

Chasing Plasmons in Flatland

ABSTRACT: Two-dimensional layered crystals, including graphene and transition metal dichalcogenides, represent an interesting avenue for studying light-matter interactions at the nanoscale in confined geometries. They offer several attractive properties, such as large exciton binding energies, strong excitonic resonances, and tunable bandgaps from the visible to the near-IR along with large spin-orbit coupling, direct band gap transitions, and valley-selective responses.

The physics becomes even more interesting when two-dimensional (2D) materials are coupled to nanostructures that support surface plasmon polaritons (SPPs) or give rise to SPPs themselves, as in the case of graphene. SPPs are formed when photons are coupled to free electrons at the interface between a metallic and a dielectric material, thus creating surface waves that propagate (typically nm to μm) and transfer the energy of electromagnetic radiation to the nanoscale. In systems like metallic nanoparticles, localized SPPs can squeeze light below its diffraction limit and induce a strongly enhanced and confined near-field. With 2D materials, the community is exploring new opportunities offered by the tighter spatial confinement and relatively longer propagation distances with the advantage of a high tunability via electrostatic gating.¹ This adds up to the possibility to reach efficient coupling of light to plasmons by compressing surface polaritons,² and the strong exciton-plasmon or plasmon-phonon coupling.³ Growing advances have been reported in the past years thanks to new site-selective fabrication and stacking methods that combine exfoliation of flakes with chemical vapor deposition (CVD) to achieve vertically and laterally positioned 2D materials with precise control over their interlayer twist angle. Such control in fabrication has enabled further exploration of the interaction between plasmonic and 2D materials, with the combination increasingly used as the active component in nanoscale photonic devices. This Virtual Issue collects more than 30 articles published in *Nano Letters* on this topic in the past decade and demonstrates the rapid evolution of the community toward a deeper understanding of the underlying physics and a clear improvement of nanotechnologies based on the combination of nanoplasmonics and 2D materials

Interfacing, Interaction, and Coupling. Several works have been reported in the past years on the interaction between 2D materials and plasmonic nanostructures. One of the main results reported was the strong plasmon-induced enhancement of the Raman signal of graphene due to the enhanced near-field interaction between graphene itself and the plasmon modes. This interaction allows for an increase in absolute light absorption of 30% with up to 700-fold enhancement of the Raman response of the graphene.⁴ This mechanism can be used also to probe with subwavelength resolution the local near-field distribution in plasmonic systems, such as nanocavities. For instance, Heeg et al. showed that a suspended single layer of graphene on top of a dimer cavity can show an enhanced Raman signal of a factor 10^3 compared to the graphene on the substrate without plasmonic nanostructures.⁵ This allowed one to squeeze light in a region in which lateral size is 1 order of magnitude smaller than the

excitation wavelength, thus increasing the subwavelength resolution that we might achieve with plasmonic nanostructures alone.

Interfacing plasmonic nanostructures and 2D materials, such as molybdenum disulfide (MoS_2), can also enhance at the same time both the Raman scattering and the photoluminescence (PL).⁶ Moreover, the fluorescence emission of 2D systems can be increased by almost 1 order of magnitude thanks to plasmons.⁷ If we then couple a specific emitter to this kind of hybrid system, we might be able to enhance energy transfer mechanisms, which are fundamental, for instance, in single molecule detection and sequencing. In this direction, it was recently shown that tungsten diselenide (WSe_2) monolayer flakes exfoliated on top of a plasmonic waveguide can boost the Purcell factor of the individual emitters in the monolayer and also couple single photons to the waveguide modes through the excitation of localized excitons.⁸ Beyond sensing and sequencing, this approach might be useful to build single-photon detectors sensitive to a specific wavelength and might have a huge impact on sensor technologies applied to photonics-based quantum computing, precision medicine, security, and so forth.

Similarly, by interfacing 2D materials to metallic nanostructures, one can also generate a novel type of plasmonic resonance, for instance, when graphene is connected to the metal surface. By exploiting the unique gate-tunable inductance of graphene, Jadidi et al. were able to produce a resonance in a metallic structure that, by itself, exhibits no resonance.⁹ Alternatively, by inserting a graphene spacer in between plasmonic nanostructures and a metallic substrate, one can create subnanometer gaps that can be used either to measure the out-of-plane conductivity in such a layered system or to tune the plasmon resonance by gating the graphene layer.^{10,11} This approach might be used also to couple single emitters to plasmonic nanostructures where the interaction between the two can be tuned at will by gating the 2D layer.

Beyond the simple interfacing between plasmonic nanostructures and 2D materials, one can also think about the possibility to couple plasmonic and excitonic resonances. Tran et al. reported on weak coupling between 2D hexagonal boron nitride (hBN) and plasmonic nanocavity arrays, thus enhancing the emission rates and reducing the fluorescence lifetimes of the quantum emitters in hBN.¹² Along this line, many groups were able to achieve a strong coupling at low temperatures between plasmon resonances and excitons in

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MoS₂.^{13,14} More recently, some groups succeeded to reach a strong coupling regime also in ambient conditions.^{15–17}

Scanning Near-Field Characterization Methods. Because of their layered nature, the possibility to suspend them on photonic cavities, and their well-known Raman spectra, 2D materials are particularly suitable for characterization methods that use highly localized light fields. Examples have combined different nanoparticle geometries and photonic crystal nanocavities with surface-enhanced Raman scattering (SERS) to study strain, doping, and the nature of defects in graphene and correlate them to devices operation.^{5,18,19} Raman enhancement up to 10³ has been shown for graphene, triggering interest in further studies using scanning near-field optical microscopy (SNOM), tip-enhanced Raman scattering (TERS), and tip-enhanced photoluminescence (TEPL) on 2D materials.^{20–23} With these techniques, the incident light is converted by the nanosized probe into a highly localized light field at the tip's apex. Depending on the sample, the near-field localization is used to stimulate the spectroscopic response of the surface or to launch radially emanating plasmons that can be back-reflected creating interference patterns.

These methods have proven themselves a spectacular source of information about local structural and optical properties, revealing propagating and reflecting plasmons in 2D materials with spatial resolution reaching ~10 nm.^{20–23} Plasmon reflection has been studied in graphene for nanometer-size gaps using scattering-type SNOM (s-SNOM), observing propagating plasmon modes that are important for potential graphene-based circuits technology.²³ More recently, s-SNOM has been used also to study hyperbolic phonon polaritons in suspended hexagonal boron nitride.²⁴ Phonon polaritons differ from plasmon polaritons for being collective oscillations of photons coupled with optical phonons (quanta of lattice vibrations). The authors probed directly wavelength differences in phonon polaritons propagating in regions with different substrate permittivity. In addition, different polariton damping is observed and correlated with the different geometries of hBN, that is, with and without a substrate.²⁴ Although these studies demonstrate the role of s-SNOM as a noninvasive method to probe propagating SPPs in 2D materials, near-field photoluminescence remains of great value to gain nanoscale information on the spectroscopic response of the sample. PL and nanospectroscopy have been used to study individual defects, such as crystal edges and twin boundaries. In WSe₂, this has been used to correlate changes in spectral intensity and shifts with different excitonic diffusion length, down to ~15 nm lateral resolution.²⁰ In addition, the bandgap of 2D materials can be changed by the applied strain, as shown for the linear decrease of the bandgap of WSe₂ with the increasing strain (in a range of 0–2%).²⁵ This indicates a possible direction for the analysis of strain using the energy of the PL peak.

Finally, the pressure modulation capability offered by the nanosized probes enables a pathway for strain control and to investigate the correlation between structural and optical properties. This resulted recently in the demonstration of local PL quenching and spectral blueshift measured in correspondence to crystal edges and nucleation sites for WSe₂.²⁰ Improving the understanding of built-in strain in 2D materials represents another important challenge for the physical characterization with potentials to unlock strain engineering for devices. Progress in this area has been pursued as a result of device fabrication or induced by the interaction with other

nanostructures. Studies included here have shown interesting crystal/plasmonic heterostructures based on MoS₂ mechanically exfoliated on Au nanostructures, as well as the analysis using TERS.²¹ The role of residual biaxial strain induced by the fabrication has been determined, whereas Au nanotriangles allowed for high Raman intensity enhancement of the MoS₂ down to ~25 nm resolution for spatially localized features. To maximize the amount of incident light that is coupled to the AFM probe, thus increasing the Raman signal purely originated by the tip plasmonic resonance, remote-excitation TERS has been demonstrated using grating-coupled excitation of SPPs.²⁶ Here, SPPs are coupled with nanostructures, such as Ag nanowires mounted on standard probes to filter out the background spectra that is generated by uncoupled light, thus improving the signal-to-noise ratio at a lateral resolution down to ~10 nm.²² In addition, regions of maximum local strain could be correlated with the areas of maximum morphological curvature thanks to the AFM.²¹

Applications for Optics and Electronics. The most widespread application of hybrid systems incorporating both plasmonic nanostructures and 2D materials is photodetection. Plasmons have been demonstrated to increase the photodetectivity of the MoS₂-monolayer photodetectors by a factor 1000.²⁷ Similar performance was achieved also by using graphene in metal–graphene–metal photodetectors, where 4-fold enhancement of the responsivity was observed, as well as an increase of 1 order of magnitude of the photoactive length.²⁸ In combination with arrays of plasmonic antennas, bilayer MoS₂ was used to obtain hot electron-based photodetection, demonstrating a photoresponsivity competing with silicon-based photodetectors without photoamplification.²⁹ In this area, Jariwala and co-workers reported on near-unity light absorption using ultrathin WSe₂ optoelectronic device (<15 nm).³⁰

Recently, the photothermoelectric (PTE) effect has been used in a novel detector for terahertz radiation, where a narrow-gap antenna simultaneously creates a p-n junction in a graphene channel located above the antenna and strongly concentrates the incoming radiation at this p-n junction for the creation of the photoresponse. The detector combines excellent sensitivity with a noise-equivalent power of 80 pW/Hz at room temperature, response time <30 ns, high dynamic range (over 3 orders of magnitude of linear power dependence), and broadband operation.³¹

In optical technology, also photovoltaic applications based on coupling 2D materials and plasmonic nanostructures have been shown. Che et al. demonstrated that silver nanoparticles attached on graphene/n-Si solar cells enabled ~11-fold plasmonic-enhancement in the power conversion efficiency.³² Nonlinear meta-lenses based on one monolayer of tungsten disulfide (WS₂) and metallic nanohole array enabled both generation and manipulation of nonlinear signals outperforming typical plasmonic metasurfaces of 2–3 orders of magnitude.³³ For reconfigurable mid-infrared beam steering, a gate-tunable graphene–gold resonator has recently enabled highly controllable reflected phase at multiple wavelengths with up to 237° of phase modulation and 23% of beam steering efficiency for reflected light for angles up to 30°. Along this line, the implementation of nanoscale nonreciprocal optical isolators is one of the main challenges of active nanophotonics. Notably, Kuzmin et al. predicted a giant Faraday rotation (up to ~100°) for SPPs propagating on graphene-coated magneto-

optically active nanowires reaching 500 nm at mid-infrared frequencies.³⁵

Finally, it is worth noticing that 2D materials can also be used as detectors of weak incoherent forces. This is the case of the WS₂-based nanoresonator operating in the strongly nonlinear regime to obtain force sensors that achieve high sensitivities at room temperature.³⁶

In conclusion, we have reported the most recent advances on the coupling between the field of plasmonics and 2D materials published in *Nano Letters* in the past decade. As well, we have discussed how we can exploit this coupling to achieve better performance in some specific areas, above all photo-detection. Future developments in hybrid systems, where excitons and plasmons are coupled on the nanoscale for emerging optical and electronic applications, are expected to gain increased importance. In addition, it is possible to exploit the intriguing nontrivial physics that arises from nonlinear optical effects.³⁷ Here, the strong interaction between plasmons and the modes supported by 2D materials can be used to extract, with subnanometer resolution, structural and dynamical features in both the layered material and in the plasmonic nanostructures. In combination, further development of in situ and in-operando analysis methods is welcome to elucidate the role of local structural, optical, and electrical properties in more complex sensors and devices based on 2D material heterosystems. Interestingly, promising results in combining metallic 1T-phase transition metal dichalcogenides and Au/Pd-MoS₂ nanosheets increase the prospects for enhancing catalytic applications using 2D materials.³⁸ These findings represent clear advances in the field, and we would like to offer also a perspective on the future of the community. It is natural to ask if the strong coupling between plasmonic and excitonic states will be used in the field of nanochemistry, for example, to affect chemical reactions and/or specific electronic dynamics on very short time scales (down to few fs). Alternatively, the ultrafast dynamics of the first femtoseconds after the excitation can be used to gain more insight on the physics underlying this strong light–matter interaction and disclose the physical mechanisms at the base of the observed effects in the steady state regime. To our knowledge, the ultrafast dynamics of all these phenomena is still unexplored representing an exciting area of research for the community in the years to come with potential to open new pathways toward an all-optical control of electronic dynamics in light harvesting and nanoenergy.

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Notes

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