Optical nano-imaging of gate-tunable graphene plasmons

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The ability to manipulate optical fields and the energy flow of light is central to modern information and communication technologies, as well as quantum information processing schemes. However, because photons do not possess charge, a way of controlling them efficiently by electrical means has so far proved elusive. A promising way to achieve electric control of light could be through plasmon polaritons—coupled excitations of photons and charge carriers—in graphene1–5. In this two-dimensional sheet of carbon atoms6, it is expected that plasmon polaritons and their associated optical fields can readily be tuned electrically by varying the graphene carrier density. Although evidence of optical graphene plasmon resonances has recently been obtained spectroscopically7,8, no experiments so far have directly resolved propagating plasmons in real space. Here we launch and detect propagating optical plasmons in tapered graphene nanostructures using near-field scattering microscopy with infrared excitation light9–11. We provide real-space images of graphene nanostructures using near-field scattering microscopy. Our scattering-type SNOM comprises an atomic force microscope (AFM) in which the metallized tip is illuminated with a focused infrared laser beam (Fig. 1a). The backscattered radiation is recorded simultaneously with the topography, yielding nanoscale resolved infrared near-field images (Methods). A representative near-field image is shown in Fig. 1b, where the tip is scanned over a tapered graphene ribbon on the carbon-terminated surface of 6H-SiC (ref. 20), illuminated by light of free-space wavelength $\lambda_0 = 9.7 \mu m$. One of the most distinct features in this image is the presence of fringes parallel to the edge of the ribbon in its wider part. The distance between fringe maxima is approximately constant at ~130 nm inside the ribbon. We interpret these fringes as follows: the near field at the tip apex locally excites radial surface waves that propagate along the surface and reflect at the edges, partially reaching the domain. However, high-resolution nanoscale real-space imaging of the plasmonic modes is of fundamental importance to conclusive unveiling of propagating and localized plasmons in graphene sheets and nanostructures.

Here we visualize propagating and localized graphene plasmons in real space by scattering-type scanning near-field optical microscopy16,19 (scattering-type SNOM). Our scattering-type SNOM comprises an atomic force microscope (AFM) in which the metallized tip is illuminated with a focused infrared laser beam (Fig. 1a). The backscattered radiation is recorded simultaneously with the topography, yielding nanoscale resolved infrared near-field images (Methods). A representative near-field image is shown in Fig. 1b, where the tip is scanned over a tapered graphene ribbon on the carbon-terminated surface of 6H-SiC (ref. 20), illuminated by light of free-space wavelength $\lambda_0 = 9.7 \mu m$. One of the most distinct features in this image is the presence of fringes parallel to the edge of the ribbon in its wider part. The distance between fringe maxima is approximately constant at ~130 nm inside the ribbon. We interpret these fringes as follows: the near field at the tip apex locally excites radial surface waves that propagate along the surface and reflect at the edges, partially reaching the domain. However, high-resolution nanoscale real-space imaging of the plasmonic modes is of fundamental importance to conclusive unveiling of propagating and localized plasmons in graphene sheets and nanostructures.

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Figure 1 | Imaging propagating and localized graphene plasmons by scattering-type SNOM. a, Diagram of the experimental configuration used to launch and detect propagating surface waves in graphene (represented as blue rings). The metallized AFM tip (shown in yellow) is illuminated by an infrared laser beam with wavelength $\lambda_0$. b, Near-field amplitude image acquired for a tapered graphene ribbon on top of 6H-SiC. The imaging wavelength is $\lambda_0 = 9.7 \mu m$. The tapered ribbon is 12 \(\mu m\) long and up to 1 \(\mu m\) wide. c, Colour-scale image of the calculated local density of optical states (LDOS) at a distance of 60 nm from the graphene surface, and assuming substrate $\varepsilon_s = 1$. Simulation fitting parameters: graphene mobility $\mu = 1,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and Fermi energy $E_F = 0.4 \text{ eV}$. 

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the tip again. By recording the local field scattered by the tip, we probe the interference of forward- and backward-propagating plasmons. Within this basic physical picture, the maxima are separated by half the plasmon wavelength, \( \lambda_p/2 \). Thus, we experimentally find a plasmon wavelength \( \lambda_p = 260 \text{ nm} \), which is about a factor of 40 smaller than the free-space excitation wavelength. As we discuss in Methods, the near-field images can be interpreted in terms of the local density of optical states (LDOS). The calculated LDOS for a tapered ribbon is shown in Fig. 1c, matching very closely the experimental results (Fig. 1b), including the features at the narrower part of the ribbon.

Our observation of a remarkably strong reduction in the plasmon wavelength, \( \lambda_p \approx \lambda_0/40 \), can directly be attributed to the two-dimensionality and the unique conductance properties of graphene. Namely, the plasmonic properties of graphene are related to its optical conductivity \( \sigma \) (for example, \( 2\pi\lambda_p = (e_r + 1)\omega/(4\pi\text{Im}(\sigma)) \), where \( e_r \) is the substrate permittivity and \( \omega \) is the frequency). For sufficiently high doping, quantified through a Fermi energy \( E_F \) exceeding the plasmon energy \( E_p \), this yields \(^4\): \( \lambda_p = \lambda_0/40 \).

\[
\lambda_p \approx \frac{2\pi}{E_p} \frac{\frac{4}{E_p e_r + 1}}
\]

We note that this simplified equation reveals a relation between the plasmon wavelength and the free-space wavelength that is governed by the fine-structure constant, \( \alpha \approx 1/137 \). The observed \( \lambda_p = 260 \text{ nm} \) is in reasonable agreement with the theoretical prediction of equation (1) for the specific substrate (for SiC: 1.9 for \( \lambda_0 = 9.7 \mu\text{m} \)), which yields \( \lambda_p = 305 \text{ nm} \) assuming \( E_p = 0.4 \text{ eV} \). This value is about a factor of two higher than the intrinsic substrate-induced doping found in earlier studies of graphene on the carbon-terminated surface of 6H-SiC (ref. 22). We speculate that narrow ribbons exhibit larger carrier densities, and this will be addressed in future studies.

In Fig. 2, we present a more detailed experimental study of the plasmon properties in graphene nanostructures by taking advantage of the strong dependence of the dielectric constant of the SiC substrate, \( \varepsilon_{SC} \), on the excitation wavelength. This allows us to tune the plasmon wavelength over a wide spectral range by just slightly changing the excitation wavelength, as the plasmon wavelength depends strongly on the dielectric constant of the substrate.

The near-field images of relatively wide ribbons are displayed in Fig. 2a, showing that the spacing of the interference fringes decreases considerably with increasing \( \varepsilon_{SC} \). This observation is qualitatively consistent with equation (1), as a larger substrate permittivity yields a smaller graphene plasmon wavelength. Quantitatively, we obtain good agreement between the plasmon wavelengths extracted from the near-field images (Fig. 3a, symbols) and the prediction of equation (1) for graphene on SiC (Fig. 3a, solid curves), using literature values for the dielectric constant of SiC (ref. 23) and an intrinsic doping \( E_F = 0.4 \text{ eV} \).

Our experimental observation of an extremely short plasmon wavelength compared to the excitation wavelength is associated with an extraordinary confinement of the infrared field perpendicular to the graphene sheet, characterized by a decay length \( \delta = \lambda_p/2\pi \) (ref. 17). This means that narrow graphene ribbons are ideally suited to confine light down to extremely small volumes. In Fig. 2b, we show near-field images of the tapered ribbons where the width \( W \) reaches values smaller than the plasmon wavelength \( \lambda_p \). These images clearly reveal two distinct localized modes (indicated by red and white arrows) that coexist with a resonant enhancement of the near-field signal, comparable to the observations in ref. 24. The resonance condition depends on \( \lambda_p \) and the ribbon width \( W \), as we observe a clear shift of the localized modes to a wider part of the ribbons for increasing \( \lambda_p \).

The width \( W \) for which these two modes occur, normalized to the plasmon wavelength \( \lambda_p \), is shown in Fig. 3b, from which we extract the resonance conditions \( W \approx 0.3\lambda_p \). These images clearly reveal two distinct localized modes (indicated by red and white arrows) that coexist with a resonant enhancement of the near-field signal.

To better understand the physical mechanisms that underlie the observation of these resonant optical modes, as well as the interference of the strong dependence of the dielectric constant of the SiC substrate, \( \varepsilon_{SC} \), on the excitation wavelength. This allows us to tune the plasmon wavelength over a wide spectral range by just slightly changing the excitation wavelength, as the plasmon wavelength depends strongly on the dielectric constant of the substrate.

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Figure 2 | Controlling the plasmon wavelength over a wide range.

(a) Coloured plots show near-field optical images taken with imaging wavelengths (\( \lambda_0 \)) of 9,200 nm (left), 9,681 nm (middle) and 10,152 nm (right), corresponding respectively to SiC dielectric constants of 2.9, 2.0 and 0.7.

(a) Images of a graphene ribbon \( \sim 1 \mu\text{m} \) wide, revealing a strong dependence of the fringe spacing, and thus plasmon wavelength, on the excitation wavelength;

(b) Images of a tapered graphene ribbon; both ribbons are on the same 6H-SiC substrate. The topography (obtained by AFM) is shown in greyscale in the leftmost and rightmost panels, and outlined by dashed lines in the central, coloured panels. The line traces in the leftmost and rightmost panels, and outlined by dashed lines in the central, coloured panels. The line traces in the leftmost and rightmost panels, and outlined by dashed lines in the central, coloured panels. The line traces in the leftmost and rightmost panels, and outlined by dashed lines in the central, coloured panels.

Red and white arrows indicate the resonant localized modes.
fringes, we calculate maps of the local density of optical states (LDOS) of the graphene ribbons. The LDOS maps calculated for two different values of the substrate permittivity $\varepsilon_{r,SiC}$ are shown in Figs 1c and 3c. As in the experimental scattering-type SNOM images, the LDOS maps reveal interference fringes parallel to the ribbon edge, and localized modes near the tip of the ribbon. The fringe spacing matches quantitatively the experimental results and the spacing increases with decreasing $\varepsilon_{r}$, associated with an increase in $\lambda_p$, as predicted by equation (1). The good agreement between experiment and theory confirms that the fringes in the wider part of the ribbon are due to plasmon interference caused by plasmon reflections at the graphene edges. We remark that both the LDOS and the experimental images exhibit their maximum away from the graphene edge, and that the fringe spacing increases slightly closer to the edge. This can be explained by the electromagnetic boundary conditions at the edges (further discussed below) and the fact that the plasmon wavevector perpendicular to the edge does not have a single value, but rather a finite distribution around $2\pi/\lambda_p$ (see Supplementary Information).

The comparison between the calculated LDOS maps and the experimental data in Fig. 2 can be used to estimate plasmon propagation distances. We observe five well-defined interference fringes away from a single edge. The peak close to the edge is relatively strong, which we attribute to enhanced near-field coupling between tip and graphene close to the edge, and multiple plasmon reflections between the metallic tip and the edge. The fringes inside the ribbon decay, owing to both the circular character of the plasmons and intrinsic losses. These observations are consistent with our LDOS calculations for plasmon losses corresponding to a mobility of 1,200 cm$^2$/V$^{-1}$s$^{-1}$ (ref. 4). This value was obtained by electrical characterization of similar graphene ribbons under ambient experimental conditions. In particular, the observed coexistence of a strong reduction in plasmon wavelength (and thus strong optical field confinement) and relatively long propagation distance is very promising, and a unique feature of plasmons carried by graphene. We emphasize that much longer propagation distances are expected for higher-mobility graphene.

In our LDOS model interpretation, the localized modes near the tip of the graphene ribbon (marked by arrows in Fig. 2b) are explained as localized graphene plasmon resonances, which occur for specific conditions $W/\lambda_p$ extracted from localized-mode measurements. Red crosses and black circles correspond to the modes indicated by red and white arrows in Fig. 2, respectively. The spatial distribution of the LDOS calculated for homogeneous ribbons of increasing width (from bottom to top), supported on a dielectric with $\varepsilon_r = 3$ (left) or $\varepsilon_r = 0.5$ (right). The ribbon width of the two lowest-order modes is shown in units of the plasmon wavelength of extended graphene, $\lambda_p$. The resonance $E_2 = 0.2$ eV and $E_2 = 0.4$ eV on a SiC-6H substrate. Green dashed line, SiC substrate permittivity. b. Experimentally obtained resonance values of the ribbon width ($W = 0.37\lambda_p$ and $0.82\lambda_p$) as well, where the strong concentration of the electromagnetic field yields an enhanced plasmon–dipole interaction and, therefore, an increase in the near-field signal. For both theory and experiment, the profiles of the two localized modes are distinctly different from those of conventional Fabry–Perot cavity modes. For example, the lowest-order mode (indicated by white arrows in Fig. 2b) exhibits field maxima at the graphene edges, whereas for a conventional lowest-order Fabry–Perot mode the field is maximum in the middle. This is because graphene plasmons are being reflected at the boundaries with a reflection coefficient of approximately one (zero phase), rather than the coefficient of minus one (π phase) characteristic of the conventional Fabry–Perot model (see Supplementary Information).

One of the most appealing advantages of graphene plasmonics is the capability to control and switch nanoscale optical fields in situ. Here we demonstrate very effective electrical control of nanoscale optical fields by applying an electric field perpendicular to the graphene sheet, which allows us to vary the carrier density in the ribbon. To this end, we have fabricated tapered ribbons based on CVD-grown graphene on a SiO$_2$ substrate with a Si backgate. By applying a backgate voltage $V_B$, we tune the carrier density and thus the Fermi energy $E_F = (V_B - V_D)^2$, where $V_D$ is the voltage that needs to be applied to offset the intrinsic doping (that is, to reach the Dirac point; $V_D$ is extracted from optical measurements, as we discuss below). The effect of changing $V_B$ on the near-field images is shown in Fig. 4a, where the Fermi energy is tuned over a wide range, from about 0 to 0.15 eV. For $V_B - V_D > 10$ V, the general near-field features are comparable to those of ribbons on SiC substrates, including the two local ribbon resonances indicated by white and red arrows in Fig. 4a. By increasing $V_B$, we find that the resonances (signal maxima) shift towards larger ribbon width, which we attribute to an increase in plasmon wavelength when the carrier density, and thus also the Fermi energy, increases (see equation (1)). The extracted value of $\lambda_p$ as a function of gate voltage is shown in the lower part of Fig. 4b (red circles correspond to the tapered ribbon shown in Fig. 4a, and green crosses correspond to additional ribbons shown in Supplementary Information). The calculated plasmon dispersion, represented by the blue contour plot in the bottom part of Fig. 4b, is based on the random-phase approximation, which includes...
Supplementary Information. Top panel, diagrams showing the Dirac cone of plasmon wavelength experimentally extracted from localized mode resonances (vertical axis), while changing arrows. The illumination wavelength is fitting the data to the model. Localized modes are indicated by white and red the left panel of Fig. 4a (corresponding to Experimentally, we clearly observe very strong plasmon damping in the ribbon compared to the thickness of the oxide (see Supplementary Information). The amplitude is shown in the near-field images in Figs 1, 2 and 4. These excitations can strongly damp the plasmons, but to first order these transitions are suppressed for \(|E_p| \leq E_p\). The data agree well with the calculated plasmon dispersion, taking into account an electrostatic enhancement of the carrier concentration due to the narrow width of the ribbon compared to the thickness of the oxide (see Supplementary Information).

The effect of plasmon damping offers the intriguing capability to actively switch graphene plasmons on and off by electric fields. Experimentally, we clearly observe very strong plasmon damping in the left panel of Fig. 4a (corresponding to \(E_p \leq E_p\)), where the ribbon does not show any signal compared to the substrate. We illustrate electrostatic switching of graphene plasmons in more detail in Fig. 4c, which shows line scans across a ribbon of width \(W = 200\) nm (vertical axis), while changing \(V_B\) (horizontal axis). At the Dirac point \((V_B = V_D)\), the near-field signal is dramatically depleted on the whole ribbon. With increasing Fermi energy, plasmon modes emerge at both sides of the Dirac point. These modes are attributed to plasmons carried by either p- or n-type charge carriers. The calculated LDOS profiles as a function of Fermi energy (lower plot of Fig. 4c) is in excellent agreement with the experimental observations.

Here and in ref. 24, electrical control of confined and propagating plasmons is demonstrated, thus providing a solution to a major problem in plasmonics, as it facilitates the design and miniaturization of active nanoscale photonic devices\(^{26,27}\). This leads to a new paradigm in optical and opto-electronic telecommunications and information processing. As an alternative to plasmon excitation and detection by (effective) dipoles, plasmons can also be resonantly excited by light in graphene nanocavities\(^{15}\), enabling strong enhancement of light absorption in graphene\(^{28}\), and a new basis for infrared detectors and light-harvesting devices.

**METHODS SUMMARY**

**Plasmon nano-imaging.** The scattering-type SNOM used for this work (from Neaspec GmbH) employs metal AFM tips as near-field probes, which are illuminated by infrared light from a grating-tunable CO2 laser. The tip acts as an optical antenna that converts the incident light into a localized near field below the tip apex\(^{11}\). The nanoscale field concentration provides the required momentum\(^{12,28}\) for launching plasmons on graphene, as illustrated in Fig. 1a. Plasmon reflection at the graphene edges produces plasmon interference, which is imaged by recording the light elastically scattered by the tip with a pseudo-heterodyne interferometer\(^{15}\). In order to suppress background scattering from the tip shaft and the sample, the tip is vibrated vertically with 50-nm amplitude at a frequency of about \(\Omega = 300\) kHz, and the detector signal is demodulated at a higher harmonic of \(\Omega\). With this technique, we record the amplitude and phase of the scattered field (see Supplementary Information). The amplitude is shown in the near-field images in Figs 1, 2 and 4.

**LDOS calculations.** In order to understand the near-field images, we model the microscope tip as a vertically oriented point dipole\(^{11}\), which is scanned 60 nm above the graphene. The dipole launches plasmons\(^{25,26}\) that are reflected at the ribbon edges. These plasmons act back on the tip, and are subsequently scattered into photons, which we detect. The detected signal is strongly correlated with the vertical component of the LDOS. We calculate the LDOS by solving the Maxwell equations for a dipole source \(p\) at location \(\mathbf{r}\):

\[
\text{LDOS} = \text{LDOS}_{\text{ref}} + \frac{1}{2\pi^2\epsilon_0|p|^2} \Im \left\{ E_{\text{ref}}(\mathbf{r}) \cdot p^* \right\}
\]

where \(E_{\text{ref}}\) is the field reflected by nearby structures and evaluated at the position of the source dipole. This is the procedure we actually follow to obtain the LDOS in this work, and \(E_{\text{ref}}\) is calculated by means of the boundary-element method for a dipole source. In order to simulate two-dimensional LDOS maps for a dipole 60 nm above a tapered graphene ribbon, we combine one-dimensional LDOS profiles of graphene ribbons of fixed width. We justify this approach on the
grounds that the ribbon width along the long axis of the graphene triangle varies adiabatically, yielding a weak reflection at the tip of the ribbon. In addition, the plasmon propagation distance is of the order of a few plasmon wavelengths. Owing to these two effects, the intensity of back-reflected plasmons from the tip of the ribbon is expected to be weak. This is consistent with the absence of modulations in the experimental near-field images.

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