Gate-tuning of graphene plasmons revealed by infrared nano-imaging

Z. Fei1, A. S. Rodin1, G. O. Andreev1, W. Bao2,3, A. S. McLeod1, M. Wagner1, L. M. Zhang4, Z. Zhao2, M. Thiemens5, G. Domínguez6, M. M. Fogler1, A. H. Castro Neto7, C. N. Lau2, F. Keilmann8 & D. N. Basov1

Surface plasmons are collective oscillations of electrons in metals or semiconductors that enable confinement and detection of electromagnetic energy at subwavelength scales1–4. Rapid progress in plasmonics has largely relied on advances in device nano-fabrication5–7, whereas less attention has been paid to the tunable properties of plasmonic media. One such medium—graphene—is amenable to convenient tuning of its electronic and optical properties by varying the applied voltage8–11. Here, using infrared nano-imaging, we show that common graphene/SiO2/Si back-gated structures support propagating surface plasmons. The wavelength of graphene plasmons is of the order of 200 nanometres at technologically relevant infrared frequencies, and they can propagate several times this distance. We have succeeded in altering both the amplitude and the wavelength of these plasmons by varying the gate voltage. Using plasmon interferometry, we investigated losses in graphene by exploring real-space profiles of plasmon standing waves formed between the tip of our nano-probe and the edges of the samples. Plasmon dissipation quantified through this analysis is linked to the exotic electrodynamics of graphene7. Standard plasmonic figures of merit of our tunable graphene devices surpass those of common metal-based structures.

In general, surface plasmons can exist in any material with mobile charge carriers whose response to electric field remains reactive—that is, whose in-plane momentum (q)-dependent and frequency (ω)-dependent complex conductivity, σ(q, ω) = σ1 + iσ2, is predominantly imaginary. Of particular interest are plasmons with high momenta, qp ≫ ω/c (where c is the velocity of light), which may be used for extreme concentration of electromagnetic energy1–3. In conventional bulk metals, the frequencies of such high-q plasmons reside in the visible or ultraviolet ranges. In graphene, they are expected to appear in the terahertz and infrared domains12. However, these high-q infrared plasmons are dormant in conventional spectroscopy of graphene. Here we used the scattering-type scanning near-field optical microscope (scattering-type SNOM) to experimentally access high-q plasmons by illuminating the sharp tip of an atomic force microscope (AFM) with a focused infrared beam (Fig. 1a). The momenta imparted by the tip extend up to a few times 1/a, where a ≈ 25 nm is the curvature radius of the tip13, thus spanning the typical range of infrared plasmon momenta (qp) in graphene11. The spatial resolution of scattering-type SNOM is also set by a, and proves to be an order of magnitude smaller than the plasmon wavelength λp. The direct observable of our method—the scattering amplitude s(ω)—is a measure of the electric field strength inside the nanoscale gap between tip and sample. Consequently, the scattering-type SNOM technique enables spectroscopy13 and infrared nano-imaging of graphene plasmons without the need to fabricate specialized periodic structures14. Our imaging data elucidate real-space characteristics of infrared plasmons in graphene such as reflection, interference and damping. All these phenomena can be readily manipulated with gate voltage—a noteworthy property unattainable in metal-based plasmonics.

To probe directly the properties of graphene plasmons, we utilize a frequency ω = 892 cm−1 corresponding to a wavelength λIR = 11.2 μm in the infrared (IR) regime where the plasmon is unimpeded by the surface optical phonon supported in graphene/SiO2/Si structures15. The nano-imaging results are shown in Fig. 1b–e, where we plot normalized near-field amplitude s(ω) = s2(ω)/s2(ω). Here s2(ω) and s2(ω) are the interference pattern close to graphene edges (blue dashed lines) and defects (green dashed lines and green dot), and at the boundary between single (G) and bilayer (BG) graphene (white dashed line). Additional features marked with arrows in e are analysed in refs 27 and 30. Locations of boundaries and defects were determined from AFM topography taken simultaneously with the near-field data. Scale bars, 100 nm. All data were acquired at ambient conditions.

1Department of Physics, University of California, San Diego, La Jolla, California 92093, USA. 2Department of Physics and Astronomy, University of California, Riverside, California 92521, USA. 3Materials Research Science and Engineering Center, University of Maryland, College Park, Maryland 20742, USA. 4Department of Physics, Boston University, Boston, Massachusetts 02215, USA. 5Department of Chemistry and Biochemistry, University of California, San Diego, La Jolla, California 92093, USA. 6Department of Physics, California State University, San Marcos, California 92096, USA. 7Graphene Research Centre and Department of Physics, National University of Singapore, 117542, Singapore. 8Max Planck Institute of Quantum Optics and Center for Nanoscience, 85714 Garching, Germany.

Figure 1 | Infrared nano-imaging experiment and results. a, Diagram of an infrared nano-imaging experiment at the surface of graphene (G) on SiO2. Green and blue arrows display the directions of incident and back-scattered light, respectively. Concentric red circles illustrate plasmon waves launched by the illuminated tip. b–e, Images of infrared amplitude s (ω = 892 cm−1) defined in the text taken at zero gate voltage. These images show a characteristic interference pattern close to graphene edges (blue dashed lines) and defects (green dashed lines and green dot), and at the boundary between single (G) and bilayer (BG) graphene (white dashed line). Additional features marked with arrows in e are analysed in refs 27 and 30. Locations of boundaries and defects were determined from AFM topography taken simultaneously with the near-field data. Scale bars, 100 nm. All data were acquired at ambient conditions.

©2012 Macmillan Publishers Limited. All rights reserved
third-order demodulated harmonics of the near-field amplitude measured for the given sample and for a Si reference sample, respectively. The near-field amplitude \( s(\omega) \) tracks real-space variations in the local electric field underneath the tip, enabling exploration of surface phonon polaritons and surface plasmons.

In Fig. 1b, we present a \( s(\omega) \) image acquired at the graphene–SiO\(_2\) interface revealing periodic oscillations of the \( s(\omega) \) signal extending along the graphene edge. Point and circular defects (Fig. 1d) trigger circular fringe patterns. Line defects (Fig. 1b) produce elongated, elliptical patterns. Furthermore, we observed fringes at both sides of the boundary between single-layer and bilayer graphene (Fig. 1c). Finally, strongly tapered corners of graphene (Fig. 1c) reveal the two-dimensional metal residing at the interface of vacuum (dielectric constant \( \varepsilon_0 = 1 \)) and a substrate with dielectric function \( \varepsilon_{\text{sub}} \).

Images in Fig. 1 are consistent with the following scenario. Illuminated by focused infrared light, the AFM tip launches plasmon waves of wavelength \( \lambda_p \) propagating radially outward from the tip. Sample edges or defects act as (imperfect) reflectors of the plasmon waves, directing them back to the tip. Therefore complex patterns of interference between launched and reflected plasmons should form inside graphene. We emphasize that our experimental technique does not capture instantaneous snapshots of these complex patterns. Instead, whereas the tip ‘launches’ plasmon waves propagating in all directions, it only ‘detects’ the cumulative near-field plasmonic signal arising underneath it. This stands in rough analogy with the operating principle of sonar echolocation. In the top panels of Fig. 2a, we sketch the plasmon interference pattern in the form of plasmon amplitude, revealing standing wave oscillations between the tip and sample edge. As the tip is scanned towards the edge, it registers these oscillations with periodicity given by \( \lambda_p/2 \), as shown in the bottom panels of Fig. 2a. Our plasmon interference interpretation is supported by theoretical estimates of the wavelength \( \lambda_p \). The plasmon dispersion of a two-dimensional metal residing at the interface of vacuum (dielectric constant \( \varepsilon_0 = 1 \)) and a substrate with dielectric function \( \varepsilon_{\text{sub}}(\omega) \) is given by the formula:

\[
q_p = \frac{i\omega\kappa}{2\pi\sigma}
\]

where \( \kappa(\omega) = \kappa_1 + i\kappa_2 \equiv [\varepsilon_0 + \varepsilon_{\text{sub}}(\omega)]/2 \) represents the complex effective dielectric function of the environment for graphene. Assuming that the conductivity \( \sigma \) of graphene takes a Drude form with relaxation time \( \tau \), equation (1) can be rewritten as:

\[
q_p = \frac{\hbar^2\kappa(\omega)}{2\varepsilon_0\varepsilon_\text{F}} \omega(\omega + i\tau)
\]

where \( \hbar \) is the reduced Planck’s constant, \( e \) is the elementary charge, and \( E_\text{F} \) is the Fermi energy. Derivation of these equations and further refinements are discussed in the Supplementary Information. The real part of \( q_p = q_1 + iq_2 \) determines the plasmon wavelength \( \lambda_p = 2\pi/q_1 \), and the ratio between the imaginary part and the real part defines the plasmon damping rate \( \gamma_p = q_2/q_1 \). In graphene, the Dirac-like dispersion of the Fermi energy \( E_\text{F} = \hbar^2v_\text{F}k^2/2m^* \) with Fermi momentum \( k^2 = \sqrt{n/[\varepsilon_\text{F}]^2} \) implies \( n^{-1/2} \) scaling of the plasmon momentum with the carrier density \( n \) at fixed \( \omega \). Here \( v_\text{F} \approx c/300 = 10^6 \text{ m s}^{-1} \) is the Fermi velocity. Finally, using frequency \( \omega = 892 \text{ cm}^{-1} \) and \( n \approx 8 \times 10^{12} \text{ cm}^{-2} \) determined from the micro-Raman probe (see below) at the graphene edge, we find \( \lambda_p \approx 200 \text{ nm} \) from equation (2), which is roughly twice the distance between fringes in Fig. 1b–e.

The images in Fig. 1b–e contain rich insights into processes governing plasmon propagation and losses on the surface of graphene. It is therefore instructive to examine line profiles along the direction normal to the sample edges. In Fig. 2b we show a plot obtained by averaging 150 such profiles—a procedure used to improve the signal-to-noise ratio. We find that the fringe widths increase from the interior to the edge of graphene, implying that the plasmon wavelength \( \lambda_p \) likewise increases. This behaviour is due to enhancement of the carrier density \( n \) near the sample edge, which is verified by our micro-Raman experiments (Fig. 2b inset). Thus, plasmonic interference patterns reported in Fig. 2b uncover the usefulness of infrared imaging for the nanoscale determination of local carrier density in graphene. In Fig. 2b we also show modelling results of plasmon profiles following a procedure detailed in the Supplementary Information. Our modelling provides a quantitative account of plasmon interferometry data. The carrier density profile (red curve in Fig. 2b inset) and the damping rate \( \gamma_p \) constitute the adjustable parameters of the model. Because plasmons in our experiments are launched and detected by the same point source (the AFM tip), the interference amplitude necessarily exhibits decay from the sample edge even when the damping rate is assumed to be vanishingly small (green trace in Fig. 2b). The best fit to the amplitude profile is achieved for significantly stronger damping, with \( \gamma_p = 0.135 \).

According to equations (1) and (2), the plasmon wavelength \( \lambda_p \) is directly determined by the carrier density \( n \). We demonstrate this
unique aspect of graphene plasmonics experimentally through imaging under gate bias (Fig. 3a). Over a range of \( V_g \) (gate voltage) values from \(+30\) V to \(-20\) V, the hole density \( n \) in our samples increases monotonically, a consequence of significant unintentional hole doping present even in ungated graphene/SiO\(_2\)/Si structures (Fig. 2b inset). This tuning of carrier density produces systematic variations in the plasmonic profiles: fringe amplitude and periodicity are both enhanced with increasing \( n \). By inferring \( \lambda_p \) directly from the fringe width, we observe a systematic decrease in \( \lambda_p \) with the reduction in hole density. Our gate-dependent data for \( \lambda_p \) approximately follow the \( \lambda_p \propto |n|^{1/2} \) law predicted for monolayer graphene\(^9\). In contrast, the plasmon damping rate does not show clear gate dependence and is roughly equal to \( 0.135 \pm 0.015 \) at all \( V_g \). This magnitude of \( \gamma_p \) significantly exceeds theoretical estimates for graphene with similar electronic mobility, \( \mu = 8,000\) cm\(^2\) V\(^{-1}\)s\(^{-1}\) (ref. 12).

It is important to understand why plasmon damping in our structures is abnormally strong. According to equations (1) and (2), two additive contributions define damping rate as \( \gamma_p = (\sigma_1/\sigma_2) + (\kappa_c/\kappa_1) \). The first term is associated with plasmonic losses inherent in graphene, whereas the second term describes losses due to the SiO\(_2\) substrate. At \( \omega = 892\) cm\(^{-1}\), we estimate \( \kappa = 2.52 + 0.13i \) and hence, \( \kappa_c/\kappa_1 \approx 0.05 \), based on our ellipsometric measurements of SiO\(_2\)/Si wafers. The resulting value of \( \sigma_1/\sigma_2 = 0.08 \) is unexpectedly high, being three to four times higher than the estimate of \( \sigma_1/\sigma_2 = (\omega e)^{-1} \) obtained from equation (2) using the relaxation rate \( \tau^{-1} \approx 20\) cm\(^{-1}\), corresponding to a typical d.c. mobility of our samples. This discrepancy has two possible interpretations. Excessive losses may originate from an enhanced electronic relaxation rate at infrared frequencies compared to that established in d.c. transport. Alternatively, losses may be unrelated to free carrier mobility/dynamics and may instead be associated with extrinsic factors such as surface irregularities. Our plasmonic interferometry data in Figs 2 and 3 provide strong support for the former hypothesis. Indeed, these images yield \( \gamma_p \) and \( \lambda_p \) (Fig. 3b) and thus allow us to determine the complex optical conductivity of graphene (Fig. 3b inset) based on the following formula: \( \sigma_2 = \frac{e \omega k}{4\pi \varepsilon_0 \lambda_p^2} \left( \sigma_1 \kappa_2 - \kappa_s \kappa_1 \right) \). We remark that these relations between plasmonic parameters (\( \lambda_p, \gamma_p \)) and the complex optical conductivity \( \sigma \) of graphene hold true for any plasmonic medium for which \( \sigma_1 \ll \sigma_2 \). Therefore these expressions apply even if the frequency-dependent conductivity deviates from the simple Drude model. The appeal of this analysis lies in establishing a link between real-space plasmonic profiles and the optical constants inferred from conventional infrared spectroscopy.

Infrared nano-imaging experiments reported here have established graphene/SiO\(_2\)/Si structures as a potent plasmonic medium that enables voltage control of both the wavelength and the amplitude of the plasmons. Higher gate voltages than used in our study will allow for the binary on/off switching of plasmon propagation with a possibility of potentially local control by a top gate or a biased tip. The plasmon wavelength in graphene, \( \lambda_p \approx 200\) nm, is one of the shortest imaged for any material, whereas the propagation length is on a par with Au in experiments monitoring strongly confined plasmons launched by AFM tips\(^{24}\). An important figure of merit, \( \lambda_{0B}/\lambda_p \approx 50-60 \), for our back-gated devices surpasses that of conventional Ag-based structures\(^2\). Intrinsic plasmonic losses in graphene that we analysed in Fig. 4 show that the unexpectedly large magnitude of \( \sigma_1 \) this result supports the notion of prominent many-body effects in graphene beyond the picture of non-interacting Dirac fermions\(^{24-25}\). Further experiments on suspended graphene, as well as devices using various types of dielectric substrates (such as hexagonal boron nitride), are needed to disentangle the roles of electron–electron and graphene–substrate interactions in the dissipation we observe at infrared frequencies. Our work uncovers an experimental path and analysis methodology for these future studies of many-body interactions in graphene.

Infrared nano-imaging experiments reported here have established graphene/SiO\(_2\)/Si structures as a potent plasmonic medium that enables voltage control of both the wavelength and the amplitude of the plasmons. Higher gate voltages than used in our study will allow for the binary on/off switching of plasmon propagation with a possibility of potentially local control by a top gate or a biased tip. The plasmon wavelength in graphene, \( \lambda_p \approx 200\) nm, is one of the shortest imaged for any material, whereas the propagation length is on a par with Au in experiments monitoring strongly confined plasmons launched by AFM tips\(^{24}\). An important figure of merit, \( \lambda_{0B}/\lambda_p \approx 50-60 \), for our back-gated devices surpasses that of conventional Ag-based structures\(^2\). Intrinsic plasmonic losses in graphene that we analysed in detail can be substantially reduced or even eliminated through population inversion\(^{25}\). We stress that plasmon tuning is realized here in the architecture of a metal-oxide-semiconductor device (we used graphene-oxide-silicon), which is a ubiquitous system in modern information processing. Furthermore, the performance of even the first generation of plasmonic devices reported here and in ref. 27 is rather promising compared to non-tunable metal-based structures\(^2\). For all these reasons, we believe that graphene may be an ideal medium for active infrared plasmonics.

METHODS SUMMARY

Infrared nano-imaging. Our scattering-type SNOM apparatus (Naspec) is based on an AFM operating in tapping mode. Measurements were performed at an AFM tapping frequency of \( \Omega = 270\) kHz and a tapping amplitude of 40 nm. The shaft of the conducting AFM tip acts as an antenna that boosts the efficiency of
near-field interaction. The back-scattered signal registered by the detector is strongly dependent on the tip–sample distance. This enables isolation of the genuine near-field contribution from the overall back-scattered signal, which is periodically modulated at harmonics of the tapping frequency . The th harmonic component of this signal , termed the th demodulated signal (here ), represents the desired near-field contribution.

**Samples and devices.** Our graphene samples were obtained by mechanical cleavage of bulk graphite and then transferred to SiO _2_/Si substrate. To avoid surface contamination by lithographic procedures, bulk graphite connected to our graphene sample was used as an electrode in our back-gating experiments. To verify the gating functionality of our devices, we first performed a spectroscopic study of the hybrid plasmon–phonon resonance at various gate voltages and found good agreement with the published data . Transport measurements of graphene samples fabricated following identical procedures indicate that the typical mobility of our graphene samples is about cm _2/_V s . Plasmon imaging experiments were completed for more than 30 graphene samples. All these structures exhibited highly reproducible behaviour and consistent values for the plasmon wavelength and damping. The data displayed in Figs 1–3 were obtained for devices with some of the weakest damping, revealing the largest number of plasmonic oscillations. Nevertheless, even in these devices the plasmonic losses are stronger than expectations based on typical electronic mobility measurements.

**Micro-Raman measurements.** According to previous studies, the position of the G-peak in the Raman spectrum of graphene is directly linked to its carrier density . Therefore, the G-peak profile shown in Fig. 2b inset reflects the range of the variation in graphene carrier density close to the edge. Plasmon imaging experiments were carried out using a Renishaw inVia Raman microscope equipped with a 50×, NA = 0.75 long-distance objective, a 1,800 lines per mm grating and an XY stage with a resolution of 100 nm. The spot size in these experiments is limited by diffraction. Therefore, the fragment of the line profile of the G-peak frequency shown in Fig. 2b inset is instrumentally broadened.

**Received 29 February; accepted 17 May 2012.**

Published online 20 June 2012.

1. Abwater, H. A. The promise of plasmonics. *Sci. Am.* 296, 56–62 (2007).
2. West, P. R. et al. Searching for better plasmonic materials. *Laser Photon. Rev.* 4, 795–808 (2010).
3. Stockman, M. I. Nanoplasmonics: the physics behind the applications. *Phys. Today* 64, 39–44 (2011).
4. Maier, S. A. *Plasmonics: Fundamentals and Applications* Ch. 4 (Springer, 2007).
5. Schuller, J. A. et al. Plasmonics for extreme light concentration and manipulation. *Nature Mater.* 9, 193–204 (2010).
6. Nagaar, P., Lindquist, N. C., Oh, S.-H. & Norris, D. J. Ultrasmooth patterned metals for plasmonics and metamaterials. *Science* 325, 594–597 (2009).
7. Lai, S., Link, S. & Halas, N. J. Nano-optics from sensing to waiguiding. *Nature Photon.* 1, 641–648 (2007).
8. Castro Neto, A. H., Guinea, F., Peres, N. M. R., Novoselov, K. S. & Geim, A. K. The electronic properties of graphene. *Rev. Mod. Phys.* 81, 109–162 (2009).
9. Wang, F. et al. Gate-variable optical transitions in graphene. *Science* 320, 206–209 (2008).
10. Li, Z. G. et al. Dirac charge dynamics in graphene by infrared spectroscopy. *Nature Phys.* 4, 532–535 (2008).
11. Vakil, A. & Engheta, N. Transformation optics using graphene. *Science* 332, 1291–1294 (2011).
12. Jablan, M., Buljan, H. & Soljačić, M. Plasmonics in graphene at infrared frequencies. *Phys. Rev. B* 80, 245435 (2009).