Feature issue introduction: Beyond Thin Films: Photonics with Ultrathin and Atomically Thin Materials

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Abstract: Rapid advances in nanotechnology have brought about the realization of material films that may comprise only as little as a few atomic layers. For example, semiconductors and metals of sub-10 nm thicknesses have been realized. Then, the graphene paradigm inspired a plethora of other 2D and quasi-2D materials. In this new era, materials have transcended beyond the traditional thin films to a domain of atomic-scale thicknesses, unleashing vast and unexplored possibilities for light-matter interactions. This *Optical Materials Express* virtual issue features a collection of thirty-five papers aiming to capture the current state-of-the art, trends and open research directions in photonics with ultrathin and atomically thin materials.

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1. Introduction

The quest to push down the thickness of materials below the sub-10 nm scale down to atomic-scale thicknesses is driven by the appealing material properties that can be obtained at these scales; properties that are typically unattainable with their bulk counterparts. For example, ultra-thin metals can be highly tunable and conducting while transparent making them ideal for optoelectronic devices [1]. Two-dimensional materials have electronic properties easily and widely tunable by a gate voltage, strain or an external electric field [2,3] leading conversely to highly tunable optical responses. Moreover, different 2D materials exhibit non-linear photonic responses spanning across the electromagnetic spectrum (EM) from microwaves to the UV [4], thus making them a fertile playground to explore non-linear photonics at the atomic scale. Furthermore, certain 2D material in the class of MXenes, can withstand unusually high fields making them ideal as saturable absorbers [5] or as scattering constituents in random nanolasers [6]. Notably, atomically thin layers of different material can be intermixed and held together with van der Waals forces into mechanically-stable heterostructures. These van der Waals heterostructures, envisioned by Geim and Grigorieva [7] pave the way for bottom-up quantum engineering of desirable photonic properties.

The recent surge in realization of sub-10 nm metals and semiconductors and the burst of emergent 2D and quasi-2D materials have opened up many uncharted domains for photonics research and devices. The possibilities and current research directions are vast. We hope with this feature issue to highlight some of the main trends in this rapidly evolving field and give a

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flavor for the current state-of-the art and exciting future research directions. In the following, we summarize the collection of papers in this feature issue in the context of the different emergent research directions.

2. Ultra-thin plasmonic materials: fabrication and applications

Continuous sub-10 nm plasmonic-films are highly sought after for applications as they can potentially exhibit photonic responses that are highly tailorable and strongly depend on the applied electric field or the dielectric environment. These properties make them ideal candidates for tunable and dynamic nanophotonics [8]. In the recent years, major milestones have been achieved in this area; for example, epitaxial 2nm-thick films of titanium nitride (TiN), a CMOS compatible plasmonic material, have been reported in Ref. [8]. However, fabrication of sub-10 nm plasmonic films still very much remains a very active open research area with challenges to meet that depend on the particular material and targeted applications.

In this feature issue, A. Milewska et al. [9] review how to control gold (Au) atom surface mobility on common dielectric substrates to achieve continuous Au films as thin as 5 nm. Furthermore, the authors discuss the optical characterization challenges of these systems with experimental spectroscopic data suggesting a non-uniform vertical Au density profile. Then, A. S. Baburin et al. [10] comprehensively review silver (Ag) material platforms within the context of plasmonic applications. The authors review a plethora of fabrication techniques and analyze the possibilities and limitations of different methods. In particular, they report in their review that continuous sub-10 nm Ag films can be fabricated on silica with the introduction of a Ge wetting layer (see also related article of Ref. [11]). In addition, they report that Molecular Beam Epitaxy (MBE)-grown Ag films on GaAs (gallium arsenide) represent the thinnest continuous Ag films that have been hitherto realized at a thickness of 1.5 nm (see also related article of Ref. [12]). The authors also review characterization techniques that have been employed for silver platforms for their surface morphology, their constitutive photonic properties and the performance of associated plasmonic devices (e.g. surface plasmon propagation length).

On the application front, A. S. Roberts et al. [13] report continuous TiN films of ~10 nm thickness as the top part of a plasmonic Fabry-Perot resonator. TiN can withstand ultrahigh temperatures (refractory material). The authors utilized this property to experimentally demonstrate selective thermal emission with the aforementioned Fabry-Perot resonator structure. Moreover, the theoretical calculations of L. J. Krayer et al. [14] show that sub-10 nm films of certain metals may exhibit near-perfect absorption of visible light when interfaced with a refractive-index-near-zero material [15]. Also, S. G. Castillo-Lopez et al. [16], studied a 1D periodic layered medium [17] comprising ultra-thin metal layers. The authors report multiple sharp absorption/transmission bands with this system that are affected by finite-size effects in the metal's optical response (see also discussion on finite-size effects in the following section).

Absorption management is key to photovoltaic applications and different schemes involving plasmonic materials have been thus far proposed to improve photovoltaic-cell performance [18]. In this feature issue, the authors of Ref. [19] employ a 20-nm-thick silver honeycomb mesh on a \sim 10 nm indium tin oxide (ITO) layer as a top contact to a hydrogenated amorphous silicon (a-Si:H) thin-film solar cell. In particular, the authors show improvement of the photovoltaic-cell performance that comes both from the reduction of the resistance of the top contact and enhancement of the absorption enabled by the plasmonic resonances supported by the honeycomb mesh.

Finite-size effects in the optical response of ultrathin and atomically thin materials

Uwe Kreibig [20] in the 1970s had extensively studied the optical properties of tiny silver nanoparticles, containing about 400 atoms or so, concluding that the imaginary part of their

permittivity increases as a result of increased electron scattering at the metal surface which reduces the electron mean-free path. Then, as metal nanoparticles become even smaller at the order of 40 atoms or less (sub-1 nm sizes), quantum-size effects set in resulting from quantum-confinement that makes their electronic energy spectrum discrete [21]. Accordingly, these quantum-size effects modify the optical responses of these nm-sized metal particles. Ultra-thin films of thicknesses of ~1 nm or less would be subject to similar quantum-size effects [22]. In sub-10 nm size ranges the permittivity response may also be spatially dispersive/non-local, i.e. may depend on both the wavevector and frequency. Different quantum and semi-classical theoretical models have been developed to-date to capture these non-local photonic responses (e.g. see Refs. [22–25]). Recent works [25,26] discuss how these different theoretical models relate to each other and with the classical Kreibig picture in the regime where the electronic energy spectrum is a quasi-continuum (particle size ranges roughly between 1 nm and 10 nm [24]).

Beyond natural materials, non-locality may emerge also in ultrathin artificial materials with effective electromagnetic properties. In this feature issue, I. V. Bondarev develops a model to capture these non-local effects in a one-layer periodic array comprising nano-sized cylinders such as carbon nanotubes [27]. Given that nanotechnology brought nanometer thicknesses in the practical realm for many materials the aforementioned finite- and quantum-size effects are ever more so relevant where theories can be put under the experimental test. In tandem with further advancement of the theoretical framework, characterization methods for the extraction and understanding of optical responses need to be revisited and adapted for ultra-thin materials. Characterization could include obtaining depth-dependent information wherever important [e.g. see feature issue article of Ref. 9]. Also, in this feature issue, R. Secondo et al. [28], discuss the intricacies of ellipsometric characterization of the optical responses of absorbing thin films with a case study for TiN.

Furthermore, Catellani and Calzolari [29] apply first-principle atomistic simulations on nm-scale TiN/(Al,Sc)N superlattices to obtain their effective optical responses. The authors' results reveal that the material environment may alter the permittivity response of the TiN constituent; this limits the applicability of classical effective-photonic-medium theories for these systems [30]. These effects are expected to become stronger as the atomic scale is approached. Actually, understanding photonic responses in quasi-2D material systems, comprising a few atomic layers is extremely important for applications and devices. In characterizing these systems development of optical techniques that would determine their thickness is crucial. Y. Wang et al. [31] report a methodology based on non-contact optical contrast measurements that rapidly estimates the thickness of hexagonal Boron Nitride (h-BN) films on a suspended device platform.

Looking into the future, this trans-dimensional regime [32], between pure 2D and bulk, challenges our traditional knowledge and opens up new research directions on finite and quantum size effects to photonic responses. Development of analytical theories can work in synergy with ab-initio atomistic calculations and experimental findings to establish rigorous understanding on these systems, presently at nascent stages. Quasi-2D/trans-dimensional systems could be systems comprising few atomic layers of the same materials but could also comprise different materials. The latter systems are known as van der Waals heterostructures [7] discussed in the following section.

4. Vertical (van der Waals) and lateral heterostructures of 2D materials: fabrication, characterization and devices

Originally 2D-material mixing was envisioned in vertical stack arrangements, with the layers holding together with van der Waals forces [7]. However, more recently lateral arrangements of 2D material for atomically thin heterojunctions have also been proposed [33]. Whether in vertical or lateral arrangements, these emergent atomically thin platforms are expected to play a

central role in photonic and optoelectronic devices offering great flexibility in bottom-up design of their properties.

In this feature issue, H. Taghinejad et al. [34] provide a comprehensive review of the current state-of-the art in realizing lateral and vertical heterostructures of 2D transition metal dichalcogenides (TMDs) and discuss their responses from photoluminescence (PL) and Raman spectra. Moreover, in Ref. [35] P. K. Sahoo et al., employ tip-enhanced photoluminescence to characterize certain 2D-TMD lateral heterostructures. The authors obtain a highly spatially resolved structural profile of the heterostructures as well as their excitonic emission spatial landscape both providing crucial information towards designing associated atomically thin devices. On the device front, the review of C.-H. Liu et al. [36] on 2D-material nanophotonic devices discusses graphene–hBN heterostructures; these heterostructures can exhibit an electric-field tunable directional excitation of hybrid plasmon-phonon-polaritons as well as serve as an electro-optic modulator when coupled with a silicon photonic crystal.

5. 2D/quasi-2D materials for non-linear photonics

2D/quasi-2D material provide a terrain to unlock unprecedented non-linear photonic properties and effects. This is because at the atomic-scale electronic confinement is strong and the material photonic responses strongly depend on external stimuli and the adjacent material environment [4,32,37].

In this feature issue, Y. Wang et al. [38] review second-harmonic generation (SHG) with different atomically thin structures based on TMDs. In particular, the authors discuss the dependence of the SHG signal on the number of stacked atomic layers, on the relative orientation of the atomic layers in twisted arrangements and on the gate voltage. In addition, the authors discuss the realization of atomically thin materials with broken out-of-plane mirror symmetry leading to spontaneous polarization and how this property can be verified from polarization-dependent SHG spectral signatures. Furthermore, H. Mu et al. [39] report a new saturable absorber medium with a strong non-linear coefficient and modulation depth with a graphene-Mo₂C (molybdenum carbide) vertical heterostructure. Finally, G. Wang et al. [40] construct a new composite comprising MoSe₂ (molybdenum diselenide) few-layer flakes dispersed in a polyvinyl alcohol (PVA) matrix at different concentrations that exhibits broadband saturable absorption properties.

6. 2D/quasi-2D materials: tunable and dynamic photonic responses

Tunability is the motivating force pushing forward atomically-thin-material research whether for electronic or photonic applications. Harnessing tunable and dynamic photonic properties is key to reconfigurability and full 4D light control that impacts a range of photonic capabilities from sensing and photodetectors to photonic integrated circuits and analogue computing platforms.

In this feature issue, P. Gopalan et al. [41] explore how to control the transient THz conductivity and photoinduced carrier lifetimes, important for high-speed modulation, in few-layer tungsten diselenide (WSe₂). The authors report that such transient properties depend on the grain size and number of layers, controllable by the growth conditions. On the other hand, Li and Naik [42] demonstrate strong refractive index tunability, as large as 0.1, with the application of in-plane DC or AC bias in 1T-phase tantalum disulfide (1T-TaS₂). The authors report that charge density waves in this quasi-2D material, present even at room temperatures, underpin such strong electric bias-based control of the photonic properties of 1T-TaS₂ thereby, indicating the promising potential of 1T-TaS₂ for tunable photonic devices.

Furthermore, A. Islam et al. [43] utilize the polarization-sensitive photonic responses of black phosphorus (BP) thin films to control the response of a mechanical resonator. In particular, the authors report that a BP drum shows a strong polarization-dependent light absorption resulting in heating-induced surface tension thereby leading to polarization-sensitive resonant responses of

the drum. In addition, V. Ryzhii et al. [44] develop theoretical models for the photoconductivity of graphene and discuss the sensitivity of the photoconductive responses with temperature under condition of no/low doping and heavy doping.

7. 2D/quasi-2D materials for photonic quantum technologies

Tailored nanoscale single-photon sources are key to the advancement of quantum technologies such as quantum communication or computing. 2D materials have emerged as an attractive platform for integrated single-photon emitters that can be controlled with electric bias [45].

For example, local strain pockets in WSe₂ flakes caused by wrinkling effects can act as single-photon emitter centers [46]. In this feature issue, D. White et al. [47], demonstrate efficient coupling of these single-photon emitters to lithium-niobate on insulator waveguide platforms; an important result towards integrated quantum photonic circuits. Moreover, C. Chakraborty et al. [48] embed WSe₂ in a van der Waals heterostructure and report electric field control of both valley polarization and valley coherence suggesting this system can be a promising architecture for valleytronic devices. Finally, S. Guddala et al. [49] propose a tungsten disulfide (WS₂)-chiral metasurface hybrid platform and show that the metasurface enables the selective outcoupling and emission of excitons at the different valleys (K, K'). This study indicates a pathway for optically addressing the valley degree of freedom which can find application in optical quantum computing.

8. Patterned atomically thin materials

The discussions of the different 2D material and heterostructures in the preceding sections manifest the truly tremendous photonic capabilities atomically thin materials exhibit in their own right. Bringing together these versatile photonic atomic-scale material systems with photonic-mode engineering based on judicious material structuring/patterning widens the range of powerful light-control phenomena and applications. The pioneering work of S. Thongrattanasiri et al. [50] demonstrating perfect absorption in periodically patterned graphene is an example of the strong potential this class of systems entails. Patterned atomically thin materials is a growing research area rapidly evolving with the increasing realization of different 2D/quasi-2D material systems.

In this feature issue, several authors have explored the familiar structuring routes from metamaterials/metasurfaces [51,52], nanoantennae (particle- or void- plasmons) [53] or periodic-multilayers [17] to introduce new light-control possibilities at the atomic scale. In particular, N. Hu et al. [54] pattern graphene into a 2D array comprising a building block of four merged two-cut split-ring resonators [51]; the authors show two near-perfect absorption windows at THz frequencies. Then, T. Liu et al. [55] demonstrate tunable and polarization sensitive absorption with a 2D hole array in graphene around a wavelength of 10 microns; a spectral range that is highly relevant to biomolecule fingerprints [56].

Furthermore, T. Guo et al. [57] consider a graphene-dielectric multilayer composite effectively behaving as a hyperbolic-type of metamaterial [58]. This effective hyperbolic metamaterial is further patterned as a grating. The authors report that the photoconductive graphene functions as a tunable gain material that enables the proposed platform to act as a tunable THz-wave amplifier. Moreover, L. Han et al. [59] construct a trimer from three BP nano-slabs in a gapped Π arrangement. By interaction of "bright" and "dark" modes in each of the nano-slabs the authors demonstrate a macroscopic electromagnetic transparency phenomenon similar to that exhibited by plasmonic nanoantennae pairs (see for example Ref. [60]). The authors of Ref. [59] report that the BP platform has the advantage not only of tunability but also strong anisotropy because of BP's in-plane anisotropic permittivity response.

As structuring of 2D/quasi-2D material for vast light-control capabilities is a growing research direction, the understanding of the effects of sides and edges is very important to the design of practical devices. In this feature issue, S. Boroviks et al. [61], have performed a

detailed examination of such edge/sidewall scattering effects for the case of hexagonal quasimonocrystalline micron-thick gold flakes. The authors note that understanding these effects may be important in characterizing the permittivity response of the system.

9. 2D/quasi-2D materials integrated with mesophotonic platforms

We use the term "mesophotonic platforms" [62,63] here to encompass a broad class of components, resonant structures or structured media with characteristic length scales similar to or smaller than the free-space wavelength, -but still quite large in comparison to the deep-subwavelength scale. Geometric optics fails in this regime and full-wave photonic approaches need to be considered to determine the responses of systems in this class. Examples are metallic or dielectric metasurfaces [52,64,65], photonic-crystal (PC) thin films [66], membrane-PC cavities and waveguides [67,68], ring [69], disk [70] or sphere [62] dielectric resonators, ridge or slot silicon waveguides [71], dielectric multilayers [17] etc.

Harnessing the synergy between the tunable responses of 2D/quasi-2D materials and resonance and wave-front shaping provided by mesophotonic platforms creates new cross-cutting avenues for light-control applicable to photonic devices. Exploring these hybrid platforms for different functionalities and applications is a blooming direction in the field. Examples are emission enhancement [72,73], spatial modulation of the properties of a 2D material [74], integrated photonics [75] as well as biosensing [76], photodetectors [77], and photoelectrochemical water-splitting [78].

In this feature issue, C.-H. Liu et al. [36], review a range of quasi-2D materials integrated with some of the aforementioned mesophotonic systems. In particular, the authors discuss emission enhancement of different TMDs integrated with PC cavities or disk resonators. Modulator devices with graphene on waveguide systems have also been reported. Furthermore, R. Maiti et al. [79] explore the impact that integration of molybdenum disulfide (MoS₂) with silicon ring resonators or waveguides has on the optical losses of these systems. This is an important step in developing further active control of heterogeneously-integrated 2D materials on silicon photonic platforms.

Moreover, Z. Xin et al. [80], utilize graphene with a liquid crystal and a perforated metallic microarray as an adaptive microlens. The graphene serves as an electrode while simultaneously facilitates the alignment of the liquid-crystal molecular axes thereby eliminating the need of alignment layers which are invasive to the operation of the microlens. In addition, Y. Cai et al. [81], embed a graphene monolayer in the dielectric spacer of a metal-insulator-metal array thereby achieving up to 44% absorption of UV light in the graphene monolayer. G. Wang et al. [82] also consider a graphene/metal-insulator-metal hybrid with the top metal layer being arrays of split-ring resonators [51]. Graphene patches are in contact with the split-ring resonators and can modulate the local reflection phase. The authors discuss how their platform can accordingly function as a 1D grating with a dynamically tunable interspacing both in terms of size and orientation.

Dielectric multistacks, also known as Bragg reflectors/mirrors [17], are one of the oldest and most elemental structured photonic material. These Bragg reflectors can possess frequency regimes of forbidden light propagation (band gaps), but also have the ability to support modes that are confined on their interfaces [83], much like surface plasmons are confined at the surface of a metal. Their planar geometry makes them attractive for integration with 2D materials.

In this feature issue, H. Knopf et al. [84], demonstrate a new approach to integrate TMDs, in particular MoSe₂ and WSe₂, with high-Q microcavities amidst Bragg mirrors. The authors report modification of the PL spectrum of the 2D material as a result of the interaction with the high-quality-factor microcavity resonances. On the other hand, L. Jiang [85] utilize the surface states of the dielectric multilayer, to localize and enhance the electric field at its surface. The authors report low-threshold optical bi-stability as a result of such electric field enhancement when

coating the interface of such dielectric multilayer with graphene. Finally, Silva and Vasilevskiy [86] add a defect on a dielectric multilayer in the form of an additional air-gapped GaAs layer, operating in its reststrahlen band. The authors report that the surface confined mode of such surface defect can be tuned by incorporating a graphene sheet and tuning its Fermi energy.

10. Conclusions

Photonics with ultra-thin and atomically thin materials is a very active field of research with multiple interleaving directions cutting across different sub-disciplines. We summarized above the collection of thirty-five articles of this feature issue. The main directions and trends that we identified are: realization of ultra-thin sub-10 nm films for tunable plasmonics and absorption/thermal emission management, further understanding of finite-size, quantum, non-local and interfacial effects as material thicknesses approach the atomic scale, conceiving and implementing new atomically thin materials with lateral and vertical heterostructures of 2D materials, 2D/quasi-2D materials for non-linear, tunable or dynamic platforms as well as for photonic quantum technologies, patterned atomically thin materials and finally, hybrid platforms comprising 2D/quasi-2D material integrated with mesophotonic structures.

The guest editors hope that this feature issue will provide the readers with a flavor of the current state-of-the art and trends in this burgeoning field and inspire most exciting research yet to come.

Acknowledgements

The guest editors would like to thank all the authors who have contributed to this feature issue. We are also grateful to the referees that have greatly contributed with their service. Our thanks extend to the OSA staff for their incredible help that was essential in putting this feature issue together. Last but not least, we heartily thank the Editor-in-Chief Alexandra Boltasseva for her insightful suggestions and guidance in forming the coverage and scope of this feature issue.

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