Dual-gated graphene devices for near-field nano-imaging

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Graphene-based heterostructures display a variety of phenomena that are strongly tunable by electrostatic local gates. Monolayer graphene (MLG) exhibits tunable surface plasmon polaritons, as revealed by scanning nano-infrared experiments. In bilayer graphene (BLG), an electronic gap is induced by a perpendicular displacement field. Gapped BLG is predicted to display unusual effects such as plasmon amplification and domain wall plasmons with significantly larger lifetime than MLG. Furthermore, a variety of correlated electronic phases highly sensitive to displacement fields have been observed in twisted graphene structures. However, applying perpendicular displacement fields in nano-infrared experiments has only recently become possible (1). In this work, we fully characterize two approaches to realizing nano-optics compatible top-gates: bilayer MoS\textsubscript{2} and MLG. We perform nano-infrared imaging on both types of structures and evaluate their strengths and weaknesses. Our work paves the way for comprehensive near-field experiments of correlated phenomena and plasmonic effects in graphene-based heterostructures.

Graphene-based van der Waals (vdW) heterostructures display a variety of phenomena including superior plasmonic properties (2–5), tunable band structures (6–8), topological edge states (9–11), and correlated phases such as superconductivity (12, 13). This large variety of electronic phases arises because the properties of graphene are strongly tunable by electrostatic gates. The optical excitations corresponding to these phases lie in the infrared range of the electromagnetic spectrum (14), where the wavelength of light, $\lambda_0$, ranges from 1µm to 100µm. Probing such heterostructures with conventional far-field optical experiments is challenging because of their limited lateral dimensions compared to $\lambda_0$. However, tip-based scanning nano-infrared experiments can overcome the diffraction limit and achieve a spatial resolution better than 10nm (15).
Nano-infrared experiments have established monolayer graphene (MLG) as an excellent platform for plasmonics because of a large confinement ratio $\lambda_0/\lambda_p$ ($\lambda_p$ is the plasmon wavelength) (17), tunability with an external gate (3, 18) and long lifetimes for the SPPs approaching 2 ps (4, 19). While MLG is well studied, the plasmonic properties of bilayer graphene are relatively unexplored (20). When bilayer graphene is gapped and the Fermi level lies in the gap, exotic plasmonic phenomena are predicted to occur. Gapped BLG under photoexcitation is predicted to amplify SPPs (21) while domain wall solitons in gapped BLG could host one-dimensional SPPs with lifetimes approaching $10^2$ ps (22).

Nano-infrared experiments have also begun to probe multilayer graphene-based Moiré systems that are known to host correlated electronic phenomena such as twisted bilayer graphene (TBG) (12), twisted trilayer graphene (TTG) (23) and twisted double bilayer graphene (TDBG) (24, 25). The electronic properties of all these systems are strongly sensitive to perpendicular displacement field. For example, the correlated insulator phases in TTG and TDBG appear only for a limited range of displacement fields (23, 24).

In transport experiments, perpendicular displacement field can be introduced using an evaporated metal layer or a graphite layer as a top-gate in conjunction with a back-gate. While transport (26) and some far-field optical experiments (27–29) can be performed on such structures, they are incompatible with nano-infrared experiments for multiple reasons. First, such layers are relatively thick (tens of nm) which make the underlying graphene layer inaccessible to nano-optics experiments. When the layers are made thinner, the presence of a high density of high mobility free carriers leads to plasmonic effects in the top-gate which modifies and obscures the behavior of the underlying graphene layer.

Recent work has shown that MLG could be used as a top gate to study Moiré patterns in vDW heterostructures (1). However, the capabilities and limitations of the top gate were not fully explored. While Ref (1) showed that two-dimensional domains in a Moiré pattern could be visualized through a MLG top gate, it’s not yet known if the plasmonic phenomena in the underlying graphene layer and one-dimensional features such as domain walls in BLG can be resolved. In this work, we demonstrate and fully characterize two approaches for nano-optics compatible top gates: bilayer MoS$_2$ and MLG. We are able to visualize the plasmons in the MLG and TBG layers underneath the MoS$_2$ top-gate. We further demonstrate a depletion of the carrier density of the underlying graphene layers with the top-gate through measurements of the plasmon wavelength and nano-infrared scattering amplitude. We then explore the use of a MLG layer as the top-gate for BLG. The doped MLG layer is a robust top-gate, but has strong optical response at mid-infrared frequencies of its own and therefore obscures the underlying BLG. However, we overcome this limitation by selectively probing the underlying BLG through nano-photocurrent imaging and are able to visualize domain walls in the BLG layer. Our work paves the way for realization of fully tunable vDW devices compatible with nano-optics experiments.
Figure 1 | Nano-optics measurements on MoS$_2$-gated monolayer graphene. (A) Schematic of our nano-infrared experiment and our device. Bilayer MoS$_2$ is contacted by Ti/Au from above while the graphene has a side contact made of Cr/Au. (B) Two-dimensional image of the nano-infrared amplitude $s$ over our device for $V_{bg} = +80$V and $V_{tg} = 0$V. The edges of MLG and MoS$_2$ are represented by magenta dashed and green dotted line represents respectively. (C) Line profiles of the nano-infrared amplitude $s$ across a graphene edge showing plasmon polaritons. Black dashed lines represent fits to a damped oscillations model (Section S3 of (16)). (D) Calculated imaginary part of the reflection coefficient $\text{Im}(r_P)$ matching the two experimental heterostructures: (D) BN/MLG/BN/SiO$_2$ (E) MoS$_2$/BN/MLG/BN/SiO$_2$. The bright contour of maximal values corresponds to the plasmon mode. The circles in (D) and (E) correspond to experimental data extracted from panel (C) (16).

Figure 1(A) shows a schematic of our experimental setup. Our first device consists of monolayer graphene encapsulated between hexagonal boron nitride (hBN) layers. The thickness of the top hBN layer is kept small (2 nm) to allow optical near-field access to the underlying graphene layer. A bilayer of MoS$_2$ is then placed on the top hBN layer for use as a top-gate while a doped silicon layer underneath the heterostructure serves as the bottom-gate. We chose MoS$_2$ because it is expected to be transparent to mid-infrared light. We study this device using a scanning nano-infrared microscope. Incident light of frequency $\omega = 1/\lambda_0 = 905$ cm$^{-1}$ is focused onto the apex of a sharp metallic tip. The amplitude $s$ and phase $\phi$ of the scattered light are detected with an interferometric method (30). The sharpness of the tip provides the momentum necessary to launch SPPs which propagate radially outward from the tip. When the SPPs encounter a physical (3, 18) or electronic (31) boundary, they are reflected and form a standing wave pattern that we directly visualize.
Figure 1(B) shows a two-dimensional map of the nano-infrared amplitude measured on our device with $V_{bg} = +80V$ and $V_{tg} = 0V$. The green dotted line marks the edge of the MoS$_2$ layer such that the area above the line does not contain MoS$_2$. We observe clear fringes parallel to the edges of the MLG (marked by magenta dashed lines) throughout the image. These fringes confirm that we are able to launch and image SPPs in the MLG layer even when the MLG is underneath MoS$_2$.

A comparison of the fringes above and below the MoS$_2$ boundary in Fig 1(B) indicates that the plasmon wavelength is smaller in the region with MoS$_2$. In Fig 1(C), we plot the line profiles extracted across the graphene edge from both regions. The line profiles confirm that the plasmon wavelength is reduced to 138 nm under the MoS$_2$ layer (blue line in Fig 1(C)) compared to 177 nm without MoS$_2$ (orange line in Fig 1(C)). This reduction is due to the large static dielectric constant of MoS$_2$ and the resulting screening. This change in plasmon wavelength is consistent with the calculated change in plasmon dispersion (Fig 1(D) and 1(E)).

**Figure 2 | Carrier density modulation in monolayer graphene with MoS$_2$ top-gate.** (A) Line profiles of the nano-infrared phase $\phi$ for various values of $V_{tg}$ at $V_{bg} = +80V$, showing a clear change in the plasmon wavelength. The data is scaled and shifted for clarity. Dashed lines follow the plasmonic peaks and are used to extract $\lambda_p$. (B) Plasmon wavelength as a function of the top-gate voltage $V_{tg}$. Circles represent $V_{tg} \leq 0V$ and asterisks represent $V_{tg} > 0V$. Inset shows the behavior of the MoS$_2$ top-gate for various $V_{tg}$ and $V_{bg}$ values. The top-gate is active in one of the four quadrants. (C) $\lambda_p$ as a function of the estimated carrier density in the graphene layer $n$. The data points for $V_{tg} > 0V$ cluster together because the top-gate is ineffective (described in text).

Figure 2 demonstrates the tuning of carrier density in the MLG layer with the MoS$_2$ top-gate. Figure 2(A) shows line-profiles of the nano-infrared phase $\phi$ for different values of top-gate bias $V_{tg}$ for a fixed value of back-gate bias $V_{bg} = +80V$. We observe a clear change in the plasmon wavelength as $V_{tg}$ is changed. At negative values of $V_{tg}$, we observe a decrease in the plasmon wavelength which is consistent with a depletion of the carrier density in the graphene
layer. When $V_{tg}$ is tuned to $+0.3V$, we observe an increase in $\lambda_p$. But a further increase in $V_{tg}$ to $+0.5V$ does not change $\lambda_p$, indicating that the carrier density in MLG does not change (Figure 2(B) and 2(C)). This limitation is the result of a pn-junction forming in the MoS$_2$ layer as described below. Taken together, our results confirm that we are able to deplete the carrier density in the graphene layer which is necessary for realizing gapped BLG.

We now turn to the BLG region of our heterostructure that is also covered by the MoS$_2$ top-gate. The BLG in our heterostructure was produced by a ‘tear-and-stack’ technique (Section S1 of [16]) which resulted in a small twist angle between the layers and a large Moiré pattern. Atomic relaxation leads to the formation of larger domains of Bernal bilayer graphene separated by domain walls (32) that host topological states (9–11). The change in optical conductivity arising from the topological states reflects plasmon polaritons leading to fringes in nano-infrared experiments (33, 34). Changing the carrier density and interlayer bias across the BLG changes the optical conductivity across the domain wall and modifies the fringe pattern (33). Figure 3(B) shows the nano-infrared amplitude over a region containing several domain walls for $V_{bg} = +80V$ and $V_{tg} = 0V$. We observe features in the amplitude that correspond to plasmons reflecting off the domain walls (34, 35). As we increase $V_{tg}$, we observe a clear change in the plasmonic pattern that confirms the changing carrier density and interlayer bias in the BLG layer. By demonstrating dual-gating and observing propagation of plasmons, we have thus shown the feasibility of performing nano-infrared studies of a dual gated system using this approach.

Figure 3 | Demonstration of top-gating effect in bilayer graphene domain walls. (A) Schematic of the heterostructure. (B, C, D) Nano-infrared amplitude image of domain walls in BLG for three different top-gate voltages with $V_{BG} = +80V$. Scale bar 400nm.

Next, we discuss the limitations of the TMD top-gate. First, we consider the performance of the top-gate at a negative $V_{bg}$. Because of the high work function of MoS$_2$, evaporated metals typically make n-type contact to MoS$_2$ (36). The geometry of our device is such that the titanium metal contacts to the MoS$_2$ lie outside the graphene region (Fig 1A). Therefore the contact resistance at the Ti/MoS$_2$ layer depends only on $V_{bg}$. At a negative $V_{bg}$, the Schottky barrier at the Ti/MoS$_2$ junction is too large and renders the top-gate ineffective. Therefore, unless doped
by local gates (37), the MoS$_2$ top-gate is only functional for positive $V_{bg}$ where n-type carriers are injected into the MoS$_2$ layer.

At a fixed, positive $V_{bg}$, the region by the contacts remains n-doped and the application of $V_{tg}$ starts to change the carrier density in the MoS$_2$ region directly above the graphene layer. When $V_{tg}$ is negative, the carriers in the MoS$_2$ layer are all n-type. However, as $V_{tg}$ becomes positive, the MoS$_2$ region above the graphene becomes hole-like. Since the carriers close to the contacts remain electron-like, a pn-junction forms in the MoS$_2$ layer along the graphene edge. This pn-junction isolates the Ti contacts from the MoS$_2$ region above the graphene layer and causes the top-gate to become ineffective. Taking the effects of the Schottky barrier and the pn-junction together, we conclude that the top-gate is effective only in one of the four quadrants in the $V_{tg} - V_{bg}$ plane, as shown in the inset of Fig 2(B).

**Figure 4 | Monolayer graphene as a top-gate for domain walls in bilayer graphene.** (A) Schematic of the heterostructure (B, C, D) Nano-photocurrent images for three different $V_{tg}$ showing domain walls in the underlying bilayer graphene. (G) Nano-infrared amplitude image at $V_{tg} = 0.7V$. Black dashed lines in (B-D) and yellow dashed line in (E) correspond to the boundary of the top-gated region. Grey solid lines in (C) indicate the domain walls in BLG. Scale bar 1µm.

To achieve full control over the properties of BLG, we could consider other materials as a top-gate. While a p-type TMD such as WS$_2$ can lead to a top-gate that is functional at negative $V_{bg}$, the pn-junction limitation would still restrict its functionality to just one quadrant in the $V_{bg} - V_{tg}$ plane. This limitation arises directly because of the electronic bandgap in a semiconductor and can only be avoided by using a gapless ambipolar material, such as monolayer graphene.

We now explore the possibility of using MLG as a top-gate for BLG. Figure 4 (A) shows a schematic of our heterostructure. Figure 4(E) shows the nano-infrared amplitude image of our device with $V_{bg} = 0V$ and $V_{tg} = +0.7V$. The yellow dashed line indicates the boundary of the
MLG top gate. We observe a nano-infrared contrast that indicates that the top gate is active but we see no other features. Figure 4 (A-D) shows the results of nano-photocurrent experiments (38) at different $V_{tg}$. As $V_{tg}$ is increased from zero, the nano-photocurrent begins to resemble the photocurrent profiles seen in other twisted BLG heterostructures (39, 40) and are known to arise from domain walls. These results suggest that even though plasmonic effects in the MLG top gate obscure the underlying graphene layer in nano-infrared experiments, nano-photocurrent experiments can be performed successfully.

Figure 5 | Direct comparison between the MLG and TMD top gates. (A) Change in the dispersion of the plasmonic mode of the heterostructure as the carrier density in the top gate is varied for a MLG top gate. (B) Similar plot as (A) for bilayer MoS$_2$ top gate. The thicknesses of the top and bottom hBN are 5nm and 30nm respectively and the underlying MLG layer is doped to $n = 6 \cdot 10^{12}$ cm$^{-2}$.

Finally, we directly compare the properties of MLG and bilayer MoS$_2$ top gates with the following hypothetical scenario. We consider an encapsulated heterostructure of monolayer graphene with a carrier density of $n = 6 \cdot 10^{12}$ cm$^{-2}$ with both a MLG top gate and a TMD top gate. We then vary the carrier concentration only in the top gate to observe how strongly the top gate modifies the behavior of the underlying graphene layer. The plasmonic dispersions in both cases are shown in Figure 5. The dispersion changes significantly with a MLG top gate while it
remains mostly unchanged in case of the TMD top gate. The large change in the dispersion with the MLG top gate is due to the carriers in the top gate affecting the plasmonic properties of the underlying graphene layer through dielectric screening. The smaller changes with a TMD demonstrate that the dielectric properties of the TMD top gate are significantly less sensitive to changes in the carrier density. These results suggest that a TMD top gate allows direct access to the plasmonic phenomena in the underlying graphene layer with minimal obscuring.

In conclusion, our results demonstrate two near-field compatible top gates for BLG. With MoS$_2$, we were able to study plasmons through scattering nano-infrared experiments and demonstrate the depletion of carriers in the underlying graphene layers. With a monolayer graphene top gate, we were able to visualize the domain walls in the underlying BLG through nano-photocurrent experiments. Our work paves the way for exploring the plasmonic properties of gapped bilayer graphene with scanning nano-infrared experiments.

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Supporting Information for “Dual-gated graphene devices for near-field nano-imaging”

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S1: Methods

Device fabrication

The results shown in Figs 1-3 were obtained from a MoS2 top-gated device (Device 1) consisting of graphene layers encapsulated in hexagonal boron nitride with a MoS2 layer on top. The stack was fabricated using the dry transfer method. A poly(bisphenol A carbonate) (PC) coated on a stamp made of transparent elastomer polydimethylsiloxane (PDMS). The two graphene layers to form the bilayer graphene were assembled by tearing a large single layer of graphene and stacking them together. The inherent strain in this process results in one of the layers twisting slightly relative to the other layer and leads to the formation of domain walls (S1). The graphene layers were contacts to the TMD layer were made of titanium while side contacts (S2) to the graphene layer were made of chromium and gold.

The MLG top-gated device (Device 2) was also fabricated with the dry transfer method but with a poly(propylene carbonate) (PPC) coated stamp. The heterostructure was fabricated in the reverse order so that the MLG top-gate was not contaminated by contact with the PPC polymer and flipped onto a SiO2/Si chip. After flipping, the heterostructure was annealed in vacuum to remove the PPC residue. Electrical contacts to the graphene layers were made with chromium and gold.

The results shown in Fig S2 were obtained from another MoS2 top-gated device (Device 3) that was also fabricated with the dry transfer method.
Infrared nano-imaging

Infrared nano-imaging was performed with a commercial scattering-type scanning near-field optical microscope (s-SNOM) based on a tapping mode atomic force microscope from Neaspec GmbH. Our light source was a quantum cascade laser obtained from DRS Daylight Solutions, tunable from 900 cm\(^{-1}\) to 1200 cm\(^{-1}\). The light from the laser was focused onto a metallic tip oscillating at a tapping frequency of around 250 kHz with a tapping amplitude of around 60 nm. The scattered light was detected using a liquid nitrogen cooled HgCdTe (MCT) detector. To suppress far-field background signals, the detected signal was demodulated at a harmonic \(n\) of the tapping frequency. In this work, we used \(n = 3\).

Plasmon wavelength and dispersion calculations

The dielectric constants for MoS\(_2\) and hBN used in the reflection coefficient calculations of Fig 1(D) and 1(E) were obtained from Refs (S3) and (S4) respectively. The plasmon wavelength used to plot the crosses was determined by the spacing between the fringes in the spatial profiles of the near-field phase (Fig 2(A)).

For the calculations in Figure 5, we assumed that the dielectric properties of a doped MoS\(_2\) layer can be described by a Drude model,

\[
\epsilon = \epsilon_\infty + \frac{\omega_p^2}{\omega^2 - i\gamma\omega}
\]

\[
\omega_p^2 = \frac{ne^2}{m^*\epsilon_0}
\]

where \(\epsilon_\infty\) and \(\epsilon_0\) are the high-frequency and low-frequency dielectric constants, \(\omega_p\) is the plasma frequency, \(\gamma\) is the damping, \(n\) is the carrier density and \(m^*\) is the effective mass of the carriers and \(e\) is the electron charge. We obtained \(\epsilon_\infty\) and \(\epsilon_0\) from Ref (S3). The band structure of TMDs is anisotropic but the out-of-plane effective mass is not known accurately. Here, we assumed that the effective mass \(m^*\) was isotropic as a worst case scenario and equal to \(m^* = 0.45\ m_0\), where \(m_0\) is the free electron mass (S5). The plasmon dispersion is insensitive to \(\gamma\).
S2: Nano-infrared images at different top-gate voltages

Figure S1 | Nano-infrared images of monolayer graphene (MLG) at $V_{bg} = +80\,\text{V}$ and various values of $V_{tg}$. The images on the left are the nano-infrared amplitude $s$ while the images on the right are of the nano-infrared phase $\phi$. The magenta lines represent the physical boundary of the MLG. The color bar limits for the amplitude images are identical for all gate voltages. The variation in the nano-infrared amplitude is consistent with a decrease (increase) of the carrier concentration as $V_{tg}$ is increased (decreased) ($S6$).

S3: Line profile fits using the damped oscillations model

The nano-infrared amplitude line profiles were fit using the damped oscillations model ($S7$):

$$ s = \frac{\cos(2qx) \exp(-2q\gamma x)}{\sqrt{x}} + \alpha \frac{\cos(qx + \phi) \exp(-q\gamma x + \phi)}{x} $$

Here, the first term represents plasmons that are launched by the tip and reflected by the graphene edge and the second term represents the plasmons that launched by the edge. The edge is assumed to be located at located at $x = 0$. $q$ is the plasmon momentum defined to be $q = \frac{2\pi}{\lambda_p}$, $\gamma$ is the damping of the plasmonic wave, and $\alpha$ and $\phi$ capture the difference in the magnitude and phase between the tip-launched waves and the edge-launched waves. The obtained fits are shown in Figure S2(A) and Figure 1(C) of the main text. The parameters derived from the fits are shown in Figure S2(B).
Figure S2 | Damped oscillations model fits for different top-gate voltages. (A) Nano-infrared amplitude line profiles for different top-gate voltages. (B) The plasmon wavelength $\lambda_p$ and the damping $\gamma$ extracted from the fits. The red asterisks correspond to the data without MoS$_2$ (red curve in Figure 1(C) of main text).

S4: Gating of bilayer graphene with MoS$_2$

Figure S3 | Gating bernal bilayer graphene with a MoS$_2$ top-gate (Device 3). (A) – (E) Nano-infrared amplitude images for $V_{bg} = 0$V various values of $V_{tg}$. The magenta dashed lines represent the boundaries of the BLG layer while the blue dotted lines represent the boundary of a gold electrode. Scale bar 2µm. (F) Dependence of the nano-infrared amplitude of the bilayer graphene layer on $V_{tg}$. The decrease in the nano-infrared amplitude as $V_{tg}$ is increased is consistent with an increase in the carrier concentration in the BLG layer (S6).
In this section, we present nano-infrared data from another bilayer graphene device (Device 2) with a MoS$_2$ top-gate. In this device, there were no domain walls. We observed a change in the nano-infrared signal when varying the top-gate voltage $V_{tg}$.

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