GRAPHENE PLASMONICS: MANIPULATING LIGHT AT THE NANOSCALE WITH A ONE-ATOM-THICK MATERIAL

N.M.R. Peres[1,2,3], P.A.D. Gonçalves[4], T. Stauber[5] and N. Asger Mortensen[3,6] – DOI: https://doi.org/10.1051/epn/2024406

[1] Centro de Física (CF-UM-UP) and Departamento de Física, Universidade do Minho, P-4710-057 Braga, Portugal
[2] International Iberian Nanotechnology Laboratory (INL), Av Mestre José Veiga, 4715-330 Braga, Portugal
[3] POLIMA – Center for Polariton-driven Light-Matter Interactions, University of Southern Denmark, Campusvej 55, DK-5230 Odense M, Denmark
[4] ICFO – Institut de Ciencies Fotoniques, The Barcelona Institute of Science and Technology, 08860 Castelldefels (Barcelona), Spain
[5] Instituto de Ciencia de Materiales de Madrid, CSIC, E-28006 Madrid, Spain
[6] Danish Institute for Advanced Study, University of Southern Denmark, Campusvej 55, DK-5230 Odense M, Denmark

Surface plasmon polaritons (SPPs), collective oscillations of the electron density coupled to electromagnetic fields at metallic surfaces, enable the confinement of electromagnetic radiation at subwavelength scales. Plasmonics, the subfield of nanophotonics that studies SPPs, leverages these capabilities to investigate the fundamental processes governing light–matter interactions and to engineer nanoscale optoelectronic devices.

Plasmons have been observed in various material platforms, including conventional metals and doped semiconductors in both two and three dimensions. Over the past decade or so, the observation of electrically tunable plasmons in graphene—a one-atom-thick material made from carbon atoms arranged in a honeycomb lattice—featuring exquisite optical properties has opened new avenues for manipulating light at the nanoscale, holding immense potential for fundamental research in quantum nanophotonics and for emergent quantum technologies.

The early days

Immediately after its discovery in 2004 [1], graphene generated significant interest among physicists due to its remarkable electrical, optical, thermal, and mechanical properties. In the initial years following graphene's isolation, most research focused on its exotic quantum transport properties, stemming from its unique electronic structure, where charge carriers in graphene behave as relativistic particles without mass, dubbed massless Dirac fermions.

The scope of research in two-dimensional (2D) materials saw a significant expansion during the 2010s. The first observation of graphene plasmons using optical spectroscopy was reported by Ju *et al.* in 2011 [2]. Their devices consisted of arrays of graphene microribbons that could be electrically doped using an ion-gel gate (Fig. 1A–B). By patterning graphene into a ribbon array, the authors effectively bridged the kinematic mismatch between light in free space and graphene plasmons, thereby exciting graphene plasmons, which imprinted well-defined resonances in the terahertz (THz) absorption spectrum (Fig. 1C). These resonances could be tuned by changing the ribbon-width as well as graphene's electronic density, in agreement with theoretical predictions made years earlier [3]. In particular, the frequency of the observed graphene plasmons scaled with the carrier density (n) as $n^{1/4}$, in contrast with the $n^{1/2}$ scaling observed in conventional 2D electron gases (2DEGs) [3]. This result is a direct consequence of massless Dirac fermion nature of charge carriers in graphene. Moreover, the observation of plasmons in graphene at room temperature was surprising, since in conventional 2DEGs these could only be observed at cryogenic temperatures.

Following this, the field evolved quite rapidly. The next challenge was to directly image graphene plasmons and spatially resolve their electric-field distribution at the nanoscale. This feat was achieved in 2012 by two independent collaborations [4]. In both cases, the researchers used a scattering-type scanning near-field optical microscope (s-SNOM) equipped with a sharp metallic tip to excite

and image plasmons in graphene. In such a scheme, the metallic tip of the s-SNOM is illuminated by an infrared (IR) laser ($\lambda_0 \approx 10$ μm), whose scattered light contains near-field components with sufficient momentum to excite highly confined graphene plasmons. The excited plasmons propagate along the graphene sheet until they are reflected at the material's edges and interfere with the "incoming" plasmons. The result of the interference between incoming and reflected plasmons leads to a standing-wave pattern. By scanning the s-SNOM over the sample, the researchers could resolve the real-space variations of the electric field below the tip, revealing interference fringes separated by half of the plasmon wavelength $\lambda_{GP}/2$. Notably, this technique gives *direct* access to the spatial properties of graphene plasmons (wavelength, confinement, damping, *etc.*), as well as to its dispersion relation $\omega_{GP}(q_{GP} = 2\pi/\lambda_{GP})$ (Fig. 2A). Both works confirmed the $\omega_{GP} \propto n^{1/4}$ dependence of the plasmon frequency on the carrier density observed earlier by Ju and co-workers, which is characteristic of graphene. Moreover, the two teams reported large confinement factors (λ_0/λ_{GP}), in the 40–60 range, thereby beating the diffraction limit ($\sim \lambda_0/2$) by more than one order of magnitude. Graphene plasmons, featuring relatively low-losses, made their real-space nanoimaging possible.

A mature field

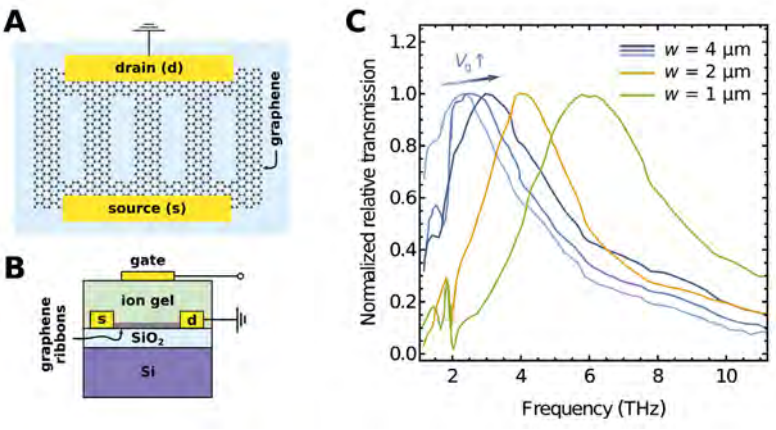
These pioneering studies sparked a new surge of interest in graphene-based plasmonics and nanophotonics, both theoretically and experimentally, further consolidating graphene plasmonics as a field of its own [5]. This enthusiasm was arguably instigated by the many alluring properties of graphene plasmons, including their capability to achieve deeply subwavelength light confinement, relatively long lifetimes, and the ability to tune the plasmon resonance *in situ* by controlling the gate voltage (even allowing switching on and off the plasmon resonances). Before long, graphene plasmons have been realized in various configurations, including graphene nanoribbons, nanodisks, nanorings, *etc.*, as well as in planar heterostructures fabricated by stacking different 2D materials on top of each other—known as van der Waals heterostructures (vdWhs)—, like atomically thin LEGO bricks. In particular, benefiting from the observation in previous transport experiments that the carrier mobility in graphene significantly improved upon encapsulating it between two thin films of hexagonal boron nitride (hBN), hBN-encapsulated graphene exhibited highly confined plasmons with unprecedentedly low damping [6], cementing graphene's reputation of a low-loss plasmonic material. Subsequently, while investigating various damping mechanisms that limit plasmon propagation (*e.g.*, electron–electron and electron–phonon scattering, extrinsic dielectric losses from the encapsulating material, *etc.*), Ni *et al.* imaged graphene plasmons with propagating lengths exceeding 10 μm (corresponding to 50 plasmon wavelengths in their experiment) in high-mobility, encapsulated graphene cooled to 60 K [7].

The large field confinement associated with graphene plasmons makes them appealing for sensing applications. While plasmons in conventional 3D metals typically display plasmon resonances around the visible region of the electromagnetic spectrum (more precisely, across the near-UV–near-IR range), plasmons in graphene span the THz to mid-IR spectral region. Besides naturally complementing metal-based plasmonics, this is particularly important because many molecules and compounds of interest possess vibrational modes in that spectral window. In a paper published in 2015, Rodrigo *et al.* demonstrated an electrically tunable graphene-plasmon-based biosensor for label-free detection of proteins [8], capable of achieving high sensitivity and outperforming conventional plasmonic sensors.

Furthermore, the unique band structure of graphene, featuring a linear energy–momentum dispersion at low energies, renders its response to external electromagnetic fields with a considerable degree of anharmonicity. This, together with the strong local field enhancement provided by plasmons in graphene has also attracted considerable interest in the exploration of nonlinear optical phenomena driven by graphene plasmons [X].

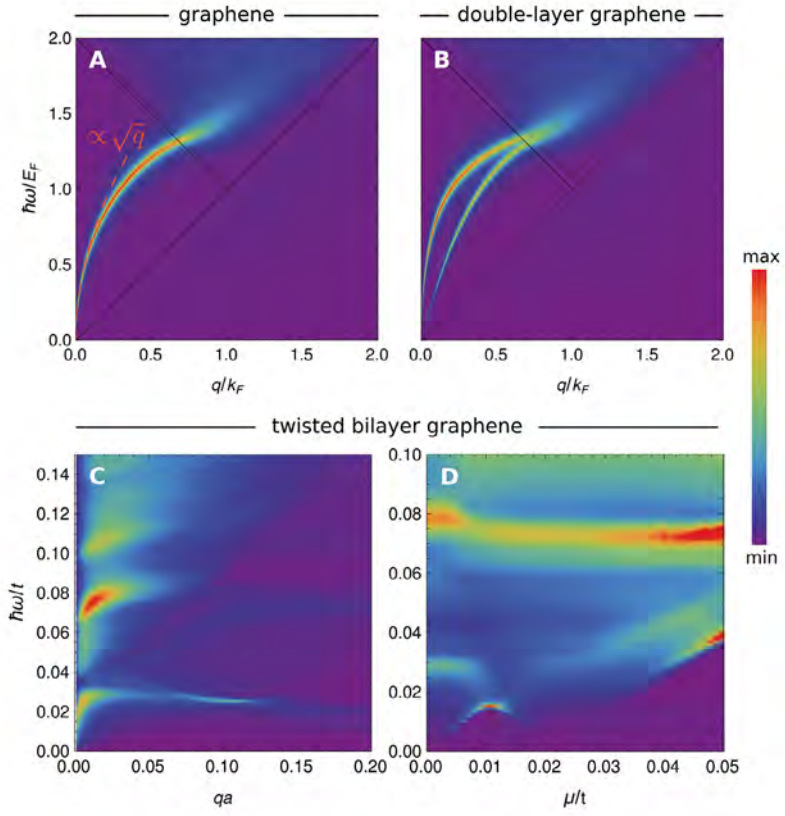
In another direction, the stacking of two graphene sheets separated by a thin dielectric spacer was also explored. In this configuration, the Coulomb interaction between plasmons in each graphene layer causes them to hybridize [5], resulting in two new coupled plasmon modes of different parities (Fig. 2B). The lowest energy mode, dubbed acoustic graphene plasmon (AGP) due to its nearly linear dispersion, was found to exhibit even stronger field confinement than what could be attained using plasmons in single-layer graphene. However, ● ● ●

▼ **FIG. 1:** (A–B) Schematics of graphene ribbon array used in Ju *et al.*'s experiment (A, top view; B, side view). (C) Relative transmission spectra (1 - T/T_{CNP}, where T_{CNP} denotes the transmission at the charge neutral point) for different ribbon widths (each spectrum is normalized to its maximum value). The control of the plasmon resonance by varying the gate voltage is shown in the 4-μm-wide ribbons. The data is taken from Ju *et al.* [2] (see original paper for details).



• • • its electric-field spatial profile makes it difficult to excite. In 2017, a study led by Koppens and Hillenbrand circumvented this obstacle by fabricating a device consisting of a graphene–dielectric–metal heterostructure [9]. In this setup, the collective charge oscillations underpinning graphene plasmons are screened by the nearby metal (image-charge effect), causing the system to behave as if it were two graphene sheets separated by twice the graphene–metal distance, thereby reshaping the plasmon dispersion and creating an AGP. In a subsequent publication, AGPs propagating at extremely slow velocities close to the electron Fermi velocity (with λ_{GP} approaching $\lambda_0/300$) were used to probe the nonlocal optical response of graphene as well as many-body effects governing the graphene electron liquid [10]. In the wake of these works, plasmon confinement down to the one-atom limit has been demonstrated in a system comprising an array of metallic rods separated from graphene by a single atomic layer of hBN [11]. Subsequently, Epstein *et al.* realized nanometer-scale acoustic graphene plasmon cavities with an impressive mode volume confinement factor of 5×10^{-10} [12] that could be excited from the far-field due to the use of silver nanocubes (which enable coupling from the far-field and create an "inverted" graphene-on-mirror cavity).

▼ **FIG. 2:** (A–B) Plasmon dispersion in single- and double-layer graphene (A and B, respectively), indicated by the sharp features in the colormap. (C) Plasmon dispersion in twisted bilayer graphene, for a twist angle of $\theta \approx 1.6°$. (D) Dependence of the plasmon energy in TBLG on the chemical potential (μ), for a fixed wavevector $qa = 0.02$. Here, $t = 2.78$ eV is the intralayer hopping amplitude (the interlayer hopping is taken as $t_\perp = -0.33$ eV). (A–B) is adapted from ref. [5], whereas (C–D) is adapted from ref. [17]. See references for further technical details.



Recent developments

Extending beyond the traditional applications of plasmonics, the ultrahigh field confinement of plasmons in graphene-based heterostructures can be leveraged to probe the intriguing quantum nonlocal (*i.e.*, momentum dependent) response of nearby materials, including metals [13] and superconductors [14].

The plasmonic properties of bilayer graphene, consisting of two electronically coupled graphene layers, can be further modified by twisting the carbon sheets relative to each other. Already in 2007, Lopes dos Santos *et al.* predicted that twisted bilayer graphene (TBLG) would exhibit a renormalization of the Fermi velocity upon rotating one the graphene sheets with respect to the other [15], inaugurating the field of twistronics. Although this prediction was experimentally confirmed about two years later, it took more than a decade for the field to be reignited. In 2018, the Jarillo-Herrero group and coworkers reported the observation of strongly correlated phenomena in TBLG when the "twist angle" between the two graphene layers was $\theta \approx 1.1°$ [16]. At this "magic angle", the electronic bands become extremely flat, suppressing the cost of kinetic energy, and thus many-body effects become important. These groundbreaking experiments led to a resurgence in interest in twistronics and prompted the investigation of moiré physics in various other van der Waals materials with no end in sight.

Against this backdrop, the investigation of the plasmonic response of TBLG was a natural development. In 2016, Stauber and Kohler discussed the formation of quasi-flat plasmonic bands of Landau-damped "interband plasmons" in TBLG [17] (Fig. 2C–D). Experimental observation came five years later [21], while plasmons in doped TBLG were already observed in 2017 [19]. In this context, the recent observation of slow plasmons in epitaxially grown TBLG [20] reflects the high tunability of the plasmonic response in moiré systems.

Final note

Many other significant achievements, both fundamental and applied, have been published in the literature. Unfortunately, it is not possible to cover all of this material in a short article. However, we hope that the references provided below offer a solid starting point for readers interested in exploring this fascinating and evolving field that merges atomically thin materials with nanophotonics. ■

About the Authors

Nuno Peres is a theoretical Condensed Matter Physicist. Co-author of 200+ papers and a book on the plasmonic properties of graphene. He has been dedicated to research on low dimensional quantum materials since 2004, focused on the electronic properties and light-matter interaction in these systems.

P. André D. Gonçalves is a postdoctoral researcher at ICFO – The Institute of Photonic Sciences, Spain. His research interests encompass several areas of quantum nanophotonics and condensed-matter physics. He obtained his PhD from the Technical University of Denmark (DTU) in 2019. He was awarded the DTU Young Researcher Award 2019 and the 2021 EPS-QEOD Thesis Prize for fundamental aspects.

Tobias Stauber is a Tenured Scientist at the Materials Science Institute of Madrid (ICMM) of the Spanish National Research Council (CSIC), Spain. His research focus is on electronic phases and nano-photonics in two-dimensional materials. He further acts as co-editor for "New Journal of Physics" and "European Physical Journal B".

In 2017, **Asger Mortensen** was appointed as a full professor and D-IAS Chair at the University of Southern Denmark (SDU). His work spans quantum electron transport, photonic bandgap materials, nanophotonics, and more. He is currently the scientific director at the DNRF Center of Excellence POLIMA, focusing on polariton phenomena.

References

[1] K. S. Novoselov *et al.*, *Science* **306**, 666 (2004)

[2] L. Ju *et al.*, *Nat. Nanotechnol.* **6**, 630 (2011)

[3] B. Wunsch *et al.*, *New J. Phys.* **6**, 318 (2006), E. H. Hwang and S. Das Sarma, Phys. Rev. B **75** (2007).

[4] J. Chen *et al.*, *Nature* **487**, 77 (2012). Z. Fei *et al.*, *Nature* **487**, 82 (2012)

[5] P. A. D. Gonçalves and N. M. R. Peres, *An Introduction to Graphene Plasmonics* (World Scientific, Singapore, 2016)

[6] A. Woessner *et al.*, *Nat. Mater.* 14, 421(2015)

[7] G. X. Ni *et al.*, *Nature* **557**, 530 (2018)

[8] D. Rodrigo *et al.*, *Science* **349**, 165 (2015)

[9] P. Alonso-González *et al.*, *Nat. Nanotechnol.* **12**, 31 (2017)

[10] M. B. Lundeberg *et al.*, *Science* **357**, 187 (2017)

[11] D. Alcaraz Iranzo *et al.*, *Science* **360**, 291 (2018)

[12] I. Epstein *et al.*, *Science* **368**, 1219 (2020)

[13] P. A. D. Gonçalves *et al.*, *Nat. Commun.* **12**, 3271 (2021)

[14] A. T. Costa *et al.*, *Proc. Natl. Acad. Sci.* **118**, e2012847118 (2021). Z. Sun, *et al.*, *Phys. Rev. Res.* **2**, 23413 (2020)

[15] J. M. B. Lopes dos Santos *et al.*, *Phys. Rev. Lett.* **99**, 256802 (2007)

[16] Y. Cao *et al.*, *Nature* 556, 43–50 (2018). Y. Cao *et al.*, *Nature* **556**, 80 (2018)

[17] T. Stauber and H. Kohler, *Nano Lett.* **16**, 6844 (2016)

[18] N. C. H. Hesp *et al.*, *Nat. Phys.* **17**, 1162 (2021)

[19] F. Hu *et al.*, *Phys. Rev. Lett.* **119**, 247402 (2017)

[20] T. Huang *et al.*, *Nature* **605**, 63 (2022)

[21] T. Stauber *et al.*, *Nano Lett.* **20**, 8711 (2020)

[22] J. D. Cox and F. J. García de Abajo, *Acc. Chem. Res.* **52**, 2536 (2019).