is not determined by the antenna even though the area of the flake is much smaller than the area of the antenna. In fact, for all samples containing an interface junction, the photovoltage peaks at a polarization orientation that is (within experimental accuracy) perpendicular to the graphene interface junction boundary, regardless of the relative orientation of the antenna (as one can see in the figure 2, and in the results presented in supplementary data). Such sensitivity to the EF field polarization persists in both the MIR and visible regions (see figures 2(b), (c) and 3). For Device #3, the effect of polarization is particularly dramatic as it tunes not only the magnitude but also the polarity of the PR (figure 2(b)).

Figure 3 shows the PR polarization dependence in the MIR and visible regions for the homogeneous Device #2 (figures 3(a)–(c)), and for the interface junction devices: Device #5 (figures 3(d)–(f)), and Device #4 (figures 3(g)–(i)). Figures 3(a), (d) and (g) show the flake geometries, orientations of interface junctions, and orientation of polarization of applied EF (shown in figure 3(a)). Figures 3(b), (e), and (h) show the details of PR for corresponding devices in MIR region, and figures 3(c), (f) and (i) illustrate the PR behavior of investigated devices in visible region. As previously mentioned, in MIR and visible regions antenna does not play a role (this is important for the antenna-equipped Device #2). The Devices #4 and #5 have no antenna.

The first important observation in figure 3 is the strong polarization dependence of the MIR response in the interface junction devices (compare figure 3(e) for Device #5 and figure 3(b) for the homogeneous Device #2) with maximum PR for polarization orientations close to 90°. The same tendency remains in the visible region (compare figure 3(c) for Device #2 and the figure 3(f) for Device #5). The detailed analysis of angular α-dependence of PR demonstrates that the response is bound to the orientation of interface junctions and is maximal when polarization (within experimental accuracy) is perpendicular to the directions of interface junction boundaries: for example, both the MIR and the visible responses of the Device #4 (figures 3(h) and (i)) are maximal when

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**Figure 2.** (a) The gate dependence of the photoreponse to 12 mW of 2.5 THz radiation of unbiased Device #1 at 12 K, measured at different orientations of linear polarization of applied radiation. Here: α = 0°—blue curve; α = 22.5°—dash dot blue, α = 45°—green, α = 67.5°—orange, α = 90°—red curve. The black curve shows the transport curve of the sample measured with 10 nA bias, with indicated position of charge neutrality point (CNP). (b) The gate dependence of the photovoltage in the presence of 16 mW of λ = 9.27 μm light, of Device #3 (bilayer/ABA trilayer junction) at 12 K for different polarization orientations. The color code for polarization orientation is the same as in (a). The black curve shows the transport curve of the sample measured with 10 nA bias. (c) The linear polarization orientation dependence of visible range photoresponse of no-antenna trilayer/4-layer interface junction Device #4. 780 nm-radiation of fiber laser, power 18 mW, sample temperature 15 K, backgate voltage 0 V. (d) The optical image of the flake area of the Device #4, showing the orientations of interface junction (red dashed line) and light polarization. Here, the area above the red dashed line is an ABA trilayer, and the area below the line is a 4-layer graphene.
polarization orientation is not exactly \( \alpha = 90^\circ \), but shifted from the precise vertical position towards angles close to \( \alpha = 90^\circ + \Theta + 180^\circ l \) where \( l \) is an integer number. The same type of shift, recorded with much higher accuracy because of higher magnitude of PR, can be seen in figure 3(f) for the Device #5: the visible PR of this device is maximal for orientations close to \( \alpha = 90^\circ + \Theta + 180^\circ l \).

This type of polarization dependence is the sign of a photoelectric response [32]. The photoelectric response plays an important role in metal–graphene–metal systems [29, 32]. In these systems, the photoelectric response occurs at locally irradiated graphene–metal boundaries [29, 32]—another variant of inhomogeneous graphene. The polarization dependence is similar to what we have observed in interface junction systems where the response peaks when light polarization is perpendicular to the graphene–metal boundary [29, 32, 33]. The key difference between these earlier results for the graphene–metal systems, and our result for the systems with interface junctions, is the alignment of polarization dependence to the orientation of interface junction, and not to the graphene–metal boundaries. If metal–graphene...
boundaries would dominate our PR, the maximum signal would be observed at horizontal polarization orientations, $\alpha = 0^\circ + 180^\circ l$. We do not see this in the THz region (compare the figures 1(c) (for the homogeneous Device #2) and of the figure 2(a) (for the interface junction Device #1 with junction orientation $\Theta = -12^\circ$), or in Mid-IR or visible (see the figure 3 and the discussion above). Our experiments show the dependence which is close to $\alpha = 90^\circ + \Theta + 180^\circ l$, but not to $\alpha = 0^\circ + 180^\circ l$. Figure 4 clearly illustrates the difference we observe in our experiments: as one can see, the Device #5 with interface junction demonstrates a very pronounced polarization dependence aligned to the direction of interface junction boundary, while the very similar Device #2 with homogeneous graphene flake shows no pronounced polarization dependence. Our explanation of the observed difference is simple. When a graphene flake is attached to opposing metal electrodes, simultaneous irradiation of both electrodes should (and does) generate the response of opposite signs. Therefore, simultaneous responses from two opposite metal–graphene boundaries effectively cancel each other, and significantly reduce the overall response. In our experiments, the irradiation spot size was bigger than the size of flakes for all explored wavelengths, therefore in all our experiments we were simultaneously irradiating both graphene–metal boundaries.

It is important to point that the polarization-dependent photoelectric response is not the entire PR in our devices. As one can see in the figure 2(a) or in the figure 3, the polarization-dependent PR coexists with well-detectable polarization-independent components: for example, the polarization-dependent response ‘oscillations’ in figures 3(l), (h) or (i) are based on a ‘background’ PR signal. We interpret this polarization-independent component of signal as predominantly photothermoelectric response [18, 28, 32].

Another difference between the devices with and without interface junctions is a strongly enhanced response of devices with graphene interface junctions. The enhancement is as large as a factor of $\sim 70$ for THz radiation, $\sim 40$ for MIR and $\sim 3–4$ for the visible regions. This is evident when comparing the magnitudes of the PR in figures 1–4. Thus the presence of interface junctions significantly alters both the polarization sensitivity and the magnitude of the PR in few-layer graphene devices.

These remarkable features, namely the enhanced sensitivity and the polarization dependence of the PR (comparing to otherwise similar homogeneous samples), underscore a novel mechanism of photovoltaic response involving interface junctions. Numerical simulations were undertaken to provide insight regarding the underlying physics. The simulations show that junction states localized at the $n$-layer/(n–1)-layer graphene junctions enable a polarization dependent PR which becomes more pronounced as the wavelength increases.
In our analysis of the PR, we modeled the graphene based devices containing the interface junctions. To enable calculation of the bandstructure we consider a graphene based nanoribbon of a finite width on each side of a bilayer-trilayer interface junction, which is translationally invariant along the junction. Figure 4(a) shows the electronic structure for such ribbon, the electron wave vector is chosen along the junction direction. One can notice the appearance of flat bands corresponding to states localized either near the bilayer-trilayer junction or near the edges [34] of the finite width ribbon. Since the latter arise because of finite-size effects in our calculation, we concentrate on the states localized near the junction. Figure 5(b) shows the probability density associated to the state marked with a red dot in the bandstructure in (a). Although in this wave-vector region the band is not as flat as it is for larger wave-vectors, the states remain quasi-localized near the junction with a localization length of a few nanometers. For this particular state with energy $\varepsilon$ and wavefunction $\Psi(\vec{k}, \varepsilon)$, highlighted in figure 5(b), the spatial dependence of the associated probability $|\Psi(\vec{k}, \varepsilon)|^2$ shows that it is mainly localized at the boundary on the top layer with some degree of localization on the other layers. The localization length of such 1D states is around 10 nm (see figure 5(b)). The calculations were done numerically based on a semi-empirical atomistic model for the junction, with details described in the supplementary data.

We find that these transverse localized interface junction states show an enhanced optical absorption particularly in the THz-Mid IR range. This is shown in figure 5(c), where the oscillator strength, a magnitude which quantifies the optical absorption, is plotted. The oscillator strengths in figure 5(c) correspond to the transitions among the bands connected with arrows in the figure 5(a), which occur mostly in the THz to MIR range. One can see that the transitions involving the quasi-localized states along the junction have more than one order of magnitude enhanced oscillator strength as compared with those among regular bands, e.g. see figure 5(c) red line. We argue that the observed enhanced PR is mostly due to these interface junction states which display an enhanced optical absorption.

The observed polarization dependence of the PR for samples containing an interface junction can be explained as follows. The interface electron states, localized in the direction across junction, form one-dimensional channel along the junction boundary. These electron states have one-dimensional electron band structure with many bands (see figure 5(a)). For electrons occupying interface junction states, the description of interaction with EF can be greatly simplified by taking into account only the inter-band optical transitions between two bands. Using a two-band model, the dynamics of interface junction electrons interacting with the EF is described by the effective Hamiltonian [10, 11] (See also the supplementary data)

$$\hat{H} = \hat{\varepsilon}(\vec{k}) + \Delta_\varepsilon \hat{c}_\varepsilon \cos(\omega t) + U(y),$$

(1)

where the so-called dynamical gap $\Delta_\varepsilon \propto \sqrt{S}$ (S is the intensity of radiation) [10, 11] determines the interaction of electrons with the EF, $U(y)$ is the coordinate-
dependent potential, and \( y \) is the direction along the interface junction (see figure 5(d)), \( \hat{\sigma}_x, \hat{\sigma}_y \) are Pauli matrices, and \( \varepsilon(\vec{k}) \) is the electron energy in the conduction band. This Hamiltonian accounts for the radiation induced direct resonant transitions between valence and conduction bands.

Resonant absorption leads to occurrence of a photocurrent of nonequilibrium electrons \( I_{ph} \), generated via rectification due to the potential step of \( U(y) \) at the interface between suspended and non-suspended parts of graphene flakes. It was shown in [12–14] that, in the limit of a low radiation intensity, the photocurrent, \( I_{ph} \propto \Delta R \). As shown for monolayer [13, 14] and for bilayer graphene (see supplementary information) the dynamical gap \( \Delta_R \propto \sin(\varphi) \), where \( \varphi \) is the angle between the polarization direction of radiation and the wavevector of propagating electrons \( \vec{k} \). Thus, the dynamical gap, and, therefore, the photocurrent, reaches a maximum when the illumination is polarized perpendicular to the propagation direction of excited electrons. Our calculations (see figure 5(b)) show, that the electrons are localized at interface junction states. Therefore the electrons are only free to move along the interface and the photocurrent is parallel to the interface junction. In this case, the electron wavevector is parallel to the interface and hence parallel to \( \Theta \), so \( \varphi = \alpha - \Theta \). In our experiments, the asymmetric potential introduced at the boundary between suspended and non-suspended portions of the device provides a built in field in a field that causes the photocurrent and converts this current into a measurable photovoltage. Our experimental observation that the maximum of the photovoltage \( V_{ph} – I_{ph} \) occurs when the polarization is perpendicular to the interface junction (see figures 2(a) and (c)—experiment, and figure (3)) therefore indicates that the dominant contribution to the PR occurs from the interface junction states, i.e. electrons propagating along the interface junction (see figure 5(d)). The asymmetric potential realized at the boundary between suspended and non-suspended portions of the device, leads to the intrinsic electric field on the interface junction (see figure 5(d)), and therefore, the photovoltaic effect [12–14] is the main mechanism to generate the photocurrent. This photocurrent converts into a measurable photovoltage. Also, the thermoelectric effects involving transport of nonequilibrium carriers and different values of thermopower in a rectifying junction, similar to that described in the [21, 35], can give additional contributions to the photocurrent A more detailed discussion is provided in the supplementary data.

Our devices demonstrate the potential to be a graphene polarization analyzer operating broadly over the THz, IR and visible spectral regions. With the exception of [18], other efforts to develop long-wavelength graphene-based photodetectors were limited to devices operating at either higher (tens of THz) or lower (300 and 600 GHz) frequencies. So far, on-chip THz polarization analyzers have been limited to very narrow wavelength regions [36] and cryogenic temperatures. In contrast, the polarization selective PR of our graphene junction devices was observable up to 250 K, as shown in figure 6.

The experimentally observed dependence of the PR on an applied back-gate voltage \( V_{gate} \) (see for example figures 2(a), (b) and 3(e)) can be explained as following. The applied back-gate voltage allows one to vary the potential barrier in the rectifying junctions. (see figure 5(d)). The barrier height \( \Delta U \) for a bilayer graphene based rectifying junction (see figure 7) is determined by the gate voltage \( V_{gate} \) as

\[
\Delta U = \frac{2\pi \hbar^2 \varepsilon_m (\varepsilon - 1)}{e m D} V_{gate},
\]

where \( \varepsilon \) is the dielectric permittivity of SiO2 layer, \( e \) is the electron charge, \( m \) is the effective mass in bilayer graphene, and \( D \) is the thickness of SiO2 layer.

The photocurrent depends on the ratio of two energy scales, \( |\Delta U| \) and the photon energy \( hf \), where \( f \) is the frequency of EF. In the limit of \( |\Delta U| \ll hf \) the most important contribution to the photocurrent originates from narrow energy region of the width \( \delta E \approx |\Delta U| \) (of deeply lying in the valence band energy states). Taking into account that the probability of EF-induced direct resonant transitions ([12, 13]) is determined as \( P \approx \Delta^2 d/|\Delta U| \), where \( d \) is the width of rectifying junction (between suspended and non-suspended parts of graphene flakes), we obtain the bipolar dependence of \( I_{ph} \) on \( \Delta U \) as

\[
I_{ph} \approx d \Delta^2 (hf)^{1/2} \left[ \frac{\Delta U}{hf} \right]^{1/2}.
\]

The electron transport and the photocurrent creation in the limit \( |\Delta U| \ll hf \) is shown schematically in the figure 7(a).

In the opposite limit \( |\Delta U| \gg hf \), the narrow region of width \( \delta E \approx 2hf \) of the energy states lying near the Fermi energy \( \varepsilon_F \) gives the major contribution to the photocurrent. Moreover, in this case the
unipolar motion of photoelectrons occurs (see schematic in figure 7(b)). Thus, in this case $I_{ph} \approx d \Delta \varepsilon (|\Delta U|)^{1/2} \left( \frac{hf}{\Delta U} \right)$ and we obtain a decrease of the photocurrent with the back-gate voltage. The maximum value of the photocurrent occurs as $hf \approx \Delta U$. Thus for the EF field with the frequency $f = 2.5$ THz the photocurrent maximum should occur when $\Delta U \approx 10$ meV. Using equation (2), we obtain the corresponding back-gate voltage $V_{gate} = 2V$ that is in a good agreement with experiments (see for example figure 2(a)).

This photocurrent mechanism that uses the potential discontinuity between suspended and supported sections of a graphene flake depends crucially on EF-induced resonant transitions. Therefore, it can be destroyed by thermal fluctuations as $k_B T \approx hf$. For example, at $f = 4.25$ THz, the gate dependence of the photocurrent should wash out for temperatures exceeding $T \approx 230$ K. This is also in a good agreement with experiments (see the figure 6).

A possible application of our polarization analyzer may be beam profiometry for lasers and other sources of IR radiation. Figure 8 shows a beam profile of a THz laser, measured using Device #3 through raster scanning the device.

**Summary**

In conclusion, we have revealed a previously unobserved type of electronic state with a relatively strong optical response tied to the interface junction between graphene sheets with different number of monolayers on each side of the junction. We have characterized the optical properties of these interface states both experimentally and from first principles calculations to find that these interface states are localized and have an enhanced dipole matrix element for interband excitation. Photo-excited electrons generate photocurrent and photovoltage at the rectifying junction between suspended and non-suspended parts of graphene flakes. Because of the one-dimensional geometry, the interface junction states support a polarization dependent photocurrent which is aligned along the junction in inhomogeneous graphene samples. The photocurrent shows the maximum value as the EF polarized in the direction perpendicular to the interface junction. We have found that in graphene flakes with interface junctions connecting source and drain electrodes, the interface junctions have a similar PR contribution to that of a nanowire placed between source and drain electrodes.

Alltogether, our results demonstrate the capability of graphene interface junctions to form 1D electronic states, whose properties can be exploited for electronic and optoelectronic applications. We discovered these
states in graphene but we anticipate that similar type of states will be important and will definitely influence physical properties for all other 2D few-monolayer semiconducting materials.

Experimental methods and techniques

Materials preparation, device fabrication, and properties of experimental samples

Five suspended graphene devices on Si/SiO$_2$ substrates used as a backgates, have been investigated. Suspended interface-layered graphene films were transferred onto highly phosphorus doped Si wafers capping 300 nm thick SiO$_2$ with pre-defined trenches via the mechanical cleavage technique \[37\]. The 150 nm deep trenches in SiO$_2$ were etched using photo-lithography and inductively coupled plasma process$^\star$. The successfully placed flakes with interface junctions were determined via optical and Raman microscopy and atomic force microscopy AFM. After localization of flakes, device contacts and antenna systems were deposited by Ti/Au (10 nm/50 nm) physical vapor deposition through a lithography-free fabrication technique (see \[30\]).

Experimental techniques

Raman microscopy measurements were performed using a Jobyn Yvon LabRam Aramis spectrometer equipped with a CCD camera. The second harmonic of a Nd:YAG laser (532 nm wavelength, 5 mW CW radiation) was used for excitation.

The PR of the devices was measured using several laser sources in different wavelength regions. In the THz range, we used the 2.5 and 4.25 THz lines of CO$_2$-pumped far-IR laser (Edinburgh Instruments’s FIRL), in the MIR region we used different lines of CO$_2$ laser (10.6 and 9.3 $\mu$m wavelengths). For the visible (VIS) region, we used the 780 nm line of a fiber laser (Eagle Yard Photonics Model DFB 780). Depending on the laser wavelength, the linearly polarized laser radiation was focused onto the samples via a 2$\pi$ diameter $f$/1 90° off-axis parabolic mirror (for THz), or via 1$\pi$ diameter $f$/2 ZnSe lens (IR and visible). In all frequency regions, the laser spot size was larger than the size of the graphene flakes such that the entire graphene flake under study was irradiated.

The samples were located in a polyethylene and Quartz (THz) or ZnSe (IR, visible) window-equipped variable temperature continuous flow cryostat (Cryo Industries Model CRC-102) in the temperature range of 8–300 K. The samples were in the vacuum, mounted at the cryostat’s sample holder that was equipped with a temperature sensor. The photoinduced changes of source-drain voltage of gated, unbiased or source–drain current-biased, samples were detected using a lock in amplifier (Stanford Research SR830) or oscilloscope (Tektronix TDS 3034B) with a voltage pre-amplifier (Stanford Research SR 560), and were recorded using LabView-driven remote control system. We used mechanical chopper modulation for THz and visible radiation. The same chopper was used in some MIR measurements. In most MIR experiments, the MIR laser was synchronized using the TTL output of a lock-in amplifier, which allowed us to have precise control over phasing and polarity of the PR.

The linear polarization of laser radiation was achieved using linear polarizers. The rotation of the laser polarization plane for THz and MIR radiation was achieved via use of two mylar (THz) or ZnSe (IR) wire grid polarizers. In the visible region, for rotation of polarization angle we used a half-wave plate.

Before every PR measurement, each sample was annealed by passing a few mA current across the graphene flakes; this was done in vacuum and at sample temperatures below 70 K. The devices were cooled down to low temperature and we performed the current annealing procedure of \[38\] to improve their quality via Joule heating effect. The annealing rate was at 0.1–0.2 mA/$\mu$m$^2$/layer. The charge neutrality point was determined at the minimum point of conductance-gate behavior, the evaluated Drude electron mobilities in our samples were of the order of $\mu \approx 15 000$ cm$^2$ V$^{-1}$ s$^{-1}$ or higher.

Using a lock-in amplifier, we measured the dark voltage noise level of the Device #3 at 10 K, with zero bias. We have found the noise at the level of $\sim 65$ nV Hz$^{-1/2}$.

Pointing control

In the PR experiments, it was very important to make sure that the observed results were related to changes of polarization orientation, and were not artifacts related to changes in the spatial intensity distribution. This problem is less important in THz measurements since the approximately 100 $\mu$m beam waist diameter of THz laser is large compared to the few micron size of the graphene flakes such that the field distribution is practically homogeneous across the flake. In the MIR and visible range, the beam diameter may be comparable with flake size. For these wavelengths the flakes were positioned in the center of the beam using the following beam alignment procedure for each polarization orientation: the samples position was scanned over a 2D area by moving the entire cryostat with motorized XYZ-stages with piezo-motor step size 0.3 $\mu$m. The laser beam profile was measured using the samples as detectors to measure the PR as a function of sample position. Once the location of the center of the beam was known, the sample was moved to this location figure 9 shows typical beam profiles in the MIR (left) and VIS (right) regions. Using this technique, we had accurate pointing control mitigating

\[ \star \] The SiO$_2$ trenches were fabricated at Nanotech-The UCSB Nanofabrication Facility.
spatial intensity distribution variations that could effect the polarization dependence results.

Author contributions

NGK and CNL designed research; LJ and CNL fabricated and tested samples; NGK, LW, MO, GCD, AG, MCW, and EAS carried out electron transport and PR measurements, ESM and LEFFT carried out the modeling, ESM performed the numerical calculations, NGK, ESM, CNL, MCW, EAS, LEFFT, MVF and KBE analyzed and interpreted data; and CNL, GCD, MCW, EAS, LEFFT, MVF, KBE, and NGK wrote the manuscript.

Funding sources

We acknowledge support from NSF (projects ECCS #0925988 and ECCS #0926056), and from the SPP 1459 ‘Graphene’ and SFB Transregio 12 by DFG, and from the Ministry of Education and Science of the Russian Federation in the framework of Increase Competitiveness Program of NUST ‘MISiS’ (K2-2014-015). The work at Sandia National Laboratories was supported by the DOE Office of Basic Energy Sciences. This work was performed, in part, at the Center for Integrated Nanotechnologies, a US Department of Energy, Office of Basic Energy Sciences user facility. Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the US Department of Energy’s National Nuclear Security Administration under contract DE-AC04-94AL85000.

Acknowledgments

We thank Stephan Roche and Francois Leonard for fruitful and inspiring discussions, and thank Emil Kadlec and Don Bethke for their help in experiments. ESM acknowledges support from FONDECYT 11130129 grant and LEFFT thanks Program ‘Inserción Académica’ 2016 of the University of Chile.

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